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Socio-technological context and recommendations on V/HTR waste & spent fuel management (Report D 33.21)

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Summary

The paper reflects the principles of sustainability for nuclear fuel cycles and radioactive waste management, especially with regards to the back-end of HTR. Sociotechnical issues are addressed, such as risk perception, acceptance of disposal sites and the proliferation resistance of HTR fuel cycles.
Typical HTR waste streams are quantified on the basis of the former ANTARES design and will be similar for pebble bed reactor types.
Preliminary performance assessments for the geological disposal of spent HTR have shown the importance of the corrosion resistance of the SiC and PyC coatings of the fuel particles. Open R&D issues are identified, for future research programmes for the back-end of HTR.
The current national approaches on HTR waste management are highlighted in the recent IAEA Tecdoc-CD-1674 and are therefore not further discussed here.

Approval

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1 Introduction

V/HTR technology is not yet commercially introduced to the nuclear energy market, although it offers many safety and efficiency improvements to the existing and advanced nuclear power reactors. A special advantage is the capability for cogeneration of heat and power as well as of nuclear process heat for many industrial applications. The present public debate on the future use of nuclear energy in many European countries and abroad often neglects the development potentials of the nuclear technologies and does not differentiate between the characteristics of the various existing and innovative reactor designs. Sustainability is generally not associated with the use of nuclear energy, but rather often exclusively with renewable energy systems, only.

Besides safety concerns, especially after the TMI, Chernobyl and Fukushima accidents, the issue of closing the nuclear fuel cycle and the final disposal of radioactive waste are the main reasons to principally discard nuclear power as a sustainable energy option.

This paper tries to establish a general framework for assessing the sustainability of different energy supply systems in a complex socio-technological context on risk perception and to focus on the back-end characteristics of V/HTR, within the current national attempts to find political consensus on constructing final repositories for radioactive High-Level Waste (HLW).

It has to be admitted that the R&D on the disposal of HLW and Intermediate-Level Waste (ILW) stemming from LWR is by far more advanced than the related work done for V/HTR, up-to-now. Thus, analogies and knowledge need to be ‘imported’ from the ongoing research on the diverse options for geological disposal, because any specific V/HTR waste will also have to comply with the waste acceptance criteria established for the main waste streams of the current reactor fleets.

It will be shown, by the example of the French situation, that despite the remarkable progress on scientific results on the retention and transport of radionuclides in a geological clay formation, the public acceptance of a final repository remains as a real challenge. This is also due to the fact that long-term predictions over hundreds of thousands years have to be derived from measurements, which can only be based on one or some decades of experimental evidence. In this context, retrievability of HLW is being discussed as an alternative final disposal concept.

Proliferation Resistance (PR) and Physical Protection (PP) is another issue, which attracts strong political and public attention and will be discussed with regards to the V/HTR conditions. A specific chapter is devoted to the sustainability evaluation of V/HTR because the spent fuel management is e.g. also influenced by thermal efficiencies and fuel burn-up or Transuranics transmutation capabilities.

A quantitative analysis of the specific V/HTR waste streams reveals that the spent fuel contains more than 90% of irradiated-graphite, raising the question of different disposal options with reduced volume requirements and adequate waste containers.

The coated-particle fuel concept is assumed to provide a ‘perfect’ enclosure for fission and activation products, over the long-term. There are at least good reasons for strongly reduced Instant Release Fractions (IRF), for this type of spent nuclear fuel. However, it is not yet evident that this feature can positively influence the socio-technical public debate. The scientific basis for the long-term performance assessment of disposed V/HTR fuel still needs to be considerably improved for a reliable and convincing comparison with disposal characteristics of other nuclear reactor types.

Finally, the paper provides an analysis of open R&D issues as well as recommendations for further R&D on the Back-End of V/HTR.

2 Sustainability Challenges

2.1 Sustainability Concept and Challenges for Energy Systems

The guiding principle of 'Sustainable Development' is gaining increasing importance on different analysis and political levels. Considering e.g. the preparations by national governments, scientific advisory councils and other non-governmental organisations for the earth summit in Johannesburg in August 2002 and thereafter, one notices that both developing and industrialised countries deal with the concept of Sustainable Development. This is meanwhile also accepted in the nuclear field by approaches such as INPRO, GIF, Sustainable Nuclear Energy Technology Platform (SNE-TP) etc.

There exist three conceptual approaches [Kuckshinrichs 2005]:

1) Weak Sustainability – the paradigm of perfect substitutability: The concept of Weak Sustainability follows the utilitarian principle and can be called the paradigm of perfect substitutability. It is based on the assumption that the entire capital stock consists of natural capital, human capital and man-made capital, and it demands that the capital stock has to remain at least constant over time. According to the paradigm of perfect substitutability, there are no restrictions for the actual composition of the capital stock. For natural capital this means, that both non-renewable and renewable resources can be diminished since the stock of other capital goods increases, and that this is not in contradiction with sustainability. A prominent approach to put this concept into concrete terms is the (economic) Capital Approach, which aims to quantify all system aspects into one (monetary) indicator. Mainly, the lack of necessary information and data prevents to use this concept.
2) Strong Sustainability – the paradigm of complementarities: The concept of Strong Sustainability calls for keeping both the aggregate total value of man-made capital and natural capital and the total value of natural capital itself at least constant. In contrast to the first concept it restricts the substitution of capital components, because artificial capital goods cannot in any case replace the services and amenities of nature. It also implies barriers to the consumption of non-renewable and renewable resources. Additionally, it assumes that services from natural and man-made capital goods are frequently complementary. A prominent approach of this concept is the Ecological Footprint which aims to assess the impacts of human activities by quantifying all system aspects into one (ecological) indicator. Mainly, inabilities to reflect important non-ecological system aspects prevent to use this concept.
3) Normative concept – indicator set: Sustainability, as defined in the Brundtland report and the Rio Declarations, is linked with ecological, social, economic, cultural and institutional development aspects of the world's societies. Viewing these different sections of sustainable development from a normative perspective for decision making, the terms "dimensions" or "pillars" have been widely established. Among the normative concepts made available so far, the Three-Pillar Model is most prominent, accounting of aspects of the ecological, economic and social dimensions under equitable ranking conditions. The normative concept can be regarded as a mixed approach which contains elements of both Weak and Strong Sustainability. On the one hand it allows substituting capital goods, but on the other hand it enables, too, to define priorities for specific components of the indicator set.

However, in practice the Three-Pillar model as shown in Figure 2.1 turned out to be helpful for quantitative assessments within tight limits only. Nevertheless, stakeholders usually can accept the mixed approach because it allows reducing complex systems to a set of its main elements; they can communicate their individual preferences and identify their main indicators. Therefore, nevertheless, different stakeholders may support different sets of indicators.[1]

[1] As an example for energy indicator sets see Vera et al. (2005).

For energy systems, different stakeholders were involved to effectively incorporate Sustainable Development and took the challenge to define indicators and to create comprehensible indicator sets. Although considerable efforts were undertaken, there was only little success to agree upon common Sustainable Development energy indicator sets. Not surprisingly, this reflects different aggregation levels of energy systems, different framework conditions and assumptions, as well as different political viewpoints and different systems of values.

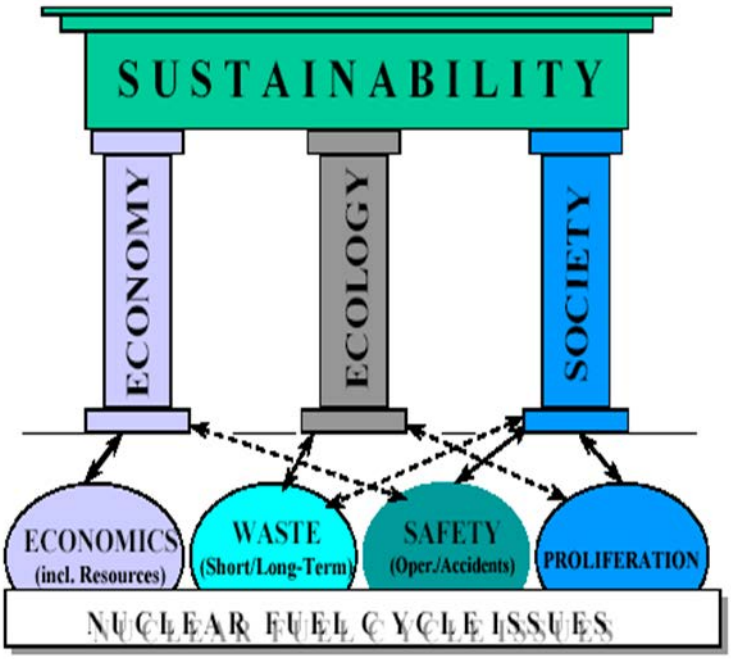

Fig. 2.1: Three Pillar model on sustainability

2.2 Concept for Energy Technologies, in particular for Nuclear Energy

Summarizing the above interpretations of sustainability concepts and its deductions the following preconditions as for sustainable energy supply systems are established. An energy supply system is sustainable, if

- the potential for provision of energy services increases or does not decrease for the next generation,
- the substances releases resulting from energy use are in line with the natural assimilation capacity,
- energy services are provided with the least possible resource input, including the "environmental resource".

Although these preconditions are widely accepted, it is the details that are most difficult. As a consequence of the first precondition, the known energy and resource base that is economically exploitable is only allowed to decrease in line with the system's ability to provide at least the same amount of energy services. This includes successful further explorations and development of advanced extraction technologies, increasing primary and final energy productivities as well as making available new energy sources. The other two preconditions show similar complexities.

For nuclear energy generation, it has to be recognized that public perception and acceptance is strongly influenced by the potential for catastrophic accidents (e.g. Chernobyl), the waste issues of radiotoxic long-lived radionuclides (minor actinides and fission products) and by the proliferation aspects of the closed or open fuel cycle. As a result, European countries actually follow different

policies on nuclear energy ranging from extended future deployment, via moratoria up to phasing-out.

Although there are specific aspects for nuclear energy generation, as shown, there is no single energy generating technology, which might be regarded to be generally sustainable, if a life cycle perspective from cradle to grave is adopted. This perspective demands not only to look at the operation of an energy generating facility, but also to keep in mind the extraction of primary energy carriers, the construction and decommissioning phase as well as final waste treatment, all of this in its ecological, economic and social aspects. Therefore, comparative energy generating technology assessment is necessary.

2.3 Sustainability Indicators for Comparative Technology Assessment

For a sustainability assessment of energy technologies solutions are to be formulated for the following critical aspects:

- What are the technical systems to be assessed?
- What indicators form a general set of Sustainable Development indicators which different actors and stakeholders can agree upon for technology assessment?
- What are adequate measurement methods for both quantitative as well as qualitative indicators?
- What are the relative weights of the indicators?
- The assessment process itself, should it be exclusively done by experts?

The generally very abstract normative goals and value concepts must be transformed into concrete "instructions for action". This requires indicators, with which a development can be characterized as comprehensively as possible. The task of indicators is to provide information about the current system state. With the aid of indicators, it should be possible to tell whether the system state observed is perceived to be satisfactory or whether changes must be made. Furthermore, indicators should be quantifiable as far as possible. Time series are required for an assessment of the dynamics of the system.

Being aware of competing electricity generating technologies and keeping in mind international research activities, it is reasonable to identify sustainability indicators which are appropriate for nuclear as well as non-nuclear technologies. The following approach is recommended here:

- Starting from a general definition of Sustainability and general sets of Sustainability indicators (UN-CSD, OECD, European Union, etc.);
- Avoiding an isolated nuclear interpretation of Sustainability;
- Evaluating different/common criteria & indicators for energy systems (e.g. German Study Commission Sustainable Energy Supply), including nuclear approaches (INPRO, Gen IV, NEA, MIT, PSI etc.);
- Identifying consensual platform (including anti-nuclear approaches);
- Being aware of the social dimensions (e.g. specific risk perception and public acceptance mechanisms);
- Identifying & defining a fuel cycle-specific indicator set (from parameters).

The previous Figure 2.1 also shows the main Sustainability issues for the nuclear fuel cycle:

- Economics
- Waste characteristics

- Safety during operation and potential accidents of fuel cycle facilities
- Proliferation resistance

It should be noted that the 'burden' on the 'Pillars' is increasing with the growing world population and enhancing standards of living.

Sustainability does not only address environmental, ecological and social compatibility but also economic and industrial aspects. Therefore, the problem of acceptability of nuclear energy and its interrelationship with technological innovation and evolution of the boundary conditions has to be discussed on several levels:

- public acceptance,
- industrial acceptance,
- internal acceptance / consensus within the nuclear community.

'Risk Perception' is being understood as a result of complex cultural and social evolutions as well as a human sense that does – obviously – not obey the risk formula (product of expected extent of damage and corresponding frequency of occurrence) being used in hazardous technologies including nuclear engineering. An attempt is being made to illustrate the mismatch of the two-parameter linear product in the definition of a technical measure of probabilistic risk and the human sentiment for the acceptance of risk in a multi-parameter interdependence of different additional aspects. A better understanding and 'acceptance' of these mechanisms might lead to an improved communication culture between the public, the industry and the nuclear community and could provide some orientation for sustainability-oriented R&D.

Although nuclear energy is a proven CO_2-free energy supply system, it has not been judged as a future option at the Global Climate Protection Conferences. Even the European Parliament rejected only by a very narrow vote (225 against 218) a statement that 'nuclear power could not be considered a safe and sustainable method of energy production and did not, therefore, belong in the policy against climate change' [Blix, 1998]. On the other hand, a study commissioned by the EC clearly states that to fulfil the commitments taken by the EU member states with regard to the reduction of greenhouse gas emissions, a massive deployment of nuclear power plants (in the order of 100 GWe) has to take place in Europe in the decades to come [EC, 1999]. This highlights the absurd discrepancy between the argumentation of proponents on the one hand and ignorance of the potential of nuclear energy and social sensitivity on the other. In this context, it is extremely important to create a 'Coalition' of sustainable CO_2-free energy systems instead of permanent confrontations and competition between nuclear, solar, wind, fossil energy resources and others.

2.4 Need for Innovation

The mechanism of the 'evolution of species' (taken here in their broadest sense) is mainly driven by changes / evolution of the boundary conditions and of those of other competitors, to which the species have to respond by viable mutation or change of attitudes. The boundary conditions for nuclear energy have evolved drastically in contrast to the expectations in the early period of its deployment. It is unrealistic to expect that the social and economic environment will adapt to the intrinsic technical evolution of nuclear technology as often claimed by the nuclear community. On the contrary, nuclear technology also has to 'mutate' quickly enough to keep pace in coping with the evolution of requirements and of competing technologies.

Nuclear energy still possesses a huge innovation potential. But this potential has not been introduced strongly enough into the discussion on the future of nuclear power and its role for the improvement of the market situation. There is a real danger that the future of nuclear power could be decided only based on a further extrapolation of the anticipated deficits of existing plants that were established some decades ago and without taking the potential for technical improvement into account. Successfully applied technical solutions, convincing operational experience, advanced commercial projects and innovative approaches complement each other to create a

long-term option on nuclear power and to provide a progressive argumentation on future prospects.

The reasons for opposition to nuclear energy are partially known from investigations / interrogations and the question arises whether technological innovations can respond to these fears, changed views and trends as discussed for some selected examples in this paper. This applies to the public as well as to political and industrial decision makers.

'Market' compatibility is a prerequisite for industrial commitment and the acceptance of 'risk' concerning the safety of investments and return of capital. 'Structural' compatibility is needed to apply nuclear energy under effective & rational licensing, manufacturing and operation procedures with no major penalities in comparison to competing systems. 'Environmental' compatibility of nuclear power is often referred to by the nuclear community in defending the need for nuclear energy but counteracted by the discussion on hypothetic catastrophic releases of radiotoxics and long-lived waste. 'Mental' compatibility of nuclear technology might even be more important for reconciling nuclear energy with public opinion again.

These goals – in a long-term view – also represent a challenge for R&D on innovative technological approaches and should be fostered by a consensus within the nuclear community despite potential different views on alternate nuclear options [SINTER Network, 1998].

2.5 Different Levels of Acceptability

The acceptability of nuclear power is mainly influenced by concerns about safety and waste and other conflicts like credibility or communication problems and different political perceptions on future energy supply systems as will be later discussed in more detail.

Another aspect is the decline of acceptability for nuclear power by industrial decision makers. The actual mechanisms of industrial decisions on investments or commitments to nuclear energy are also a result of evolutionary – possibly irreversible – changes in the attitudes of industrial leadership, management and criteria (e.g. shareholder value, short-term return of capital etc.) as well as the uncertain competitiveness of nuclear against low-priced and highly effective fossil-fired power plants. The 'safety' of investment is not only judged on a purely technical and financial basis but also taking into account e.g. the 'risk' of changes in market structures, needs for backfitting and the danger of early shut-down due to political influences or even draw-backs in other business areas as a consequence of the "nuclear engagement". Normally, the loss-of-investment risk and damages in case of nuclear accidents are also partially not accepted by insurance companies as they would be for other industrial projects.

The field of industrial acceptability of nuclear power is often assessed with a sentimental view back to the 'good old days' when industrial leaders claimed for long-term perspectives and developments as well as for large investments with return of capital in the long-term. Monopolistic structures in electricity generation are also shrinking in favour of free markets, competition and opportunities for independent energy suppliers. The actual situation has therefore to be analysed and 'accepted' as a basis for 'tuning' the technical concepts and economic features of nuclear power plants. The economic attractiveness of conventional power plants is also a result of drastic improvements in efficiency and cost reductions using the benefits of series production and low-cost manufacturing of equipment in the world market. Long and complicated licensing procedures impose additional handicaps and risks on nuclear energy.

Another – but interconnected – area of 'risk perception' consequences is the still underdeveloped consensus on common strategies within the nuclear community especially with regard to innovations. Evolutionary incremental improvements may reduce technical risks and the need for extensive and expensive demonstrations, but they may possibly not keep pace with the evolution of requirements. Addressing innovative approaches from a dualistic perspective by the attribute 'revolutionary', indicates that innovations are judged to endanger existing technologies and installations.

It is rather essential that the nuclear community defines a consensus on the coexistence of different generations of established technologies and different options on innovative technologies and concepts. Such an 'internal' consensus and a visible willingness to also respond to the evolution of the social environment (e.g. by flexible innovation strategies) may be a precondition for an effective communication towards an 'external' public and political consensus.

The drastic beneficial changes of globalization and e.g., of regional unifications (e.g. EU) also have to be accepted and actively transposed into new collaboration structures on the industrial, administrative and R&D side instead of the familiar former structures on national levels.

2.6 Mechanisms of Risk Perception

Although probabilistic risk assessments (PRA) have proven to be an adequate tool for balanced technical improvements of the safety level, argumentation based on the results of PRAs did not lead to improved public acceptance in the long run. And even much smaller probabilities for catastrophic events or quantified comparisons of risks associated with different energy supply scenarios might not really change the situation.

The reason might be that the risk defined as a linear product of damage and probability does not respond to the functional dependencies of the very essential human sense for the recognition, handling and avoidance or acceptance of danger. This human sense has permitted a survival of the human species for millions of years even under more dangerous and hostile conditions. All other human perceptions mainly behave in a non-linear way, so why should this one be linear?

Table 2.1: Parameters influencing risk perception

Kind of risk constraints	**Nature of risk source**
• voluntariness • possibility for avoidance • distribution over time • geographic distribution • degree of being affected • potential for control, active influence • social control • degree of familiarising / knowledge • scientific/technical maturity	• natural or artificial risk • possibility for fleeing • sentiment for danger • distance to the source of danger • reversibility / irreversibility • human / social risk • alternate possibilities • peaceful or military / terroristic use
Dimension of risk consequence	**Nature of benefit**
• fatal vs. limited damage • delayed vs. prompt damage • catastrophic vs. continuous damage • somatic or genetic effects	• exclusivity of benefit • public vs. private benefit • distribution of benefit • short vs. long-term benefit

It is a fact that the hemispheres of the human brain operate in different ways. Whereas the left side mainly balances rational intellectual input and experiences, the right side mainly reacts in an associative way. Both judgements influence the acceptability of an action or a decision. This fact cannot be ignored by the proponents of nuclear power and must be accepted as a God-given natural mechanism with which the use of nuclear energy must comply. Not vice versa!

Everybody can check for himself whether the perception of a danger or a benefit has the same power as the probability of its occurrence. Nobody would play a lottery if one only judged it using probabilistic methods. There is a large personal potential benefit, with a limited amount of money

as venture against a negligible probability for a significant prize. Obviously, the probability for that chance is strongly overestimated in the expectation of a huge benefit although a smaller gain is more probable. If the potential financial risk were very much higher – e.g. the income of a whole month or even a year – the participation in lotteries would be reduced to some individuals only who accept that game. Even in the case of very tempting benefits, there will always be an upper limit for the venture or perceived damage, otherwise it will not be accepted at all. Higher potential damage – even the loss of life – is accepted, if one judges oneself as being able to influence the procedure by own skills either in games or, e.g., by driving a car.

This is also willingly accepted even for a higher potential number of fatalities when, e.g., flying in a large aeroplane due to the trust in the skills of the pilot and the reliability of the plane and the airline. But panic reactions might result at the very moment when minor unusual disturbances occur. Accordingly, an 'Acceptance Formula' has to be enlarged by some other parameters describing the voluntariness, reliance on personnel / equipment and control of both compared to the 'Risk Formula'. The Institute for Decision Research in Oregon [Renn, 1984] referred to some more comprehensive qualitative risk and benefit characteristics as shown in Table 2.1.

Normally, only some of these parameters are selected for simplified or statistical reductions. However, for a complex acceptability problem like the acceptability of nuclear the full set of influences with different weights has to be kept in mind. Cultural / national influences may lead to different weights or variations but the basic mechanisms are the same, at least in a latent manner.

It is obvious that the risk formula does not at all respond to this complexity of judgements that human beings are able to perform in an intuitive way. This excellent result of human evolution also has to be respected within the argumentation on nuclear energy and to be transposed into technical and structural requirements for reconciling nuclear with the mechanisms of acceptability.

2.7 Technological Consequences of Risk Perception Mechanisms

Some of the risk perception mechanisms can indeed be addressed by technological / structural convergence or innovation as, e.g., claimed for the HTR designs.

The few examples in Table 2.2 show that the trends towards advanced and innovative designs (such as HTR) are principally on the right path for coping with these anticipated parameters and that recent licensing requirements requesting restriction of maximum releases and the inclusion of core melt accidents in the design as well as the need for a co-operative communication can be correlated logically. It is also important to note that passive safety systems might have the 'charm' to control an artificial risk by natural means.

The technological innovation potential to address these factors is often not discussed in an open way mainly because of the 'risk' perceived by the nuclear community itself with regard to potential negative consequences of upcoming, but not yet available concepts for the operation of older designs.

The risk / technology controversy is not a temporary pathologic effect but characteristic of modern societies. It is very important to handle this conflict in a positive way and to develop – as a 'social innovation' – a culture of communication and dispute. It is essential to respond by technological innovations to the mechanisms and needs of acceptability and sustainability. Measuring new technologies against the sustainability criteria as defined above may be an appropriate way to balance the advantages and disadvantages of each alternative in a consistent and fair way and to derive a transparent decision basis for synergetic solutions.

An attempt is made in the following chapter, to apply these principles to the disposal of spent fuel and radioactive waste taking the situation in France into account, where nuclear energy is deemed to be more accepted, at least from the view and perception of other countries. The reflections also apply to HTR, as the spent HTR fuel also needs to be either directly disposed or reprocessed. In the latter case, the waste stemming from reprocessing will be similar to LWR and most probably also vitrified.

Table 2.2: Examples of technical response to risk perception parameters (non-exhaustive)

Qualitative Risk / Benefit Characteristics	**Potential Technology Response**
Possibility for avoidance	Independent barriers, self-acting safety systems
Potential for control	Accident management measures
Social control	Transparent technology and surveillance
Degree of familiarisation / knowledge	Neutral competent information
Scientific / technical maturity	Open information on the technological progress
Fatal vs. limited damage	Limitation of maximum releases
Delayed vs. prompt damage	Longer grace periods
Catastrophic vs. continuous damage	Reduction of inventory, transmutation of long-lived waste into short-lived material
Extreme catastrophes	Deterministic exclusion / limitation by design
Natural or artificial risk	Protection via natural laws (passive systems)
Possibility for escaping	Slow accident progression
Sentiment for danger	Open information on all abnormal situations
Reversibility / irreversibility	Restricted max. release, accident management, Retrievability of disposed radioactive waste
Alternate possibilities	Neutral comparisons, sustainability criteria
Distance to the source of danger	Siting, underground construction
Distribution of benefit	Lower local electricity cost near to NPP

Only in case of direct disposal of spent HTR fuel, there might be a benefit from the different fuel design and the coating of the fuel particles. This will be discussed, later. However, direct disposal is generally not perceived to be compatible with the principles of sustainability. Proliferation resistance is another issue of concern and will be addressed, too.

3 Social acceptance of nuclear waste disposal

3.1 From Laboratory Data to the Safety Analysis and Societal Concerns

The following chapters intend to illustrate the conflict between R&D results, scientific evidence and societal concerns, by concentrating on the geological disposal, which is a precondition to close the fuel and waste cycles for any kind of nuclear reactors, including HTRs. After more than 30 years of research and development, there is a broad technical consensus that geologic disposal will provide for the safety of humankind, now and far into the future. Safety analyses have demonstrated that the risk, measured as the exposure to radiation, will be of little consequence. Still, there is not yet an operating geologic repository for highly radioactive waste, and there remains substantial public concern about the long-term safety of geologic disposal. In the present chapter, we assess the meaning of the technical results and derived models for long-term safety, considering model validity and credibility not only from a technical perspective, but in a much broader historical, epistemological and societal context. Safety analysis is treated in its social and temporal dimensions. This approach provides new insights into the societal dimension of scenarios and risk, and it shows that there is certainly no direct link between increased scientific understanding and a public position in favor of or against different strategies of nuclear waste disposal. This is not due

to the public being poorly informed, but rather due to cultural cognition of expertise and historical and cultural perception of hazards to territories destined for disposal. The societal and cultural dimension does not diminish the role of science: scientific results become even more important in distinguishing between the conflicting views of the risk of nuclear waste disposal.

Nuclear waste is one of the key environmental concerns regarding the use of nuclear energy. As early as 1978 it is stated that *"Unless a solution can be found for the permanent disposal of radioactive waste and spent fuel, there will not be the consensus needed to go ahead with nuclear power as an energy source. … We have to convince the public, those who know little about nuclear power but fear radiation"* [US House of Representatives, 1978]. Geological disposal is considered by most national and international bodies and large parts of the scientific community to be the best strategy to reduce the long-term risk of highly radioactive wastes arising from nuclear energy production [Blue Ribbon Commission on America's Nuclear Future, 2012; European Council, 2011; Ministry of Trade and Industry (Finland), 1994; OECD/NEA, 1995; Republique Française, 2006]. Regarding geological disposal, the OECD/NEA Radioactive Waste Management Committee issued a Collective Statement [OECD/NEA, 2008)] noting:

"*The overwhelming scientific consensus worldwide is that geological disposal is technically feasible. This is supported by extensive experimental data accumulated for different geological formations and engineered materials from surface investigations, underground research facilities and demonstration equipment and facilities; by the current state of the art in modeling techniques; by the experience in operating underground repositories for other classes of waste; and by the advances in best practice for performing safety assessments of potential disposal systems*."

Nevertheless, serious doubt remains in the public mind and are expressed by Non-Government Organisations, NGOs, [Wallace, 2010]. Industrial solutions for the storage of low-level waste [ANDRA, 2011] or transuranic radioactive wastes [WIPP, 2010] are operative in various countries, however, no geological disposal site exists for most of the radioactivity generated during nuclear energy production. This waste mostly exists as spent nuclear fuel or nuclear waste glass in countries that reprocess their spent nuclear fuel. Both forms of waste generate a considerable amount of heat during the first few thousands of years and remain radiotoxic for several millions of years.

The question remains whether sufficient societal consensus and confidence [OECD/NEA, 2013] can be established as regards the long-term isolation of radioactive waste by means of passive natural and man-made barriers that will not need later human intervention. Until now, society has never pursued such an enterprise. The debate on the fate of nuclear waste is going on for more than 35 years [Rosa, 2010], even though as early as 1982, it was commonly believed and stated by government representatives that the issues were not scientific, but rather political, and that the remaining challenge was for scientists and engineers to convince the public of the safety of a geologic repository. For example, in the US, the Nuclear Waste Policy Act (NWPA), amended in 1987 – which tied the entire U.S. high-level waste management program to the fate of the Yucca Mountain site – has not been followed to produce a timely solution for dealing with the U.S.'s most hazardous radioactive materials [Blue Ribbon Commission on America's Nuclear Future, 2012].

The evolution of a geological isolation system over many hundreds of thousands of years is not predictable in detail and various evolution scenarios have to be developed [OECD/NEA, 2001] to analyze all feature, event and process combinations which might occur in the future and which might compromise the required isolation performance of the disposal sites. Among the key questions which need to be addressed are how long will containers remain 'water tight' in the face of inevitable corrosion by groundwater, at which rates will radionuclides be released from the waste into groundwater, whether and to which degree contaminated groundwater will be 'decontaminated' by interaction with the surfaces presented by minerals contained in the engineered barrier materials and rocks, and how long will the migration of radionuclides through the rock take, and when and at which rates will they eventually enter the biosphere.

Any assessment of long-term waste facility performance requires thorough understanding of the physico-chemical mechanisms which govern either retention of radionuclides or their mobilization, of the hydraulic properties of the confinement system, of the temporal evolution of the geochemical and geological system etc., and all of this must be represented in detailed mathematical simulation

models constrained by, and coherent with, the best available knowledge. Sensitivity and uncertainty analyses must be performed to assess confidence levels. An example for the type of necessary data and their technical interpretation is given in [Paper I]**.**

The only problem, but a major one from a societal point of view, is that the large number of processes, models and parameters involved in the formal demonstration of repository safety does not offer any spontaneous indicator which would be understandable to politicians and citizens. Furthermore, due to the long times for which predictions need to be performed, the overall scenario cannot be verified entirely as it is deduced from many individual observations made in many laboratories and by field observations, all of which always simplify certain aspects. This poses a number of questions which will be addressed in the following*:*

- How can one deduce from short-term experimental data measured on centimeter to decimeter sized rock samples the long-term behavior of hundreds of meters scale systems?
- How to be sure that a scientific model or theory derived from experimental data is applicable under repository conditions?
- What to do if there are different models to explain the same data? How to combine simple models for various individual processes into an overall vision of the performance of the waste isolation system as well as for understanding the performance of individual barriers as a function of time?
- What are the appropriate indicators for safety and risk? How credible are predictions for hundreds of thousands of years?
- Is there any use in carrying out predictions for hundreds of thousands of years, if all data indicate that the risk will be negligible for the next tens of thousands of years?
- Is there a direct impact of an increase in scientific understanding on the perception of risks from nuclear waste disposal?

The challenge of this chapter is to contribute to answering some of these questions by combining the experimental approach of simulating repository properties with safety analyses and a societal approach, considering safety not only as a technical but as well a societal challenge, as addressed in the chapter 2 on Sustainability and Risk Perception.

The challenge is also that of a scientific method, which presents a concise experimental approach intended to validate models to simulate nuclear waste isolation features of a repository. The natural science approach requires that all data, model hypotheses and experimental conditions are thoroughly documented. But the link between the technical data, concepts and derived models with the temporality of environmental and health risks is not trivial. Here we show that we need to avoid the easy and self-sufficient shortcut, believing that comparison of model and data assure by themselves credibility and validity for the long term and that temporal scales of hundreds and thousands of years have any self-evident meaning outside of the scientific community. In this chapter 3, we address the validity, credibility and temporality of our models in the much larger historical, epistemological and societal context. Finally, we reflect risk and safety in its social dimension avoiding to consider it as pure technical issue, as it is normally done.

3.2 From Experimental Data and Models to Safety Evaluation

The relation of the technical data and models to safety is not trivial. In the present subchapter, the attempt is made to assess model validity, interpretation strategies and procedures used for long term prediction in direct confrontation to the technical data to assess the credibility of models for disposal safety for unprecedented long times. This confrontation covers both predictive capacity and safety assessment methods.

As is the case for all empirical observations, our data correspond to a specific experimental set up and any generalization made for deducing information as to the behavior of a geological repository requires a theoretical and modeling frame work. Data and models have inherent uncertainties. The experiments are conducted with the objective of parameterizing such models. In turn, the question

must be posed whether the experimental conditions and sample geometry are representative and whether these models and the associated parameters (diffusion law and diffusion coefficients, Darcy's law and permeability values, solubility of minerals, mass action equations of species in solution etc.) derived from small samples are also valid and applicable to the real situation, and for which the rock samples were assumed to be representative.

Model Validity

How do we know whether a model derived from short-term experimental data of cm-sized rock samples is correct and applicable to long-term assessment of the barrier function of a geological repository? Models need to remain valid and credible during scale up in space by a factor of 10^2 to 10^3 and in time by a factor of 10^5 to 10^6.

General concerns on model validity

Claims of model validity are frequent in literature but the term "validity" needs to be used with caution in this significant scale up. As early as 1979, it was stated that biosphere transport-dose models were validated sufficiently [Burkholder, 1979], but research is still ongoing some 35 years later. Some authors [Oreskes, 1994] even suggest that verification and validation of numerical models of natural systems be principally impossible, since confirmation by the demonstration of agreement between observation and prediction is inherently partial. Others suggest that we should think of model validation as an ever continuing process [Hassan, 2003] allowing to eliminate false models rather than qualifying a model as validated. The question of model validation has been addressed in the larger context of science and society and the meaning of truth [Nordstrom, 2012]. Based on correspondence theory of truth, scientific laws, theories, models are more certain (more valid?), the more their consequences correspond to reality. But how can we measure correspondence and how to compare it with a reality in the very far future? Alternatively, coherence theory of truth refers to the consistency of a theory with the large body of scientific truth, but such a theory would limit any new discovery in conflict with previous knowledge. We may better think [Heidegger, 1997] of truth in the old Greek tradition as "aletheia" or disclosure and not in terms of the roman "veritas" which always implies comparison of thought and world. Nordstrom suggests thinking in terms of the usefulness of models and he insists correctly that a model is always a simplification which by definition is never complete or exact.

Some examples

Taking the geochemical clay pore water model as an example: Can we say that it is validated since it corresponds to the data presented in the [Paper I] Indeed great progress has been made in the geochemical field as regards to predicting groundwater composition and water-rock interaction based on thermodynamic data [Glynn and Plummer, 2005]. This is not to negate that there are inherent short-comings which may affect the application to our data and the ability to predict, such as incomplete data bases, extrapolation of short-term laboratory data to the long term, missing kinetic constants of mineral reactions or the incapacity to describe future groundwater flow and associated uncertainties correctly, etc. [Ewing, 1999]. Still, while admitting the inherent partial character of model confirmation, characterized by incompleteness (not all trace elements are described) and non-uniqueness (different mineral phase assemblages and solid solutions may explain the same water composition), our geochemical pore water model adequately describes experimentally observed non-trivial evolution of solution concentrations, indicating the usefulness of the model.

Another example is the water transport model and the hydrodynamic parameters derived from it. In the 1990's there was a very passionate debate on whether groundwater transport models can be validated at all. According to some authors [Konikow, 1992] the predictive capacity of such models can be improved but even agreement of model and reality is no proof of model validity and the accuracy of prediction can only be assessed once the predicted period of time has passed. More practically, others [Marsily, 1992] have argued that, since groundwater transport models are based

on mass balance and Darcy's law, both of which have shown applicability in a huge amount of cases, if a model based on these principles reproduces the observed behavior, it can also be used for predictions. As the data show that Darcy's law is applicable under the conditions of the applied high hydrodynamic gradients, it is evident that the law can be used to predict the rate of water transport driven by realistic low hydraulic gradients (i.e. the same parameter applies for all gradients). Since the Peclet numbers would be rather similar under laboratory and field conditions (see [Paper I], it is indicated that some mixed diffusion and advection control also has to be considered for HTO traced water transport under realistic repository conditions.

With the limitation of partial validation, which is true for all empirical science, we can deduce from the correspondence between model and experimental reality that the models parametrized by the percolation test results can be applied to large-scale repository conditions, since they are describing consistently the observations based on "the large body of scientific truth" (diffusion law, Darcy's law, solubility of minerals, mass action equations of species in solution, etc.).

Credibility of Predictions

Application of the partially validated model to large time and space scales has the character of a prediction. One may prefer the term "long-term assessment", since one does not intend to predict a real future evolution and many evolutions are possible; one only tries to assess the consequences for a wide range of possible evolutions. The question is how credible predictions can be? The use of a valid model for wrong boundary conditions will certainly not allow credible predictions.

General concerns on credible predictions

According to Nordstrom, we can distinguish between phenomenological and chronological predictions [Nordstrom, 2012]. The first applies to time-independent phenomena such as thermodynamic solubility. But the prediction of the migration of radionuclides in a repository is a chronological prediction.

It is the nature of all empirical science that it does not provide absolute truth or completely validated models; yet scientific work remains largely credible. All predictions are faced by inherent uncertainty and flaws but many predictions remain credible. The mere notion of "credibility" implies that we do not know the truth or complete model validity. We need to be aware of what science can do and what it cannot. Projection into the future is an intrinsic part of human action, part of any identification of radiological risks and of any associated fear. Scientific understanding can increase the credibility of predictions, despite the empirical character of the data regarding radiological health impact, of our laboratory data on the migration behavior of radionuclides in clay cores, etc.

With respect to credibility, one needs to distinguish between strong and weak predictions: strong predictions are based on natural laws (Fick's law, Darcy's law…), on deduction, on complete induction or on interpolation between known states. Weak predictions are those based on induction, extrapolation and analogy.

Strong predictions and interpolation

Strong predictions are applicable for simple systems and for simple aspects of complex systems. The confidence in science since the Greeks, and even more evident since the renaissance has considerably benefited from successful strong predictions even though false predictions probably outweigh correct ones. An example is the prediction of the solar eclipse of 28th May 585 BC or the prediction of the return of the comet observed by Halley in 1682.

Predictions based on radioactive decay or of heat conduction in a repository are typical examples for strong predictions. There is general agreement on using the law of radioactive decay or Fourrier's Law for predicting future inventories of radioactive waste or temperature evolutions in a repository, provided that decay constants, thermal conductivities and the initial conditions (radioactivity inventory…) are known with the requested accuracy. Another example is the

migration of radionuclides in the repository rock due to chemical and hydraulic gradients. Strong predictions require not only partially validated models but also that these gradients are known and that the variability of rock properties (diffusion coefficients, porosity, and permeability) along the transport path of 70 m in the Callovo-Oxfordian clay rock allows interpolation between properties of known rock samples. The number of samples in the study being limited to four (see table 3, in [Paper I], the migration parameters will have relatively important uncertainties. Within this range of uncertainties, the rock samples studied are representative, since some extreme cases in carbonate mineral contents and hydrodynamic characteristics are considered which might be encountered along the transport path.

Extrapolations, mechanism and analogy

The situation is more complicated with extrapolations considered in the present context as a weak prediction. It is an inductive reasoning from a known situation (laboratory experiments for few days to years) to an unknown one (repository in far future…). Between the known and the unknown situation, there needs to be some similarity or analogy. Steel denounced an extrapolator's circle where for a serious extrapolation one needs strong similarity of known situation and target, while the target is actually not known [Steel, 2008]. A typical case is the kinetics of nuclear waste glass dissolution: Direct extrapolation of the observed laboratory trends of glass dissolution rates made 30 years ago [(Altenhein, 1982; Altenhein, 1983; Altenhein, 1984)] has led to predicted glass life times of hundreds of thousands of years. From the viewpoint of our current knowledge, one can certainly qualify such a prediction as weak, since experimental data collected for one year were fitted by an empirical mathematical expression of ever decreasing rates extrapolated to an unknown situation millions of year ahead. If we had used the type of data as from the G experiment in [Paper I] for such an extrapolation, it would as well be a weak prediction even though repository rock was present.

A sound scientific base for extrapolations may be given by identifying the physical chemical mechanisms and the environmental factors capable to interfere with these mechanisms, according to which events are causally related when there is a mechanism that connects them [Glennan, 1996]. For the problem of extrapolations from laboratory to repository, the idea is that the same physico-chemical causes shall produce the same effects even in one million years. However, there always remains a difference between the laboratory and the target situation. Guala suggested as criteria the existence of some empirical knowledge of model situation and target [Guala, 2005], but such empirical knowledge is impossible to gather for situations in the far future. Some fundamental problems of the mechanistic approach were recently addressed [Howick, 2013], in particular the fact that knowledge on mechanisms is always incomplete.

Comparative Process Tracing

To assess whether a mechanism can be used as a base for extrapolation, Steel proposed a procedure which he called "comparative processes tracing" [(Steel, 2008)]. Applied to nuclear waste glasses, one would first have to study all important mechanisms which are operatives in the model system (mm to cm sized simulated nuclear waste glass exposed to various simulated environments in the laboratory). The reliability of comparative process tracing then depends on correctly identifying the points, at which significant differences between the laboratory studies and the canistered nuclear waste glass in a future repository are likely to occur. Significant differences are those that would make a difference to whether the causal generalization to be extrapolated remains valid in all of the evolutions which might occur to the glass and its environment during disposal.

In this sense, large experimental research programs have been conducted worldwide since more than 30 years studying effects of glass composition, surface area, solution volume and flow rate, temperature and pressure and solution composition, presence of repository materials like iron, cement, rock samples, radioactivity, etc. In general, the results have shown very low long-term glass dissolution rates in water in a confined space like that expected in a repository near field. General mechanisms were developed from model systems to explain the impact of all these

variables on the model system by fundamental physico-chemical processes and parameters like diffusion, solubility, transition state rate laws, activation energies, etc., hence, allowing strong predictions. There was some debate whether affinity terms or protective surface layers were responsible for the observed decreasing reaction rates, but today agreement is obtained that both processes are operative [Frugier, 2006; Grambow, 2001; Iseghem, 2007]. These studies allow today to address differences between laboratory model and repository target: geometric constraints like size and fracturing of the industrially produced highly radioactive glass block, the hydrodynamic conditions, the radiation fields, and the contact of iron from canister. In this sense, the G experiment of [Paper I] was conducted to validate the applicability of current mechanistic and quantitative understanding of glass corrosion to the clay–water system.

Study of Natural Analogue Systems

The largest difference between model and target is the exposure history to an aqueous environment. This cannot be studied in the laboratory but rather by studying so-called “natural analogue” glasses. Concerning analogy, it certainly plays an essential role in conceptualization of experimental data, in developing evolution and exposure scenarios for repository performance in a qualitative way. Many natural analogue studies were performed [Alexander, 1990; Brandberg, 1993] for example using volcanic basalt glasses [Crovisier, 2003; Crovisier, 1987; Ewing, 1979; Mazer, 1992] as an analogue to study the long-term performance of nuclear glasses in natural environment and it certainly increases credibility of arguments in favor of repository safety. For example, it can be shown that

(1) natural basalt glasses behave under similar conditions similar to nuclear waste glasses;

(2) natural basalt glasses can survive under natural water conditions for millions of years [Malow, 1984]; and

(3) that low rates observed under laboratory conditions in confined silica saturated environments are also observed with natural glasses [Grambow, 1986].

While not allowing strong predictions, natural analogue studies are a strong qualitative support to the safety case [IAEA, 2012].

3.3 Bounding Case Predictions

Detailed prediction of the release of radionuclides from a nuclear waste repository cannot be considered as a strong prediction. It is impossible to predict the evolution of a geological system over geological times, due to variations in the initial and, above all, boundary conditions, since the system is open for exchange of mass and energy. Also, there is the possibility of rapid non-foreseeable events etc. In the case of a nuclear waste repository, it is neither possible nor necessary to do an exact and quantitative prediction of the transfer of radionuclides from the disposal location to the biosphere in space and time. Also the ICRP states [Weiss, 2013] that the scientific basis becomes very uncertain for assessments of health impacts over very long times into the future indicating that the strict application of numerical criteria may be inappropriate. It is sufficient to demonstrate that dose and risk thresholds are not exceeded at any given time for whatever evolution of the disposal system and its environment. The prediction of the ‘non-exceeding of a bounding dose value’ is not a prediction of a future evolution, since many future evolutions may be compatible with such a bounding value. An absurd example may illustrate this type of bounding case prediction: It is certainly impossible to predict the exact temperature for tomorrow, but most scientists (and laymen) will agree that one can predict that the average temperature in Europe over the next 10,000 years will not exceed 50°C. If such a type of information is sufficient for a given purpose, a credible prediction can be made, despite the open and empirical character of the system. It is even a strong prediction, since many scientific facts are incorporated in assessing this bounding case. Certainly, such a prediction is not free of potential error. A large asteroid may strike the earth invalidating the 50°C bounding value. Applied to our case of a nuclear waste repository, one can try to do strong prediction of a bounding dose value

assuring that the evolution under a range of normal (no asteroid) circumstances will never exceed this value even when taking into consideration all plausible variability.

Before applying this concept in the use of the type of experimental data of paper 1 we must recall some very useful recommendations from the paper of Nordstrom [Nordstrom, 2012]:

(1) Model computations must be explicable to non-modelers. For example explaining geochemical model computations to non-modelers and especially to non-technical audiences is a test of how well the modeler understands the computations.

(2) The main conclusions or arguments of a complex computation should be reproducible in a simpler manner by 'back-of-the-envelope' calculations.

We add: these recommendations are even more important when deriving safety implications from experimental data, since safety concerns many people.

How to Use Data for Assessing Radionuclide Mobility in a Repository in Clay Rock?

Identification of Subsystems

We may divide the overall system of nuclear waste disposal in clay rock into subsystems which represent certain safety functions, for which one can develop strong bounding case predictions based on physical and chemical laws. Such subsystems may be described in a sufficiently simple manner allowing 'back-of-the-envelope' calculations as suggested in [Nordstrom, 2012]. Subsystems can represent parts of the real system or they can represent bounding cases (worst case conditions…) for radionuclide behavior. We may couple mathematically or logically the subsystems representing bounding cases to obtain an overall bounding case picture for repository performance.

Subsystem "Solubility in Groundwater Close to Waste"

For example: a first subsystem may be the groundwater or the pore water in the vicinity of the waste and its contamination by radionuclides. The degree of potential contamination will depend on detailed scenarios of waste / water interactions, the distance of the water from the waste, the time after disposal, etc. Maximum degrees of contamination can be assessed by predicting the solubility limits governing maximal radionuclide concentrations in groundwater, since radionuclide concentrations in water only can become higher than this limit in presence of colloids. Solubility constraints are independent of time, except if the geochemical conditions change. Hence, taking the terminology of Nordstrom, prediction of solubility is a phenomenological prediction. Thermodynamically well-founded solubility predictions for given chemical boundary conditions are strong predictions if the temperature, pressure, the nature of the solubility controlling solid phases and their composition and the solution concentrations of all complexing substances and corresponding stability constants are known, including those for organic species. Knowing solubility constraints and the water transport rate, one can then determine a bounding constraint for a maximum flux of radionuclides at a given position in the isolation system. Because of uncertainties in solution concentrations and stoichiometry of complexing substances and in the nature and the composition of solid phases, only bounding values can be predicted in a strong manner. For deriving a bounding solubility value, one needs a bounding model for the nature of solution complexes including representative organic complexes as well as the solubility controlling solid. We may take as an example the solubility of radionuclides in the pore water in clay rock. Highest radionuclide concentrations are expected close to the nuclear glass, but it is not the glass which is the solubility controlling solid phase. Instead, solubility may be controlled by one or more secondary solid alteration phases which often form on the glass surface in an ill-defined amorphous state. A theoretical assessment of empirical solubility constraints of actinides at the glass/water interface is given in [Rai, 2011] for pH and redox conditions, relevant also for clay rock porewater compositions.

At a distance of a few meters away from the waste, radionuclide concentrations are typically very low and in this case, neither solubility limits are reached nor solid solutions are formed. For the

example of tetravalent actinides, uncertainties due to unknown solubility controlling phases may lead to uncertainties in maximum degrees of water contamination of many orders of magnitude. Normally, in a series of potentially solubility controlling phases, the phases with lowest solubility should control solution concentrations. But this is only true if the solubility controlling phase forms rapidly. For example, oxides like PuO_2 have much lower solubility (are more stable) than amorphous hydrated oxides like PuO_2(am), but precipitation of the crystalline oxide is very slow. Hydrous amorphous phases can then be used for bounding solubility constraints. In the pH range of clay pore water and its potential modification by glass or steel corrosion (pH 7-9) the solubility of $Pu(OH)_4$ is about 10^{-9} M. In the literature, examples are given where solution concentrations of radionuclides are higher than solubility values due to colloid formation. This is not possible in clay rock: colloids may form but they cannot be transported since the pore space acts as a very effective filter. Hence predictions of thermodynamic solubility of fast forming phases like amorphous hydrous oxides, hydroxides or carbonates in clay rock pore water can be considered as strong predictions of bounding maximum solution concentrations of contaminants.

The Subsystem "Sorption of Radionuclides on Clay Rock" and the Problem of Non-Uniqueness of Models

Another subsystem concerns the sorption of radionuclides on clay rock surfaces. Once released from the emplaced waste, the most toxic radionuclides, the actinides, contaminate the contacting clay pore water, the maximum concentration possible being given by the solubility constraints discussed above. The dissolved radionuclide species will diffuse away from the waste and enter into contact with hitherto non-contaminated clay rock surfaces. A smaller or larger fraction of these radionuclides will then interact with the clay rock surface of the pore space and become strongly attached to these surfaces so they become retarded in their migration to the biosphere, in most cases to an extent that radioactive decay occurs well before the radionuclide in question reaches the biosphere. Large sorption databases have been created involving various radionuclides in contact with a clay powder or clay blocks in the presence of clay pore water.

Sorption models are non-unique. Different models like the K_d model or various models for surface complexation and ion exchange are frequently used. A key hypothesis is that there is a thermodynamic equilibrium between the concentrations of radionuclides sorbed on the solid and the concentrations of the same radionuclide in the contacting aqueous solution. This idea of equilibrium implies reversible exchange, a hypothesis which is not always tested and which is not always true since there may be irreversible entrapment of radionuclides on the solid, for example of Cs on frayed edge sites of illite [Poinssot, 1999]. If there is irreversible fixation of radionuclides on the solid, there will be a smaller quantity of radionuclides available for migration to the biosphere. Hence, the non-consideration of irreversible entrapment allows for a bounding case calculation, providing as a result a quantity of migrating radionuclides quite higher than the real case. Nordstrom stated that predictions of complex events are ambiguous because of model non-uniqueness. However, this statement is only correct if wrong models are used. Since various sorption models are correct (describe correctly the observations for a given frame of conditions), model non-uniqueness does not necessarily diminish the credibility in terms of a strong prediction. As long as a given model describes sorption in agreement with the experimental data quantitatively and independent of time one may use it for strong predictions of bounding effects. Sorption models may vary largely in their range of operational validity. A phenomenological K_d model may only be able to describe sorption in nuclear waste if the conditions of the disposal system are identical to the set of experimental conditions used to determine this K_d, while a model based on mechanistic description of surface complexation and ion exchange may be able to describe the variation of sorption as a consequence of the variation of initial conditions. A K_d model may become incorrect if mineral or water composition changes, while a surface complexation model may be able to capture such differences. In some cases, molecular modeling and spectroscopic characterization helps understanding formation of surface complexes. A geological host formation like a clay formation of hundreds of meters varies little with time but shows some spatial variability. Hence, it is sufficient to measure the K_d in a sufficiently large quantity of statistically representative samples. This approach may not be feasible and a full surface complexation approach may be necessary in the case of

close vicinity of the waste, since chemistry of pore waters may vary largely due to release of waste package materials (organics…).

Subsystem "Water and Radionuclide Movement in Clay Rock"

One can predict for clay rock and for natural hydraulic gradients of Callovo-Oxfordian clay that diffusion dominates over advective water flow. Taking the French clay rock of the proposed disposal site as an example, the residence time of water in the formation above the Callovo-Oxfordian clay layer are reported to be between 300,000 and 1.5 million years [Fourré, 2011]. To know the maximum amount of radionuclides which can be transported by water in clay rock, one needs to know the radionuclide concentration at the source (potentially solubility controlled), the distribution ratio (K_d) of the radionuclide between the clay rock and the pore water, the diffusion coefficient, and the accessible porosity. This means that we need to couple the subsystem "solubility" with the subsystem "sorption" and both with subsystem "diffusion". The minimum time necessary (the "breakthrough time") for radionuclides to migrate the 70 m from the waste via the clay rock to overlaying geological formations, were calculated in [Paper I] for different radionuclides: 250,000 years for the fastest moving nuclides (iodine-129) up to complete retention prior to radioactive decay for uranium, the transuranic elements, and for cesium-135. Even large uncertainties in retention parameters do not change these findings. For example for iodine-129 transport through clay, the use of the experimentally observed K_d values would allow to calculate breakthrough times of about 500,000 years. Due to uncertainties on this K_d value, one can define a bounding case value K_d of zero leading to the above mentioned 250,000 years for breakthrough.

3.4 Safety Assessment

What do our model estimations of radionuclide behavior in subsystems of the disposal system tell us about the safety of a future repository in clay rock? Whether, when and in which quantity (is it safety relevant?) radionuclides from the waste may migrate towards the biosphere? Does the reasoning of our data and models in the context of subsystems of the disposal system provide already a procedure to evaluate the effectiveness of nuclear waste isolation systems in preventing adverse radiological effects to present and future human beings and their environments?

The knowledge collected to allow long-term predictions respond to a question of the type "what if?". Answers are always given for a given set of boundary conditions and scenarios. Safety relevant phenomena and boundary conditions of natural, human and waste/repository origin must be studied all together in safety assessments. Safety assessment, the quantification of radiation dose and risks that may arise from the disposal facility, is an integral part of a general safety case which has to be made to demonstrate safety of any geological disposal concept for radioactive waste: "*The safety case is the collection of scientific, technical, administrative and managerial arguments and evidence in support of the safety of a disposal facility, covering the suitability of the site and the design, construction and operation of the facility, the assessment of radiation risks and assurance of the adequacy and quality of all of the safety related work associated with the disposal facility*" [IAEA, 2012].

Safety assessment approaches

The risk assessment approach combines classical risk analyses (consequences of events leading to release of radionuclides multiplied by the probability of the event) with systems analyses (description of processes leading to dose to humans). Probabilistic systems analyses (PSA) is used in some cases to represent combined natural and engineered barrier systems, whose parameters are not entirely characterized [Ewing, 1999]. A PSA includes 6 essential steps:

(1) identify performance measures,

(2) characterize the way how the system affects performance,

(3) build mathematical models describing performance,

(4) quantify uncertainties of model parameters by probability distribution functions,

(5) propagate uncertainty through the system, and

(6) compare performance with real system.

Probabilities can be assigned explicitly, e.g., by probability density functions or implicitly through the selection of likely and less likely scenarios. However if probabilities of expected behavior are not known, probabilistic system analysis becomes meaningless [Konikow, 1999]. In this case, safety assessment can also be done in a deterministic manner based on expected (best estimate) evolution of the repository system or on worst case scenarios.

Assessment may be divided in analyses of scenarios (how are containment barriers breached?) and of consequences (which doses?). Scenario analyses determine systematically [OECD/NEA, 2001], by which chain of features, events and processes (FEP) is it possible for radionuclides to migrate into the environment. The analyses of safety functions over time concern any barrier against water circulation including the geological environment and the disposal architecture, any barrier that immobilizes radionuclides in packages and containers in the disposal cells, and any barrier that delays and reduces the migration of radionuclides that were released outside the boundary of the disposal cell.

One needs to consider the groundwater access to the waste (*when?, how much?, vapor or liquid?*), the evolution of the container (*when will it breach by corrosion?, are there containers with initial fabrication faults?*) and the waste matrix (*thermal effects, which rate of corrosion of the matrix in water?*), the evolution of the engineered barriers in the near field (*are all voids filled to avoid accumulation of large gas or water volumes?, is there retention of radionuclides on engineered barrier materials?, which are the physical and chemical interactions at materials interfaces?*), the transport of solutes (*diffusion or advection control?, which impact of preexisting fractures and fractures created by repository construction?, which variation of hydrodynamic properties in space?*), the geology (*which frequency of tectonic events?, which geomechanical properties?*), the behavior of radionuclides (*which radionuclides are released?, which radionuclides are retained by solubility constraints and sorption?)* and the transfer to the biosphere considering the impact of natural phenomena, human activities, and the effect of the waste on the repository.

Performance Indicators: Radiotoxicity, Risk, Dose

There are various performance indicators to assess the capacity of a geological disposal concept to protect future generations against radiological risks. There is a vast literature on comparing the radiotoxicity of the radionuclide inventories of the waste with the toxicity of the ore body used to generate the corresponding nuclear fuel. With reprocessing, it takes some 20,000 years, without some millions of years until toxicity decreases to that of the ore body [U.S. Congress, 1985]. But toxicity inventories are not a measure of risk, since they are not related to exposure scenarios. For a given exposure scenario, it is common to distinguish between predicted risk and predicted dose (in dose predictions, there is no reference to the probability to be exposed to a given dose). Dose thresholds are given in $mSv \cdot yr^{-1}$ for potential exposure by ingestion or external irradiation. Recently, for the exposure from a geological disposal site, a dose limit of 0.3 $mSv \cdot yr^{-1}$ and a risk value of $10^{-5} \cdot yr^{-1}$ were recommended as “planned exposure” [Weiss, 2013].

Radiological dose thresholds are in some sense also risk thresholds as there is a conversion factor of $5.5 \cdot 10^{-5}$ $cancer \cdot Sv^{-1}$ [ICRP, 2007] between dose and risk of cancer using the typical linear-no-threshold LNT hypothesis. Only if the probability of a given exposure scenario is also taken into account, one can talk about a risk assessment.

A Short Non-Exhaustive History of Assessments

It was stated as early as 1979 that for “reasonable” isolation systems expected doses are lower than natural background levels [Burkholder, 1979]. In the European Community, performance

assessment started in the early 1980's with examples being the projects PAGIS (1982-1989), PACOMA (1989-1991), EVEREST (1992-1996) and SPA (1996-1999) using a common methodology applied to repositories in clay, granite and salt considering data and process uncertainties by sensitivity analyses and uncertainties in the future evolution of the geological isolation system by scenario analyses [Storck, 2000]. In the PAGIS project for radionuclide inventories for 30 years of nuclear power generation, calculated dose rates were in all cases lower than national regulatory limits. Resulting doses calculated in the PACOMA project on alpha bearing waste were not much lower, even though radionuclide inventories were orders of magnitude lower, indicating that there is no direct link between radionuclide inventory and expected dose. The SPA project has shown that differences between calculations by different organizations for a given repository type such as granite can easily amount to more than one order of magnitude, corresponding to different hypotheses taken regarding container failure rates and groundwater travel times. A much higher actinide inventory in a deep repository does not lead to higher dose rates, since flux to the biosphere is controlled by solubility constraints at the waste surface independent of actinide inventory. Hypotheses for transfer to biosphere are important: water taken from wells gives higher doses than from rivers due to dilution effects. The food chain is also important: eating fish gives higher doses than drinking the water. Comparing various international approaches, the EVEREST project has shown large effects of conceptual and model uncertainties on final results [Orres, 1997].

The recent European Commission Project PAMINA brought together 27 organizations from ten European countries [Bailey, 2011] and studied the impact of uncertainties on the safety case [Galson, 2012]. Uncertainties are classified depending on whether they are model, data or scenario related, all of which can be of random or epistemic (knowledge based) nature. Some uncertainties are not important to safety, since safety is controlled by other processes or the probability of an event is extremely low. Uncertainties can be reduced in some cases by adding additional barriers. Other uncertainties need to be included explicitly in the analyses or one needs to develop a bounding case and demonstrate that safety remains acceptable.

Safety assessment is now a routine exercise used at various stages in the development of repository projects to compare alternatives: designs, layouts, materials, geological environments and repository sites. It offers a means of obtaining an overall view of the robustness of a given disposal concept. The principal role of safety analyses is not so much to obtain the numerical results (dose, risk) but the identification of key factors influencing safety.

Role of Results in Safety Analyses

If we conclude as above from our type of data and models that it takes more than 250,000 years for transport of the most mobile radionuclides across the Callovo-Oxfordian clay rock barrier, does this mean, if the waste is placed in this geological formation, that we can already demonstrate that the biosphere is protected from the nuclear waste for at least this time period? Certainly, these results are an indicator for safety or a piece of evidence. But we can easily imagine scenarios with faster radionuclide transport. Mobile anionic radionuclides may migrate through plugs and seals or through damaged clay rock if these are not tight. Large gas pressures generated for example by anoxic iron container corrosion or by radiolysis may create new pathways for groundwater. These alternative evolution scenarios are typically studied one by one, whether it is realistic that they occur, etc. This leads to studies on the mechanisms of how fissures form and how they heal, for example during excavation. Large research projects [Johnson, 2013; Shaw, 2013] deal with these questions, but it is outside of the scope of the present communication. Also external forces might compromise the isolation system such as earth quakes (one can select geological formations with little probability) and mineral exploitation might occur or geothermal resources might be looked for in the future (one has to look for sites with little of such interest). We will see further below that even social scenarios can have an important impact.

Another case: our results show that glass is a strong barrier against radionuclide release also in compact clay rock environments. Times for complete alteration of more than 200,000 years should then also be valid under such conditions if the radioactive glass is immersed in clay pore water. But the question of the scenario of the temporality of water ingress to the waste is also important. We

know that glass dissolution rates in fast flowing water are much higher, but the scenario of fast waterflow is incompatible with the compact low permeability (see Table 2 in [Paper I]) of the clay rock. To recall from paper I: we needed pressures of tens to hundreds bars for pushing some milliliters of water through the rock. Also one needs to develop a scenario on how water will come in contact with the nuclear glass. For the first thousands of years, there will be no water access to the glass and hence, no glass corrosion and no radionuclide release since the container is expected to be tight. It may fail only by corrosion combined with geomechanical pressure and this process may take thousands of years. Indeed slow iron corrosion rates even in the presence of microorganisms under repository conditions are confirmed, among many data from other research groups, by the S experiment. But even after container failure, liquid water may still have difficulties to enter into the container, since due to anoxic corrosion of steel containers, overpressures of hydrogen gaz are produced, potentially blocking the inflow of liquid water to the glass inside of the corroding container. Under such conditions, glass corrosion occurs under vapor phase conditions. The present data are not directly applicable to this scenario, but complementary research is underway to study the consequences of this scenario and first results show that corrosion rates in vapor are at least as fast (if not faster) as those in liquid water [Neeway, 2012].

3.5 Safety and Society

One of the astonishing results of more than 30 years of safety analyses is that despite important scientific progress and work on stakeholder confidence [OECD/NEA, 2013], the public confidence in the results has not significantly increased. If a repository concept is convincing like in Sweden or Finland, where the safety relies strongly on the very robust copper container, there is no major problem either with safety analyses.

Safety assessment is today generally considered by research organizations, by agencies for nuclear waste management, and by technical support organizations for regulators as a pure technical issue. Social sciences are rarely, if ever, involved. This separation ignores that safety and risk are societal concepts, and it leads to shortcomings, some of which are discussed in the following.

The Social Dimension of Repository Evolution Scenarios

Impact of man on repository safety is often studied under the title “human intrusion” compromising the isolation system by voluntary or involuntary actions like the search for mineral or geothermal resources. There is a debate as to conserving the memory of the site. Not studied at all is the inverse case: what happens, if man does not act at all and abandons the disposal site before closing it permanently?

Let us take another look at the scenario of migration of radionuclides from the waste through the clay rock to the biosphere. The very long isolation times of more than 250,000 years for the fastest moving anions remains only valid if solute diffusion through the clay rock is the principal transport mechanism. This requires that plugs and seals be tight and that galleries be backfilled all the way to the entry shaft from the surface down to repository level in a manner to leave only very small void spaces. The just described scenario requires human action: after waste emplacement, in case of provisions for reversibility after some hundreds of years, the future decision makers will have to decide to mobilize large financial, human and material resources to close the site correctly. However, if the society has already lived for more than hundred years with an open and reversible disposal site, what will motivate decision makers to invest billions of euros? They may want to use the financial resources for the potential more urgent needs to win elections etc.? This leads us to suggest another scenario which merits a careful technical analysis: the termination of backfilling and closing of the disposal site. One may only speculate about the consequence since a detailed assessment is missing: it may be that galleries will become very difficult to access with time, fissures may become larger and water from overlaying geological formations may eventually infiltrate. The migration distance of about 70 m for radionuclides from the confined location of waste emplaced in the center of the Callovo-Oxfordian clay rock formation to the overlaying

geological strata with faster ground water circulation may then not be the preferential migration path from the waste to the environment, but the about 10 m from the waste through plugs and seals towards the galleries. Still many tens of thousands of years of isolation are probably to be expected for the fastest moving anionic radionuclides as well as complete isolation of actinides until their decay, but the very large isolation times of >250,000 years may then not be realistic.

Safety between Scientific and Societal Temporalities

Predictions for hundreds of thousands of years are unprecedented in human technological history. Can one reduce the time, for which a quantitative evaluation is necessary? The US-EPA requested predictions which shall be quantitative for the first 10,000 years and qualitative thereafter [US EPA, 1985]. The US Board of Radioactive Waste Management stated [National Research Council, 1990] that predicting "accurately the response of a complex mass of rock and groundwater as it reacts over thousands of years to the insertion of highly radioactive materials is not possible". In [Ramspott, 1993], it is stated that there is no technical basis for setting a time limit of 10,000 years on the regulated performance of a nuclear waste repository. Total system performance assessment (TSPA) of the Yucca Mountain repository project gives maximum doses to public well beyond 10,000 years. Already in 1999, a review panel [Peer Review Panel, 1999] stated that it is unlikely that the TSPA viability assessment, taken as a whole, describes the long-term probable behavior of the proposed repository. Also the international joint evaluation [NEA-IAEA, 2001] stated that, while compliance to performance criteria in the 10,000-year period was attained, work should continue beyond this regulatory period. In 2004, the D.C. Circuit Court of Appeals ruled that the 10,000-year compliance period was not “based upon and consistent with” the recommendation of the National Academy of Science that compliance be assessed at the time of maximum risk, within the period of geologic stability of the site. A few years later, Yucca Mountain was abandoned as repository project.

But still we are confronted with the question whether or not risk prediction has any meaning for hundreds of thousands of years. The physical concept of time, more or less in the sense of Newton as a time, in which things happen, serves as a universal temporal framework to quantify and to communicate the risk associated with the future release of radionuclides to future man. The physicists, chemists and geologists working on geological disposal projects are convinced that decisions on disposal should be made on the basis of this concept of time. In the tradition of safety analyses, no distinction is made between the significance of exposure risks which occur early or late in the disposal time. For example, even if safety analyses show that after repository closure, there is essentially zero risk for the first ten-thousand years, waste management and technical support (TSO) organizations tend to focus on the maximum dose which may occur after hundreds of thousands of years. In contrast to our everyday live, the different times for a risk to occur seem to have no impact in the evaluation analyses. The reason is probably that the time in question is so long that its meaning for today's decisions remains unclear. The citizen is not interested in predictions for millions of years. Risks are perceived by our citizens typically related to a time scale for a few months to years. The mere fact that one is obliged to assess performance for such a long time increases the perceived risk.

Of course, the concept of "risk" implies always a projection into future and it cannot be separated from the intrinsic vulnerability of man, beyond our own existence. The temporality of risk refers on the one hand to physical time (which may extend to millions of years) and societal time. The first temporality provides the frame for our capacity to predict and the second governs our concerns. Decisions may take both concepts of time into account since these are not concepts from which we can choose, they are simultaneously present, despite being conflicting. Although our ability to forecast is limited, our responsibility is not. To the degree to which we are able to predict risk we have the duty to provide protection for future generations. Radioactive waste disposal risk assessment pushes the conflict between physical and human temporality to an extreme level, deprived of any unifying horizon.

Societal Dimension of Radiological Risk

The various reference values used to compare and to quantify risk are related also to social choice and they are not pure scientific parameters, as was already discussed in chapter 2. Risk is not a pure scientific fact but scientific facts are used in the social definition of risk. Dose may be related to life expectancy. But effective dose loses its direct connection to health detriment for doses in the future after a time span of a few generations, given the evolution of society and human habits [ICRP, 2007]. Also, the understanding of the relation between dose and health risk may vary in the future.

The debate on risks associated with nuclear waste is a typical example of the fact that risks are social constructions with conflicting views on the definition of risk [Beck, 2007]. Even though safety analyses may show that there is only a small risk, opposition to disposal remains strong. Often this problem is presented as a problem of communication, or of a difference between risk and risk perception. It is essentially the psychometric theory of risk perception (see previous chapter) which stresses the subjective aspects and which maintains the sharp cleavage between the rational expert and the 'irrational' citizen [Jasanoff, 1998]: Various important properties for the subjective judgment of the risks of technologies are identified [Fischhoff, 1978]: the question whether risks are voluntarily taken, whether the effect is immediate, whether there is sufficient knowledge of the person which is exposed to the risk, whether there is scientific knowledge regarding the risk, whether one has control over the risk, whether the risk is new, chronic or catastrophic and how severe are the consequences. Indeed, the question of control is central to the perception of risk related to geological disposal. Despite the fact that geological disposal provides an additional barrier (the geological barrier) against radiological exposure of populations, when compared to current interim storage of the waste at the earth's surface, the perceived risk of geological disposal may still be higher since disposal may be perceived as an abandoning of the waste with no further possibility of controlling it. In contrast, it is indicated in [Sjöberg, 2004] that risk perception of populations is not irrational and is essentially driven by ideological concerns and attitudes, such as attitude or beliefs that a technology is interfering with Nature. Cultural theory interprets the conflicting attitudes towards risk as conflict of different world views on social organization [Douglas, 1982; Poortinga, 2002; Schwarz, 1990].

The "cultural cognition of risk" refers to the tendency of individuals to form risk perceptions that are congenial to their values [Kahan, 2011]. Two opposite expert views on nuclear waste disposal were presented to a test group: high risk: "Using deep geologic isolation …would put human health and the environment at risk", and low risk: "Radioactive wastes from nuclear power plants can be disposed of without danger to the public or the environment". Individuals systematically overestimate the degree of scientific support for positions they are culturally predisposed to accept: People with hierarchical and individualistic attitudes accept the low risk position much easier as an expert's view, while egalitarian and communitarian position consider the high risk position as expert opinion.

The question of siting a disposal facility introduces a new paradigm in the attitudes relative to the risks of long-lived waste. Based on the work of Mary Douglas, it is possible to hypothesize that the environmental problematic of nuclear waste together with a certain conception of the nature radicalizes the view of a certain part of civil society relative to disposal. Indeed, the dissociation of the industrial site of nuclear waste production from the site of disposal presents the waste problem from a cultural perspective as an environmental problem and not as a security problem. Hence, the "social acceptability" of the technical solution becomes a problem in itself, independent of the characteristics of the technical solution. Thus, the question of disposal does not advance towards the extension of the technological controversy but towards its reduction over time to its "social" dimension. The analysis of the statements produced by opponents of geological storage puts into perspective the cultural dimension of the problem in the representation of risk. It is the symbolic issue of contamination of a pristine nature, which is one of the main arguments forwarded against disposal, not arguments about safety. In this perspective, we understand why the qualification of the site for the radwaste and the time factor become social rather than strictly scientific issues. Indeed, the debate is not about the robustness of scientific hypotheses but rather regarding a public choice confirming an irreversible disposal and therefore potentially beyond human control in the very long term. The relation of the population with its territory is historically and culturally

constructed. Thus, the qualification of a new site beyond industrial production sites as the nation's radwaste dump may be seen as a risk to this pristine territory even if there is no real radiological risk in any foreseeable future, just as our and many other experimental data seem to indicate at least for many thousands of years.

While recognizing the role of cultural cognition of scientific results and expertise, as well as acknowledging the historical and cultural perception of hazards to territories destined for disposal, one should avoid cultural relativism where everything becomes subjective and a social construction, and where an increase of scientific knowledge does not lead to an increase in rationality towards radiological risk assessment from nuclear waste disposal. One should not confound the diminishing credibility of experts with a diminishing credibility of science. Just the opposite, scientific results become more and more a common object in the conflicting views on the definition of risk [Beck, 2007], and they are also important for assessing hazards for the historically grown relation between citizens and their territory. However, despite large public interest, scientific results on nuclear waste disposal are still very poorly known among non-technical stakeholders in the field of nuclear waste disposal.

Translation of Scientific Risk Assessment to the Political Context

If the technical feasibility associated with the disposal of long-lived waste can be based on sound scientific data such as those exposed in this paper, the decision process for disposal remains a subject of controversy. In the United States, as well as in France, the problem of the choice of disposal sites for long-lived radioactive wastes and the territorialization of the problem contribute to a situation of non-decision [Barthe, 2006] in which two concepts of the risk are opposed. On the one hand, scientific modeling develops a performance assessment based risk prediction, on the other hand, the politicization of the project questions the simple fact of irreversible deposition of long-lived radwaste. These are two opposed lines of argument without common ground. In the French case, the scientific assessment of long-lived radwaste disposal poses no real problem as long as it is not subject to a political decision.

Indeed, as the work of Yannick Barthe demonstrated, scientific modeling is a way to keep the issue of disposal outside of space and time. As far as the risk is defined as an "anticipated hazard" [Lascoumes, 1993] it can be quantified by experimental programs such as ours, field studies and probabilistic calculations. These technical analyses serve as a base for expert opinion intended to sustain potential future decisions. The "discourse of safe management" [Gilbert, 1996] applied to risk assessment provides predictive assessments and standards. The decision for disposal would become a logical sequence combining present and future. Yannick Barthe indeed shows how technical temporality masks public perception of risk of nuclear waste disposal and adopted technical solutions remain undecided since they escape the public debate similarly as scientific observations as those provided in the paper. Until the 1990's the owners of the waste were responsible for finding a solution and disposal was considered as a scientifically and industrially acceptable solution. The "transformation into politics" [Barthe, 2006] changes both the mode of decision and the temporal character of the scientific and technical data and models on which it is based. The instruction of the research process by parliament ended a decision process based on the present / future relationship by establishing a process based on the exploration of the past to basic policy decisions.

The decision is part of a system of actions, in which time is a mobilized as a resource by different actors. The entry of waste management into politics, for example, has put the long-term perspective of disposal in competition with more short-term oriented options like transmutation or interim storage. The work of Yannick Barthe and Remi Barbier points out that "*the time is a resource of action against which the various stakeholders are not all equivalent*" [Barbier, 2007]. They wonder about the capacity of nuclear institutions to dispose of a "proper place" in the sense of Michel de Certeau, that is to develop a very long-term strategy to deal with opponents [Certeau, 1990]. In fact a geological disposal site may become such a "proper place", ideally isolated from history and protected by strong barriers from any exchanges with our world, governed by geological temporality extending to millions of years. Societal temporality is reintroduced in action and decision to configure this closed temporal space. The whole question is whether and how such

an isolated place can be created, buried in the history of the concerned territories as an isolated place beyond space and time.

3.6 Conclusions on Waste Disposal Acceptance

The simulation by percolation tests of some key safety features of a geological disposal site has provides important indicators for the safety indicators for clay rock. Credibility of the models used is discussed in the overall context of scenarios for the evolution of the safety of the barrier system. It has been shown that societal and technical evolution scenarios sometimes need to be mixed. Yet there is no direct link between increased scientific understanding of the repository system and a public position on nuclear waste disposal. Social sciences need to be more strongly involved into safety assessment, since safety is not a pure technical but as well a societal term. The political request for an alternative 'retrievable' waste disposal concept can be seen as a consequence of the ongoing socio-technical dispute. For the HTR waste management, it has to be acknowledged that the experimental evidence for the long-term behavior of disposed HTR spent fuel and irradiated graphite still needs to be further developed, for being comparable with the scientific knowledge base of spent LWR fuel. This will be discussed later.

4 Waste Streams Arising from V/HTR

HTR fuel elements can be either pebbles, each the size of a billiard ball, or prismatic blocks, each about 0.8 m tall. Pebble bed reactors contain hundreds of thousands of pebbles, while prismatic reactors contain about 1000 blocks. Each concept has advantages and disadvantages. Pebble bed concepts can use on-line refueling while block type reactors use batch refueling. Fuel distribution is controlled in block reactors in a deterministic way but only randomly in pebble bed concepts. The limiting power level determined by passive cooling constraints in accident conditions is about 50% higher than for pebble bed reactors. Block type cores also normally have significantly lower pressure drops. The waste streams for this type of reactors have been quantified in [Cellier 2006].

The prismatic block concept was selected for the ANTARES of Areva [ANTARES 2006]. The reactor system consists in a solid graphite core containing an annular ring of fuel elements comprising four basic regions: an inner replaceable reflector, the graphite substrate of the fuel, an outer replaceable reflector, and an outer permanent reflector (Fig. 4.1).

4.1 HTR Waste Management for ANTARES

Overall Strategy

A key challenge for the ANTARES project was to develop HTR technologies that efficiently manage radioactive waste, especially irradiated graphite because graphite is a key material for the HTR systems. This challenge had to be met through application of the following principles:

- Minimize the creation of waste through design,
- Minimize the impact of the waste once it has been created,
- Recycle to the maximum extent possible.

As graphite serves as both moderator and reflector, it makes up about 98% of the core volume. It was thus evident that managing the graphite waste stream was a key element in the successful commercialization of HTR. While limiting the impurities in the graphite has the greatest influence on the radionuclide content of the irradiated waste, a "back-end" solution was needed anyway, no matter how "pure" the graphite used was.

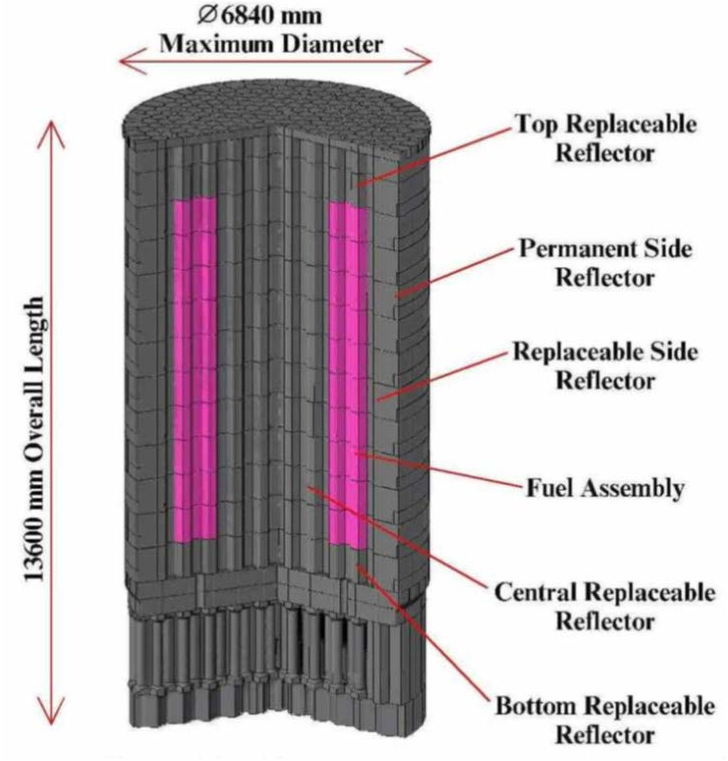

Fig. 4.1: Core structure of ANTARES

From a plant perspective, spent fuel elements and irradiated graphite reflector elements represent, by far, the largest component of the HTR waste stream. The total mass of graphite in the ANTARES including the graphite in the fuel compacts was 729 tons. Regarding fuel utilization, one-half of the core (510 assemblies) was discharged each refueling. Based on an 18-month fuel cycle, one HTR module would produce 20,910 spent fuel assemblies over its 60-year life. The graphite wastes produced include defueled graphite host blocks, irradiated replaceable reflector blocks, and secondary graphite wastes. The total lifetime discharge of irradiated graphite assuming 60 years of operation was approximately 6100 tons.

Several recommendations for future actions and overall strategic direction for the spent fuel storage program were developed. According to US practice, it had been assumed that spent fuel be stored on site. Initially, the spent fuel was to be stored in water-cooled storage modules adjacent to the reactor, then – after a short duration for cooling – transferred to either dry cask storage or other dry-type modular, expandable storage for intermediate / long term storage to take advantage of the potential economic gains of this configuration.

Ultimately, closure of the fuel cycle would have permitted full optimization of fuel use, especially when considering the symbiotic relation that could exist between LWRs, FBRs and HTRs relative to spent fuel and waste management. ANTARES spent fuel would have been eventually transported to a repository or reprocessing facility. The U.S., French and IAEA requirements/regulations that govern transport of LWR spent nuclear fuel would have applied to ANTARES. It was considered that HTR spent fuel transportation services would be similar to LWRs thus the current service options provided to the LWR industry could be used with minor adaptation. Eventually, the existing reprocessing capabilities could also be adapted to HTR fuel, albeit with the attendant development required. The work performed in that field is not reported here.

Irradiated Graphite Waste Management

The graphite fuel, moderator and reflector blocks have to be periodically removed and replaced with fresh material. The anticipated replacement frequency was three years for the graphite fuel, six years for both replaceable reflectors, and 60 years (life of the plant) for the outer permanent reflector. Radioactive graphite wastes would have required handling and disposal like other radioactive wastes.

Regarding irradiated graphite waste management options, the basic problem to be faced was two-fold. First, the irradiated graphite contains a radionuclide inventory (H-3, Be-10, C-14, Cl-35, Cl-36, Co-60, Ni-63, etc.) resulting in a waste classification that would be US Class C and French MAVL. Second, there was already a large worldwide inventory of irradiated graphite waste. With limited space and strict controls on waste facility radioactivity limits, direct disposal was not the favored option and with the significant amount of C-14, simple incineration was not feasible except maybe in the US.

Anyway, options did exist or would have existed to dispose radioactive graphite wastes, and facilities were needed for storage/disposal of irradiated graphite. Disposal options included surface burial, sub-surface burial, or deep geological emplacement depending on the requirements of the disposing nation.

Table 4.1 provides irradiated graphite discharge data for an ANTARES-like 600 MWt HTR that operates for 60 years.

Table 4.1: Irradiated graphite discharge quantities for a 600 MWt HTR operated for 60 years

Graphite Waste (Tons)

Reactor component graphite	Mass of component	Replacement period (yrs)	No of times replaced	Lifetime discharge
Fuel assemblies including compacts	116	3	20 (1)	2320
Inner/outer reflector + graphite above/below	353	6	10 (2)	3530
Permanent reflector	260	60	(3)	260
Total graphite	**729**			**6110**

(1) Half of assemblies in first and last cores operate 18 months
(2) Includes nine replacements plus the graphite remaining at license expiration
(3) Permanent reflector is not replaced but rather disposed of on decommissioning

Separation of the fuel compacts from their graphite host blocks would have reduced the volume of graphite that requires disposal as high level waste by more than 80%. Facilities for compact removal could have been located at a reprocessing plant, centralized facility, or even co-located at a HTR site. However, fission product contamination of the graphite host blocks could have led to dispose of them as high level waste. Methods to either avoid or mitigate the impact of potential contamination thus had to be developed. They include a de-fueling system design that minimizes the possibility that the compacts are damaged during removal, or if damage occurs, one that mitigates the consequences, and fuel blocks designed to facilitate compact removal.

Design/licensing of facilities for disposal of irradiated graphite was required in accordance with the regulations for radioactive waste disposal established by the various countries holding inventories of radioactive graphite wastes, knowing that requirements for irradiated graphite disposal differ from country to country. Direct burial of radioactive reflector blocks could have been possible in the U.S. because of less restrictive radioactivity limits and space availability. In Europe, the requirements for disposal may be more stringent (i.e., requiring disposal of this material in a sub-surface facility).

At-reactor storage of irradiated graphite was needed anyway to permit the decay of shorter-lived radionuclides (e.g. ^{3}H) prior to disposal offsite. The actual storage duration was governed primarily by the rules and disposal costs for the available disposal options.

Transport requirements for these wastes would depend on the level and type of activation (e.g., ^{14}C), possible contamination, if any, of the defueled host blocks, and acceptable sizes of blocks and particulates. In the U.S. it is likely that the graphite wastes would meet Class C waste requirements or less and could be packaged and transported accordingly. In Europe, packaging and transport requirements for graphite wastes would likely be commensurate with disposal requirements and may be more stringent because of the disposal requirements for ^{14}C.

It was also possible that graphite waste could be recycled. From a general point of view, whether from newly irradiated stock or legacy waste, graphite recycling would have offered a means for resolving the irradiated graphite waste problem. Although there would be impediments at each phase of graphite incineration/reforming, production, fabrication, and implementation, it was considered that the introduction of recycled graphite into HTRs might not be as difficult as it appears. With remote refueling, the handling of slightly irradiated fuel or reflector blocks should be viable and if the radioactivity level of the recycled graphite is low enough, it may be possible to construct the entire core from recycled graphite. Even if not, dummy graphite reflector and fuel blocks from virgin graphite could have been used to construct the first core, and during the commissioning phase, the recycled fuel and reflector blocks could have been introduced. The dummy blocks could have been used for successive reactor modules or for other plants, further reducing graphite wastes.

Thus recycling could have significantly reduced or even eliminated the need to treat irradiated graphite as a waste product. Some methods for recycling irradiated graphite waste that eliminate much of the volumes to be disposed of as waste were investigated in the ANTARES program:

- Annealing: whole blocks are annealed to sufficiently restore properties
- Pulverizing: grinding to a powder for processing and re-manufacturing into blocks with similar properties
- Reforming: blocks undergo pyrolysis in a high temperature reformer where the carbon as CO and CO_2 can be captured for further processing or used as feedstock to create new but still radioactive graphite.
- Incineration: blocks are burned and the CO and CO_2 generated are captured for further processing[2].

Although some of these methods appeared promising, they were in the early stages of development and required much R&D before they can be considered viable options. It must be noted that the FP7 CARBOWASTE program was dealing with some of these methods.

4.2 Spent Fuel Management

Main Requirements

The operational characteristics were such that the spent fuel management system should support refueling of four reactor modules on the same site within the minimum envisioned fuel cycle of 12 months. That is, the fuel elements discharged from a single reactor module should be placed into storage within approximately ten months after their discharge from the reactor and the time required to place the elements into the dry storage system can be no more than one month if there is a desire to avoid handling spent fuel during refueling activities. This assumes that placement into the system had to take place between successive module's refueling-related activities.

[2] Because of the significant amount of C-14, simple incineration of irradiated graphite – despite being technically feasible and radiologically acceptable – was considered to not be publically acceptable unless it was accompanied by a back-end process that separates C-14 from C-12 and precludes its release.

In order to support various fuel and core management strategies, retrieval of fuel elements from storage and replacement into subsequent operating cores did not have to be precluded (i.e., the design had to accommodate retrieval, however, not necessarily easy retrieval).

The initial capacity of the system had to be sufficient to accommodate the anticipated number of fuel elements discharged from four reactor modules over their first ten years of operation. This translates into a system capacity of approximately 15,000 fuel elements.

The system had to be capable of expansion to provide for storage of all of the fuel discharged from four reactor modules over the expected 60 year life of the plant without disruption of normal plant operation. This translates into approximately 100,000 fuel elements. In order to meet this goal, a separate, longer term storage facility had to be used; transfer of the fuel between storage facilities had to be taken into account and not impact the plant operation.

Storage System Assessment

The existing spent fuel storage facilities and systems were first studied in order to judge their applicability to the storage of ANTARES fuel elements and to try to estimate the design, economic and operational impacts should such systems be adopted for the ANTARES plant. Two existing storage systems might have been amenable to modification for storage of ANTARES fuel without significant design impacts. These systems are the Modular Vault Dry Storage (MVDS) system in use at the Fort Saint Vrain facility and the Transnuclear NUHOMS® canister-based storage system in use at many light water reactor sites.

Both systems could have easily supported the goals related to expandability and lifetime fuel storage. In addition, both were expected to require fairly extensive fuel element preparation and handling prior to final disposal. Nevertheless, considering in particular expected system costs, it was considered that neither system would represent ultimately an optimal storage solution and that exploration of fuel storage options not based on existing systems was warranted in that their expected higher design and development costs should be outweighed by lower construction and operational costs.

On-site Intermediate Storage Facility

A lot of work was related to the development of an on-site intermediate spent fuel storage system. This system receives irradiated fuel blocks from the reactor systems and stores them until they are shipped off-site. This system consists of a Core Offload Storage Facility (COSF), a Fuel Canning Facility (FCF), an Actively Cooled Storage Facility (ACSF), a Canister Loading Facility (CLF), and a (optional) Dry Cask Storage Facility (DCSF). With the exception of the DCSF, all of these facilities are located in the reactor service building.

An alternative to an ACSF was a passively cooled facility that is independent of the reactor services building. This passively cooled alternative would have been a modular “add on” design that could be continuously expanded to provide long-term storage in lieu, for instance, of NUHOMS depending on economic and/or other considerations. However, with a passive cooling system, some additional lag storage (e.g., enlarge the COSF) to permit spent fuel thermal aging could have been required prior to transfer to such facility.

The general flow of materials through the on-site intermediate spent fuel storage system was planned as follows: Irradiated fuel elements are received from the reactor systems by way of the fuel transfer tunnel and placed in either the COSF or the FCF. The fuel elements are sealed into FSCs in the FCF. At that point, the heat output of the Fuel Storage Containers (FSCs) might have been too great for them to be loaded into larger storage modules. In that case, the FSCs would have had to be stored in the ACSF until their heat output decreases sufficiently. When sufficiently cool, the FSCs could have been loaded into large-capacity canisters which are licensed for both storage and transportation.

Cask Systems

In the course of the ANTARES program, the design of the large-capacity canisters was actually based on the existing NUHOMS® canisters for LWR fuel. The canisters would have provided containment and convenient handling of multiple FSCs but would not have been designed to provide shielding.

The large-capacity canister could have stored 114 fuel elements in a 19-cell array of fuel storage tubes. A 19-cell array appeared to be an optimal size for an ANTARES canister. Each fuel storage tube would have contained one FSC. A fully loaded canister was projected to have a mass of about 43,000 kg.

Two designs were considered for the large-capacity canister. One is a tube-and-disk design (Figure 4.2), the other one a honeycomb design. The honeycomb design might be more efficient in that it provides a tighter spacing between FSCs. On the other hand, with a tube and disc design it might be possible to eliminate the hexagonal storage cells and simply machine openings in the stacked fuel basket disks to accommodate the FSCs. Both designs would have included shield plugs at each end to provide shielding during later transfer operations.

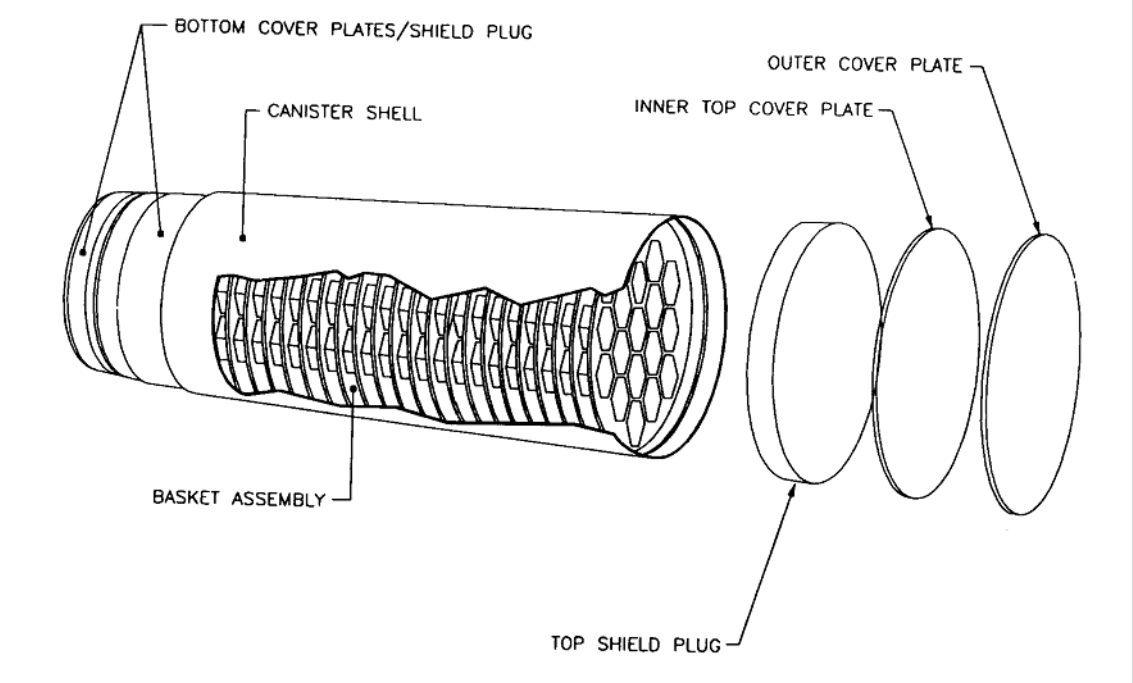

Fig. 4.2: Tube-and-disk design for large-capacity canister

If the irradiated fuel was to be stored on-site in a DCSF, the canister would have been placed into a transfer cask, the transfer cask and canister being placed in the high-radiation area of the CLF. In that design, the transfer cask is a thick-walled vessel designed to provide shielding during closure and transfer operations. The large-capacity canister is loaded with FSCs, the top shield plug is inserted, and the transfer cask and canister are moved to the low-radiation area of the CLF. The top covers are placed and welded to the canister. Finally, the transfer cask cover is bolted in place.

The DCSF includes a number of concrete HSMs. Each Horizontal Storage Module (HSM) can store one large-capacity canister. An HSM is a box-like, prefabricated, heavy walled structure with a removable shielded portal in the front to permit horizontal loading of the canister. Following loading, the canister rests on rails located inside the module. The heavy reinforced concrete side walls and top provide biological shielding and protection against natural phenomena. The modules are vented at the top and bottom to permit removal of the decay heat generated by the spent nuclear fuel stored therein. The projected dimensions of an HSM were approximately 5 m high × 3.4 m wide × 6.1 m long.

A transfer cask for a large-capacity canister would range from 2.4 to 2.7 m in diameter depending on the thickness of the neutron and gamma shields and would be at least 5.2 m long. The transfer cask would be larger than comparable LWR transfer casks because the ANTARES canisters were larger than their LWR counterparts. However, the weight of a transfer cask loaded with a canister of 19 FSCs (hook weight) would have been about the same as some of its LWR counterparts because less gamma shielding was needed for ANTARES fuel, and because an LWR transfer cask contains water when it is removed from the fuel pool. The mass of an empty transfer cask would have ranged from 54 to 59 tons.

If the irradiated fuel was to be moved off-site immediately, without storage in a DCSF, the canister loading operations would have been similar, but the transfer cask would have been replaced with a transport cask. In consideration of the projected dimensions and weight of a loaded transport cask, transport would have been performed by rail. The mass of a loaded transport cask might have reached 115 tons or more with impact limiters. These dimensions / masses are comparable to LWR spent fuel containers, such as CASTOR containers, and could encounter similar transport problems as, e.g., experienced in Germany.

4.3 Irradiated-graphite management issues

The ANTARES waste management program was dealing with irradiated graphite and spent fuel issues. In particular, developing a solution for irradiated graphite management was a prerequisite to the acceptance of HTR technology by utilities and plant operators. Nevertheless, it was considered that the irradiated graphite from the operation of the ANTARES could have been readily managed within the spectrum of disposal options currently available, because the question with irradiated graphite waste management was more of a perception than a technical issue, and would have not constrained the development of ANTARES. In addition, separation of the fuel compacts from their graphite host blocks in a dedicated facility would have drastically reduced the volume of graphite that requires disposal as high level waste.

Thus, options did exist or would have existed to store/dispose of radioactive graphite wastes. These options would include surface burial, sub-surface burial, or deep geological emplacement depending on the requirements of the disposing nation. Regardless of its source (i.e., fuel blocks or reflector blocks), the irradiated graphite is similar and, at the time of the Project, the options for its management were limited to on-site storage, as evidenced by the large graphite stockpiles that do exist (e.g., in France, United Kingdom et al), or to direct disposal in a surface repository. Other than the public concern over the mounting quantities of irradiated graphite being stored at multiple reactor sites, storage of irradiated graphite has not been problematic in the US. But with limited space and strict controls on waste facility radioactivity limits, direct disposal was an option only if dedicated surface disposal or sub-surface disposal facilities become available.

Recycling could have also significantly reduced or even eliminated the need to treat irradiated graphite as a waste product. But each of the form of processing envisioned warranted further study by the end of the ANTARES Project. Irradiated graphite, whether from new HTR or legacy graphite waste, could be considered a resource if suitable processing and fabrication programs could be developed and deployed.

However, it must be noted that the overall irradiated graphite discharge of 6110 tons per HTR module over its lifetime will represent a major waste management challenge. One four-module unit, e.g., will generate about 25,000 tons of irradiated-graphite, which is about the same mass as accumulated by all French graphite-moderated reactors having been operated in the past. In France, it is considered to build a special near-surface repository to dispose of this amount of irradiated graphite, as illustrated in Figure 4.3. Such a repository would be needed for each four-HTR-module unit, if no attempts would be undertaken to reduce the i-graphite discharge. It is evident that the shear amount of i-graphite discharges can be an acceptability issue for the HTR system and that the recycling and re-use of i-graphite may help to support a broader deployment of HTRs.

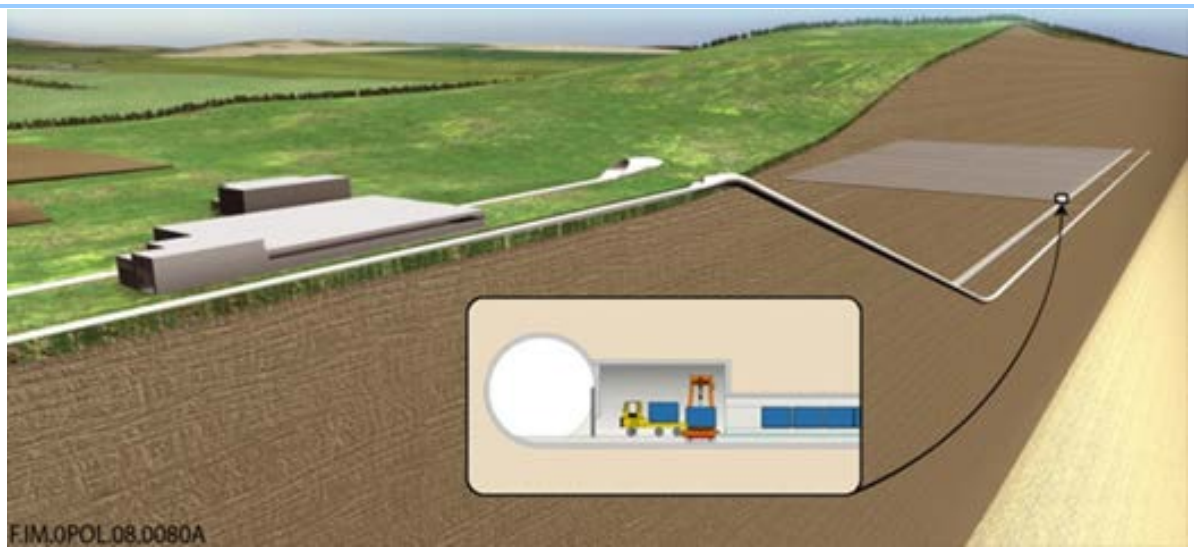

Figure 4.3: French design of an i-graphite repository for 25,000 metric tons capacity (source ANDRA)

5 Proliferation Resistance and Physical Protection (PR&PP)

Amongst the criteria for sustainability and public acceptance of nuclear reactors, the proliferation resistance of the fuel cycle and the physical protection of the nuclear plant against sabotage is regarded as a major issue and evaluated in the following chapter [Moses 2010] taking the methodologies of GIF and IAEA into account [GIF 2006, INPRO 2007, Petrunin 2008].

The design basis for all the HTR is targeted to achieve passive safety avoiding the release of fission products under all conditions of normal operation and accidents including beyond design basis events. This passive safety aspect of the design should make the HTR less vulnerable to a significant risk of "radiological sabotage" through malevolent acts and proliferation of fissile material. However, it must be noted that the uranium enrichment of HTR fuel is typically much higher as compared to LWR.

5.1 V/HTR design characteristics

Description of Block-type Fuelled HTR

The TRISO-coated particle fuel (see Figure 5.1) has a spherical ceramic fuel kernel of either uranium oxide or uranium oxycarbide, or mixed oxides of other actinides. The kernel with small-diameter (nominally 200-500 µm typically varying inversely with fissile inventory) is coated with four coating layers consisting sequentially of low-density porous pyrocarbon, inner high density pyrocarbon (IPyC), silicon carbide (SiC)[3], and outer high density pyrocarbon (OPyC). The coatings on the fuel particles serve as the primary containment preventing the release of fission products, and plant configurations and operating conditions are being designed appropriately to limit fuel temperatures during both normal operations and accident conditions so as to preclude the release of fission products. For block-type fuel, the coated particles are loaded into fuel compacts (sticks) held together by graphitized carbon. The fuel compacts are either loaded into holes in the hexagonal prismatic block fuel elements as in the US fuel design of stacked in a fuel pin which is then positioned into the bore holes of the block fuel elements as in the Japanese fuel design. Fuel elements are stacked in the reactor core with fissile and neutron burnable poison loadings tailored so that the power distribution is peaked toward the top of the core where the inlet cooling gas has the lowest temperature and the power density is lowest in the bottom of the core where the temperature of the outlet coolant is highest. The fuel and burnable poison loading patterns are set to maintain a preferably homogeneous radial temperature profile and to keep the peak fuel temperature below the limit for normal operation, which is 1250ºC for TRISO-coated fuel particles with SiC coatings.

[3] On-going research focuses on replacing SiC coatings with zirconium carbide (ZrC) coatings to achieve higher temperature limits (~2000ºC) for fission product retention during accidents and to reduce diffusion of radioactive silver.

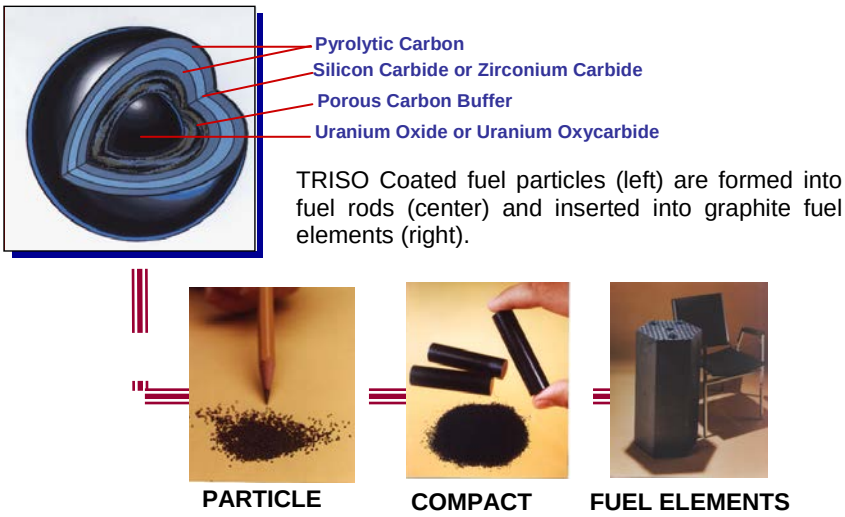

Figure 5.1: Illustration of coated particle fuel in the prismatic fuel element of US design

As described in the previous chapter, the spent fuel is retained in cooled storage containers that are embedded below grade and located adjacent to the reactor cavity. Prismatic spent fuel, which is unloaded from the core during periodic refueling shutdowns, can be tracked remotely by cameras viewing the serial numbers on the fuel elements during handling and storage operations. Since each fuel element is loaded with less than 4 kilograms of LEU, the plutonium content at full burn-up (~120 GWD/MT) will be small (~60-70 grams) and isotopically degraded compared to weapon-grade plutonium.

The current concepts for the energy utilization from the prismatic V/HTRs are based on either direct Brayton cycle for electricity generation or steam generators producing superheated steam or the use of an intermediate heat exchanger for heat utilization in either a cogeneration mode or a secondary Brayton cycle. The vessel configuration for the direct cycle GT-MHR and the reactor building option for the GT-MHR are illustrated in Figure 5.2 [General Atomics 2002].

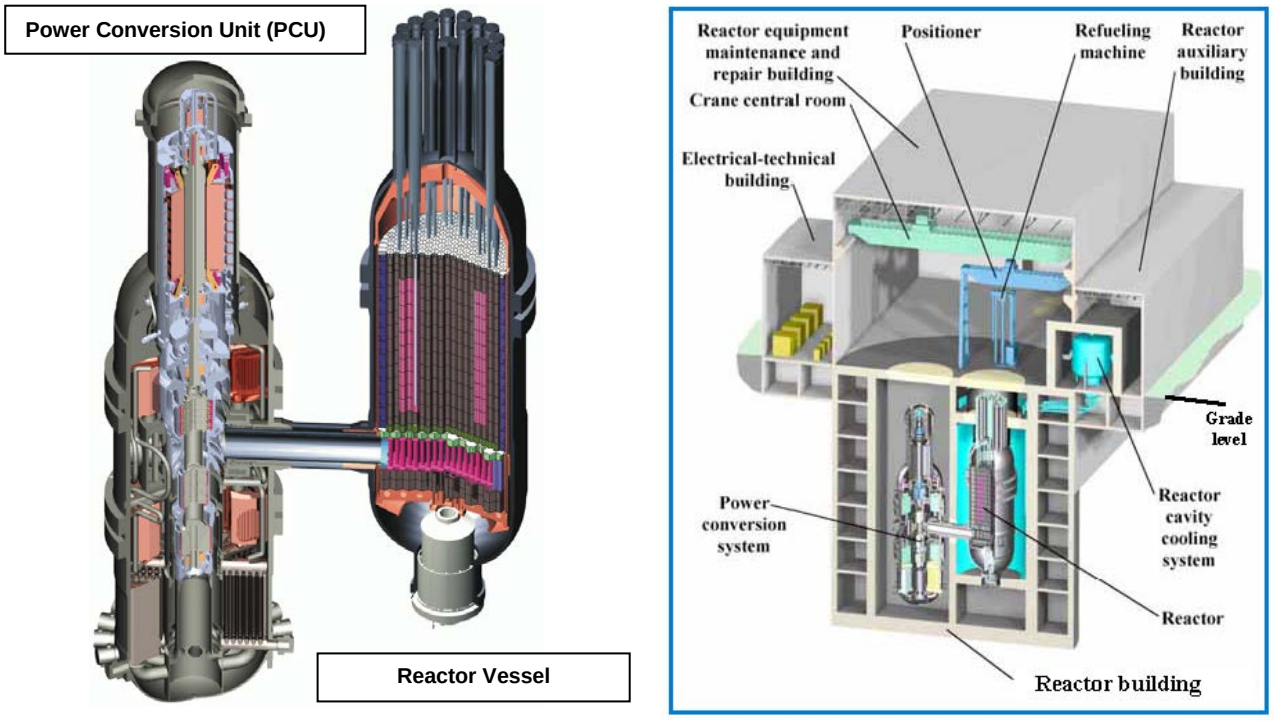

Figure 5.2: GT-MHR Reactor, cross-duct and PCU vessels (left) and fully-embedded reactor building (right)

For the GT-MHR, the reactor vessel and power conversion unit are placed sub-surface to improve and enhance the physical protection of the plant.

Description of the Pebble Bed V/HTR

There were two national programs for a pebble bed V/HTR that have been or are being pursued. The final choice on the fuel element design (prismatic or pebble bed) for the NHDD project of the Republic of Korea has not been made, yet [Chang 2007].

- South Africa: The Westinghouse and South African PBMR (Pty) Ltd. pebble-fuel, water-cooled RCCS, filtered confinement Pebble Bed Modular Reactor (PBMR) [Slabber 2004] that has been designed as a 400 MWt direct Brayton cycle plant as an NGNP candidate has recently been changed to a 400–500 MWt steam plant utilizing two 200–250 MWt reactors. The core for the 400 MWt PBMR was to be annular with an inner cylindrical graphite reflector; the 200–250 MWt core design would be cylindrical.
- Peoples Republic of China (PRC): The China Huaneng Group in a consortium with the China Nuclear Engineering & Construction Group (CNEC) and Tsinghua University's Institute of Nuclear and New Energy Technology (INET) is developing and preparing near-term (starting in 2010, commissioning in 2014) construction of the 250 MWt , steam-cycle High-Temperature Reactor-Pebble-bed Module (HTR-PM) [Zhang 2009] the HTR-PM, which builds on the success of the Tsinghua University's HTR-10 test reactor, is envisioned to be constructed in two module units producing 500 MWt and 200 MWe. The HTR-PM core is to be cylindrical. The reactor vessel is placed above grade level but the fuel handling system is situated below, underground.

The pebble bed reactors share the same passive safety features as the prismatic V/HTRs but have less excess reactivity due to on-load refueling. The LEU fuel for the pebble bed V/HTRs is contained in the same type of TRISO-coated particles compacted in small, 60 mm diameter spheres as illustrated in Figure 5.3.

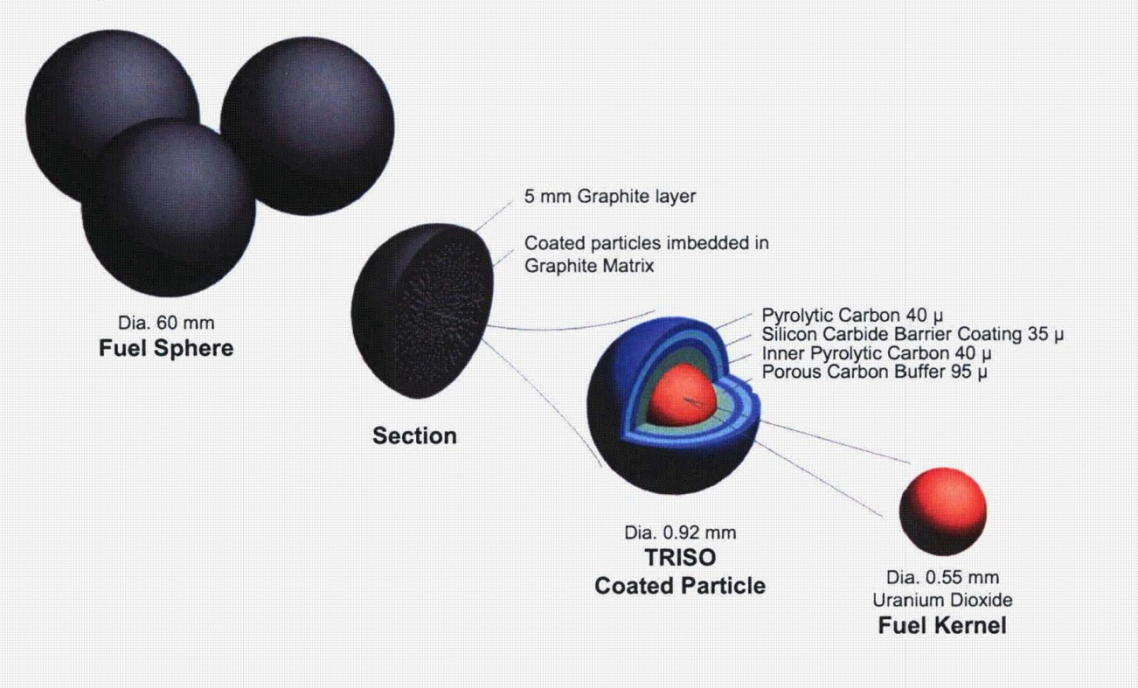

Figure 5.3: Illustration of coated particle fuel in a spherical fuel element

The pebble fuel is not tracked individually by serial number as in the prismatic core, but elements are counted, characterized, and checked following each of multiple recirculation until they achieve

the target burn-up based on radioactivity measurements. Following about six passes of each pebble through the core (valid for the 400 MWt PBMR) during on-line pebble recirculation, when pebble radioactivity indicates sufficient burn-up, the pebble is transferred to a storage container with a record kept of the number of pebbles transferred. Once pebble spent fuel is in the storage container, radiation monitoring is used to quantify by inference the amount of spent fuel present since, with no more than 0.12 grams of plutonium per pebble, it would take several tens of thousands of pebbles (or several metric tons by total mass and cubic meters by volume) to be diverted to constitute the basis for recovering a significant quantity of plutonium since a fresh PBMR pebble only contains 9 grams of LEU and a fresh HTR-PM pebble 7 grams. Furthermore, at a burnup around 90 GWd/t, the plutonium isotopic composition in the pebble spent fuel is degraded significantly from that of weapon-grade plutonium.

The reactor building and vessel arrangement for the 400 MWt Brayton-cycle PBMR concept is illustrated in Figure 5.4 showing the partially embedded reactor with the horizontal gas-turbine to the right of the reactor vessel and the associated spent fuel storage locations below-grade to the left of the reactor vessel. An additional confinement volume above the reactor supports the controlled and filtered venting in case of depressurization of the primary circuit.

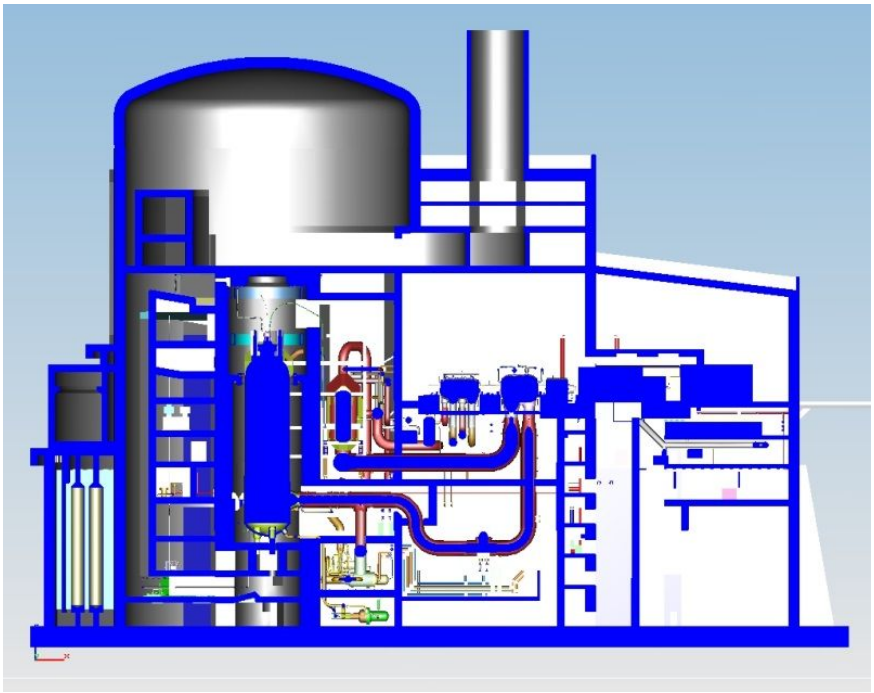

Figure 5.4: 400 MWt PBMR partially embedded reactor building with reactor vessel and turbine lay-down

The reactor vessel and vessel arrangement for the 2 x 250 MWt steam-cycle HTR-PM of China is illustrated in Figure 5.5 with the steam generator below and to the left of the reactor vessel but without indicating the location of spent fuel storage that is understood to be below grade [Zhang 2009].

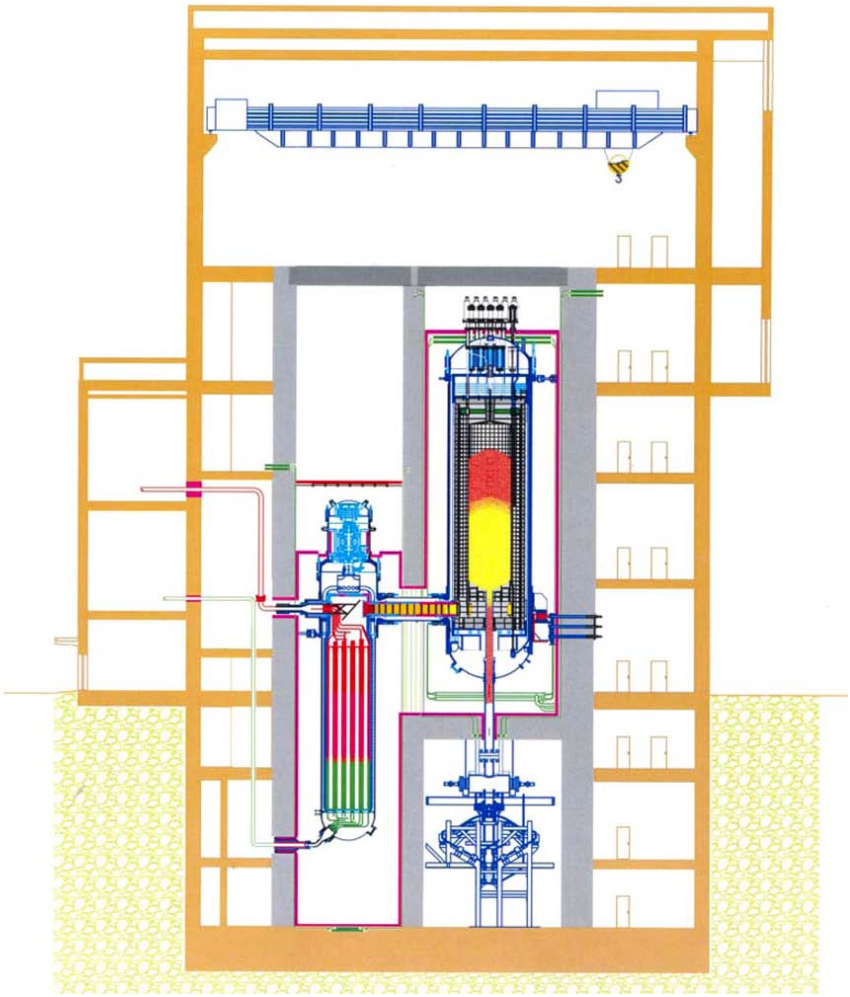

Figure 5.5: 250 MWt module of the two-module HTR-PM;
reactor building elevated above ground level with steam generator; spent fuel storage not shown

Overview of Fuel Cycle(s)

The baseline fuel cycle for the first generation V/HTR is the once-through fuel cycle using LEU fuel. The Russians are simultaneously pursuing the GT-MHR as a “deep-burn” option for weapon-grade plutonium (Pu) disposition. The use of high enriched uranium (HEU) as HTGR fuel, as was done in the past, is no longer considered acceptable due to the very low proliferation resistance of HEU although the HEU-Thorium cycle generates much less Minor Actinides (MA) as compared to LEU. Both GA and Areva were considering a range of other fuel cycle options for future reactor deployments including plutonium disposition and Transuranics (TRU) / MA transmutation and the use of thorium (^{232}Th) as a fertile component for high-conversion fuel. Each of these options, including the so-called deep-burn options, is based on an initial once-through irradiation without recycle, although technologies to reprocess and recycle TRISO fuel are also under either consideration or initial development and were studied extensively in the past at laboratory and pilot-scale for HEU/Th fuels. The ongoing research and development and the past experience provide a reasonably sound basis to have confidence in the ability to close the V/HTR fuel cycle in the future, if needed.

The fuel cycle options for V/HTRs can be categorized in three ways described as follows.

(1) V/HTRs can operate with either pebble or prismatic fuels. Pebble bed reactors operate with on-line refueling. This enables operation with very low excess reactivity, typically only sufficient to overcome the neutron poisoning effects of xenon that occur following power reductions. Prismatic fueled reactors require periodic refueling outages and thus operate

with substantially higher average excess reactivity, but allow substantially greater flexibility in fuel zoning and shuffling.

(2) V/HTR fuel cycles can be categorized by the types of fuel particles used, as follows:

a. LEU fuel particles with or without natural uranium fertile fuel particles

b. Pu fuel particles

c. TRU or MA fuel particles

d. ^{233}U fuel particles (or ^{233}U with ^{238}U)

e. Thorium (or thorium with uranium) fertile fuel particles

f. Pu/^{232}Th and/or Pu/^{238}U in mixed oxides (MOX)

The first four types of particles contain fissile isotopes that are required to support criticality of the reactor. The LEU particles also contain the fertile isotope ^{238}U and may contain in some designs fertile particles of natural uranium. However, with the V/HTR's thermal spectrum, thorium has substantially better properties as a fertile isotope, so, for core designs that add fertile material, thorium fuel particles may replace the use of natural uranium in the future. This thorium may be mixed with a small amount of uranium to dilute and "denature" the fissile ^{233}U produced by neutron absorption in thorium. In general, it can be expected that future V/HTR reactors will operate with fuels composed of some mix of the six particle types listed above. Each particle type involves specific technical issues for fabrication, with some being more challenging than others.

(3) V/HTR fuel cycles can be categorized by whether or not the spent fuel is discarded or recycled. Recycle may occur with either aqueous or pyro-reprocessing methods, and recycled materials may be returned to V/HTR's or sent to fast reactors.

Except for the LEU once-through cycle and the historic testing and use of HEU/Th, all other fuel cycles for the V/HTR represent future possibilities given also that there is likely to be several years and a significant financial investment for supporting research (including irradiation testing of laboratory-scale, pilot-scale and industrial-scale fabrications of candidate fuels) to qualify the fuel forms for the alternative fuel cycles. Currently, only LEU fuel is being tested for qualification so alternative fuel options are likely years away in development. Further, reprocessing technologies for V/HTR fuels are not currently developed except the specific head end process to separate the fuel particles from the graphite matrix and fuel kernels from the coatings. The grind-leach, burn-leach and electrolysis in nitric acid technologies studied for reprocessing HEU/Th fuels must still be developed beyond laboratory and pilot-scale and yield large quantities of ^{14}C-contaminated CO_2 or carbon sludge that must be treated, conditioned and disposed of safely.

5.2 PR&PP Relevant System Elements and Potential Adversary Targets

Key high-level information defining the V/HTR system elements includes (see Figure 5.6):

- Material types existing within each system element or grouping of system elements

– Fresh fuel: LEU in currently planned V/HTRs; weapon-grade plutonium in future Russian GT-MHR; plutonium or TRU in future deep-burn V/HTRs; and LEU/Th or Pu/Th MOX in future converter V/HTRs. As raw material for fresh fuel fabrication (LEU – uranium hexafluoride, nitrate, or oxide; Pu-bearing – plutonium metal, nitrate or oxide; TRU – nitrates or oxides; and LEU/Th – hexafluoride, nitrate or oxide), the material is most attractive since the least amount of effort would be needed either to enrich indirect-use uranium or to divert plutonium during nuclear weapon dismantling. Once encased in graphitized carbon as TRISO-coated particle fuel in fuel elements (pebbles or prismatic blocks), recovery becomes more difficult involving processes similar to reprocessing except for the presence of fission products and activation products such as ^{14}C, ^{60}Co etc.

– Irradiated and spent fuel with fission products: Irradiated LEU with small amounts of plutonium in currently planned V/HTRs; irradiated/isotopically-degraded plutonium in future Russian GT-MHR; irradiated TRU in future deep burn V/HTRs; irradiated LEU/Th with both plutonium and ^{233}U in future converter V/HTRs; and radioactive or other hazardous wastes especially those solid, liquid and gaseous materials collected for storage and/or disposal during operations and maintenance of any reactor type. Reprocessing is required to recover usable weapon-grade material, but, as is the case for all spent fuel, breakage and dispersal of radioactive irradiated fuel is potentially attractive as a means of effecting radiological sabotage.

- Operations envisioned to occur in a system element, and whether (and how) these operations can be modified or misused:

– Fresh fuel: Operations include fabrication by supplier, shipment from supplier to user, receiving and storage at user's site. Theft is an issue in each element dealing with fresh fuel for the acquisition of either indirect-use ^{235}U in the current baseline concepts or direct-use plutonium in future plutonium and TRU-fueled concepts, but recovery of weapon-usable material is more difficult involving processes similar to reprocessing except for the presence of fission products and activation products such as ^{60}Co. During fuel fabrication, a deliberate act or inadvertent error in quality control can lead to fuel failures and radioactive releases from the damaged fuel during irradiation as an act of radiological sabotage. Fabrication also involves scrap recovery and recycling within the supplier's fuel fabrication facility with non-recoverable scrap stored for disposition as low-level radioactive waste. Broken fresh fuel elements would be stored separately by the user for shipment back to the supplier for recycling as unirradiated scrap.

– Irradiated fuel: Operations include irradiation in reactor, recirculation (pebble bed) of irradiated fuel, reloading (prismatic) of irradiated fuel into reactor, irradiated and spent fuel storage, radioactive waste handling and storage, loading irradiated and spent fuel or radioactive wastes into shipping casks for off-site shipment, shipment of spent fuel or radioactive wastes for disposition, and processing of spent fuel for disposition by supplier. Diversion and theft of irradiated material is the principal threat for the acquisition of potential nuclear weapon-usable material (enriched uranium, plutonium, or ^{233}U) that still must be recovered from irradiated fuel elements by reprocessing and for uranium by both reprocessing and subsequent enrichment. Theft of smaller quantities of such material can result in their use for radiological sabotage. Sabotage of the reactor during fuel irradiation or shutdown operations or sabotage of spent fuel storage tanks or shipping casks can potentially lead to radiological releases to the environment as an act of radiological sabotage. Special handling of damaged irradiated fuel would likely include separated canned storage at the user's site with a negotiated disposition path including shipment back to the supplier.

- Material movements in and out of a system element:

– Fresh fuel fabrication: Raw constituents of fresh fuel are brought into the fuel fabrication facility (LEU – uranium hexafluoride, nitrate, or oxide; Pu-bearing – plutonium metal, nitrate or oxide; TRU – nitrates or oxides; and LEU/Th – hexafluoride, nitrate or oxide), and fabricated fuel elements (prismatic blocks or pebbles) containing graphitized-carbon-encased TRISO-coated fuel particles are shipped out. Fuel scrap is recycled internally at the supplier's fabrication facility with non-recoverable fuel scrap-waste stored on-site and ultimately shipped off-site for disposal as low-level radioactive waste.

– Fresh fuel shipment to, receipt by, storage at, and irradiation by user facility: Except for irradiation, all movement in and out of the listed system elements is that of fresh fuel; damaged fresh fuel is collected, stored and returned to supplier for scrap recovery. Diverted or stolen fresh fuel must still be processed using the same methods for reprocessing spent fuel to remove the carbon and SiC in order to recover the fissile material content.

– Irradiation, handling of irradiated fuel for recirculation (pebbles) or reloading (prismatic) or spent fuel storage, spent fuel storage, loading for off-site shipment, shipment for

disposition, and the handling and storage of radioactive wastes: Except for irradiation and radioactive waste handling and storage, all movement in and out of the listed system elements is that of irradiated or spent fuel. Radioactive waste handling and storage involves all fluids and solids that are contaminated with fission and neutron-activation products that end up in either the off-gas system or the solid and liquid waste systems for storage and ultimate disposition as low-level radioactive waste; the wastes are primarily generated by the helium purification system and routine maintenance of the reactor and auxiliary systems.

- Safeguards and security envisioned to exist in the system elements:

– All facilities and operations will be subject to the provisions of the supplier and user states' Safeguards Agreement with the IAEA, the Additional Protocol, the Convention on Physical Protection of Nuclear Materials for international shipments of nuclear material, the Agency's guidance on The Physical Protection of Nuclear Materials and Nuclear Facilities, and the nuclear material, equipment and technology transfer commitments of the members states of the Zangger Committee[4] and the Nuclear Suppliers Group[5].

– Within facilities, measures shall be taken to assure Containment and Surveillance (C/S) and the Continuity of Knowledge (CoK). For prismatic fuel V/HTRs, CoK shall be established by the visual tracking of serial-numbered fuel elements from fabrication to disposition. For pebble fuel V/HTRs, CoK shall be established by counting of fresh fuel elements and by bulk accountability methods that may include for spent fuel both counting elements and active neutron interrogation techniques to quantify the fissile inventory to within acceptable levels of uncertainty.

Assuming that the most likely adversary targets are those presented in the user state, then fresh fuel is the most likely target for theft or diversion. For this reason, LEU and LEU/Th fuels are best suited for export to user states containing only indirect use ^{235}U in LEU with the use of Pu-bearing fuels reserved for use in supplier states for weapon material disposition and deep-burn of TRU. However, recovery of usable nuclear material from either fresh or irradiated V/HTR fuel involves the processes needed to remove large quantities of carbon for a small yield of nuclear material.

[4] Zangger Committee!

The Committee, named after its first Chairman Prof. Claude Zangger, was formed following the coming into force of the Nuclear Non-Proliferation Treaty (NPT), to serve as the "faithful interpreter" of its Article III, paragraph 2, to harmonize the interpretation of nuclear export control policies for NPT Parties.

The Committee has been focussing on what is meant in Article III.2 of the Treaty by "especially designed or prepared equipment or material for the processing, use or production of special fissionable material." The Zangger Committee maintains a Trigger List (triggering safeguards as a condition of supply) of nuclear-related strategic goods to assist NPT Parties in identifying equipment and materials subject to export controls.

Today the Zangger Committee has 39 members including all the nuclear weapon States.

[5] The Nuclear Suppliers Group (NSG) is a group of nuclear supplier countries that seeks to contribute to the non-proliferation of nuclear weapons through the implementation of two sets of Guidelines for nuclear exports and nuclear-related exports.

The NSG Guidelines also contain the so-called “Non-Proliferation Principle,” adopted in 1994, whereby a supplier, notwithstanding other provisions in the NSG Guidelines, authorises a transfer only when satisfied that the transfer would not contribute to the proliferation of nuclear weapons. The Non-Proliferation Principle seeks to cover the rare but important cases where adherence to the NPT or to a Nuclear Weapon Free Zone Treaty may not by itself be a guarantee that a State will consistently share the objectives of the Treaty or that it will remain in compliance with its Treaty obligations. At present, 48 member states.

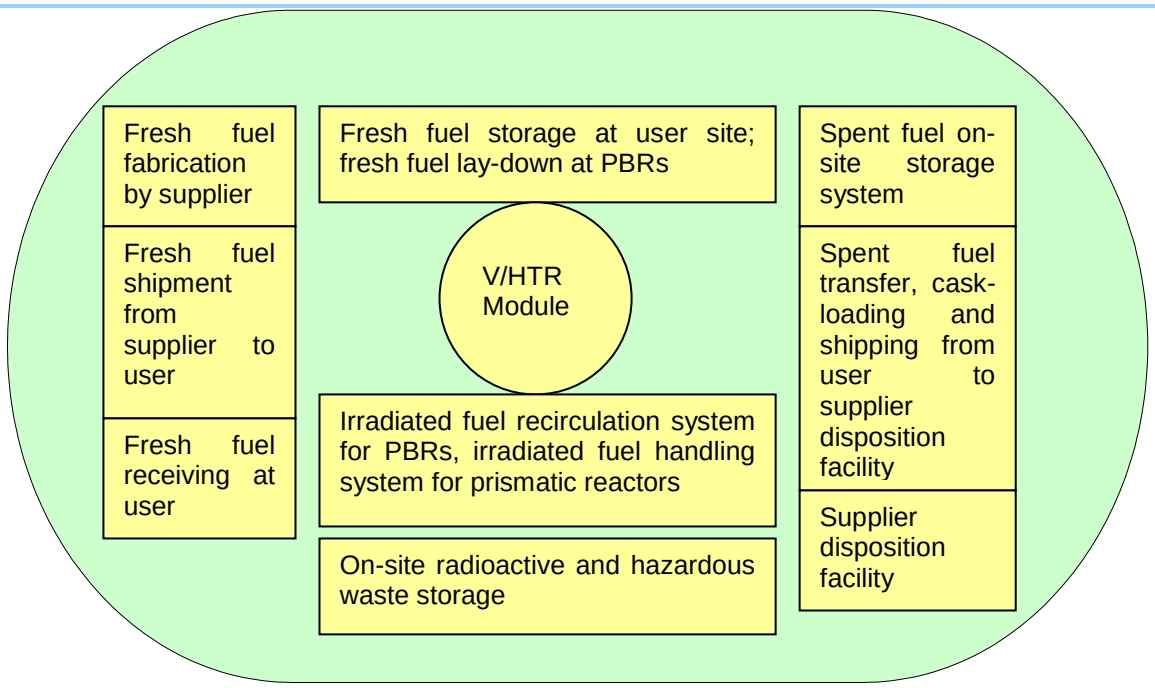

Figure 5.6: Diagram of V/HTR nuclear system elements

5.3 Proliferation Resistance Considerations Incorporated into Design

The key proliferation resistance feature of the V/HTR fuel system is the fuel itself. To obtain a significant quantity of either indirect-use ^{235}U from LEU or direct-use plutonium, one has to process several metric tons and tens of cubic meters of carbon encasing coated particles using either grind-leach, burn-leach of electrolysis in nitric acid. The most attractive fuel form would be weapon-grade plutonium oxide proposed for plutonium disposition in Russia, and this fuel form would most likely not be exported out of the country. If, in the future, MOX or TRU oxides for deep-burn were to be qualified as licensable fuel forms, such fuel would also be less likely to be exported until LEU availability runs out, by which time advanced safeguards will have been developed and implemented for the export of potential breeder fast reactors.

Concealed Diversion or Production of Material

For large quantities of materials production (plutonium or ^{233}U), this would likely be detectable by spent fuel accountancy based on radiation monitoring or fuel element counting, by containment and surveillance on fuel storage, or by recorded reactivity deviations in reactor operations. The production of small quantities for research would not likely be detected so readily. The V/HTR does not produce readily accessible, attractive fissile material, and the technologies for reprocessing coated particle fuels are both more complicated and still require development.

Breakout

As noted before, reprocessing has yet to be demonstrated for the coated particle fuels on an industrial scale. If there are multi-lateral contractual provisions for the supply of fresh fuel and the take-back of spent fuel for an exported V/HTR, the issue of breakout is further mitigated since there will be either no such material or limited quantities of material to be reprocessed in the user state, but this depends of course on the frequency of take-backs and either IAEA or bilateral inspections of the spent fuel storage facilities.

Production in Clandestine Facilities

The most likely breakout is the use of the skills obtained in operating a foreign-sold V/HTR to construct and operate a much simpler graphite-moderated production reactor. The use of a multi-billion dollar exported V/HTR under international safeguards and with a fuel take-back contract likely offers only one real opportunity for proliferation, and that is the proliferation of enabling technical knowledge to do something much simpler to achieve the production of weapons-usable plutonium outside the view of international on-lookers. However, this type of information proliferation is unavoidable since it is needed to assure the host country's ability to operate the V/HTR both safely and economically. Even if the supplier-state provides the operators, the host state would still likely require the training of a small staff of knowledgeable regulators unless the IAEA or some other international group is to provide the independent regulatory oversight which would still likely have staff members from non-weapons states including the host state**.**

5.4 Physical Protection (PP) Considerations Incorporated Into Design

This section provides a qualitative overview discussing those elements of the V/HTR system design that create potential benefits or issues for potential sub-national threats.

Theft of Material for Nuclear Explosives

Any plutonium, or ^{233}U in future LEU/Th cycles, would be in highly radioactive spent fuel encased in coated particles with fission products, where the material of interest would be quite dilute so that the theft of a significant quantity would require the theft of metric tons of ^{14}C-contaminated (along with leaked fission products and other minor activation products) graphite and/or graphitized carbon containing the coated particles. Obtaining access to a significant quantity of plutonium or ^{233}U in the stolen spent fuel would require substantial effort of both mechanical and chemical processing. The resulting product would possess less than desirable nuclear characteristics, namely, either plutonium with a high inventory of the heavier plutonium isotopes or ^{233}U with hundreds of ppm of ^{232}U. The latter would be highly radioactive and require further chemical cleaning to remove radioactive decay products that would then reappear within a matter of hours to days after processing. It is judged that the intrinsic qualities of V/HTR spent fuel would not make it a desirable target for theft by a subnational group for nuclear explosives. Deep-burn fuels containing Pu or TRU/MA and thorium fuels containing ^{233}U without ^{238}U diluent could be potential targets for theft, particularly during transportation. In this case pebble fuels may have a PP advantage, since it is possible to partially irradiate them with an onsite reactor before transportation, which could use the same shielded canisters used for the return of spent pebble fuel. Another alternative would be to add radioactive spikants to the fuel during fabrication as was previously considered for HEU/Th fuels, but this would increase fabrication costs and need to be assessed. The most economical and practical approach for exported fuels is most likely the use of LEU fuel.

Radiological Sabotage

As discussed above, the V/HTR is designed based on achieving a high degree of passive safety by the use of a robust fuel in a reactor that maintains the fuel temperature below fuel damaging temperatures under all conditions of normal operations and accidents including beyond-design-basis events. The design vision used is that, even if the safety-related RCCS is compromised, heat will still be dumped from the external wall of the reactor vessel such that sufficient heat is removed into surrounding structures so that fuel temperatures in the core do not exceed the levels that would cause the loss of the primary containment provided by the SiC (or ZrC) coatings on the fuel particles.

However, the possible objectives for "radiological sabotage" by insider threats could include:

- Sabotage of fuel quality and quality control/assurance checks at the fuel fabrication plant in the supplier state.
- Sabotage from an insider or intruder threat in the host state either by causing a core heat-up accident by sabotage of shutdown cooling systems or by the deliberate introduction of corrosive chemicals into the primary coolant with simultaneous sabotage of primary coolant contaminant detectors/instrumentation and simultaneous sabotage of the helium purification system.

Neither of these strategies would be expected to cause significant off-site consequences but could be very expensive to recover due to lost operations and repair costs and would be highly detrimental to public confidence.

Another potential insider or intruder threat resulting in radiological sabotage outside the reactor boundary might best be accomplished by stealing small quantities of spent fuel and then explosively smashing it into smaller chunks or pieces in small but strategically-located areas.

Based on the discussions above, the most crucial aspects of preventing radiological sabotage in reactor operations are those steps, including physical protection and access controls to sensitive areas on-site to preclude both insider and intruder threats, to assure the following:

- Quality controls at the fuel fabrication plant in the supplier nation.
- Proper maintenance, inspections and protection of (i) the helium supply and the helium supply station to prevent the introduction of corrosive chemicals, (ii) the primary coolant contaminant monitoring equipment to detect the introduction of such chemicals, and (iii) the helium purification system to remove contaminants.
- Careful maintenance, inspections, testing and protection of reactivity control systems to assure the capability to achieve safe hot and cold shutdown and, if required, accomplishing the same function from a secure remote location.
- Careful maintenance, inspections, testing and protection of the safe-shutdown cooling system circulator, heat exchanger and power supplies so that, while not safety-related equipment but rather investment-protection equipment, loss of normal cooling upsets do not always lead to reliance on the safety-related reactor cavity cooling system, in which fuel temperatures can approach their limits for accident conditions.
- Physical protection of and controlled access to fresh and spent fuel storage locations and to the inbound and outbound transportation loading systems and the transportation of the fresh fuel from the fuel fabrication facility and of the spent fuel to its processing or disposal facilities.

5.5 PR&PP Issues, Concerns and Benefits

The key areas of known strength in the V/HTR concept at this time are its robust fuel, high burnup, and the use of the once-through LEU fuel cycle. Future plans for integration and assessment of PR&PP for the concept need to address (1) emerging issues from the PR&PP aspects of alternative fuel cycles that have yet to be realized and (2) the inclusion of PR&PP aspects in the development of coated particle fuel reprocessing technologies that are as yet not fully developed and demonstrated.

For proliferation resistance, the coated particle fuel encased in graphite and/or graphitized carbon is the major barrier to the ease of recovery, but additional aspects such as adding a radiation barrier or an enhanced radiation barrier not easily removed requires attention both from basic R&D and from engineering development that will impact the cost of fabrication and handling fresh fuel. The key question is whether such radiation barriers are needed or will make the cost of using the fuel too expensive for practical implementation.

In addition, the benefits of a below-grade siting in terms of enhanced physical protection versus cost of construction needs to be assessed in detail for each concept.

In conclusion, it can be stated that spent HTR fuel is not a desirable target for theft due to its intrinsic qualities. Spent fuel is highly radioactive. Obtaining a significant quantity requires the theft of metric tons of contaminated graphite and/or graphitized carbon containing the coated particles. Accessing to Pu or ^{233}U requires substantial effort of both mechanical and chemical processing with a resulting product of less-than-desirable nuclear characteristics, namely, either plutonium with a high inventory of the heavier plutonium isotopes or ^{233}U with hundreds of ppm of ^{232}U, making it highly radioactive and requiring further chemical cleaning to remove radioactive decay products that would then reappear within a matter of hours to days after processing. Deep-burn fuels containing Pu or TRU/MA and thorium fuels containing ^{233}U without ^{238}U diluent could be potential targets for theft, particularly during transportation. Partially irradiating them with an onsite reactor, or adding radioactive spikants to them before transportation can protect them from theft.

6 The Option of Geological Disposal of Spent V/HTR Fuel

The generation of nuclear waste from nuclear power plants including the High-Temperature Reactor (HTR) and the Very High Temperature Reactor (V/HTR) is a major concern with regard to sustainable nuclear energy use. Waste management and disposal issues have to be incorporated into the design of future reactors fuels and their associated fuel cycles, and will be an indispensable component of the licensing process, in many countries. This issue is specifically identified as a key technology goal of Generation IV nuclear energy systems [USDOE 2005], namely to:

"Minimize and manage their nuclear waste and notably reduce the long term stewardship burden in the future, thereby improving protection for the public health and the environment."

Generation IV reactors, including the V/HTR therefore need advanced back-end concepts improving the isolation capabilities and reducing the disposal volume requirements in final repositories. Certain aspects of the back-end of V/HTR reactor fuel cycles were already studied in the former EURATOM Framework Programmes as well as, e.g., in Germany. The objective of further R&D was to provide an overall assessment and optimization of disposal options, such as embedding of coated-particles in suitable matrices, development of container concepts, etc., and the spent HTR/V/HTR fuel disposal performance.

To study various disposal options of spent V/HTR fuel waste packages, it is necessary to analyze the behavior of advanced coatings (ZrC) as well as alternate kernel compositions (UCO) to investigate failure mechanisms of coated particles during geological disposal by applying fuel modeling codes and to improve geochemical modeling of HTR/V/HTR fuel.

The investigations were restricted to consideration of low enriched fuels (LEU) in the once-through fuel cycle which may be an initial choice until interim solutions (separation of spent coated-particles and moderator) or closed cycles will be introduced for a larger fleet of V/HTR [Grenèche, 2004]. Techniques for closing the HTR fuel cycle and the 'Graphite Cycle' were investigated in the CARBOWASTE project [Lensa, 2008].

6.1 Characteristics of V/HTR Waste

The largest fraction of radioactive waste activity is located in the spent HTR fuel elements. There, the coated fuel particles retain the fission products and residual uranium, plutonium and Minor Actinides. The coated fuel particle is a microsphere of around 0.9 mm diameter. The inner kernel consists of either uranium oxide, carbide or a mixture of the two. This is surrounded by a low density carbonaceous buffer layer, followed successively by layers of pyrolytic carbon (PyC), silicon or zirconium carbide (SiC or ZrC), and PyC. The layers act as primary boundary for the retention of fission products both in the reactor and during disposal. The coated particles are embedded in the moderator made from further carbonaceous materials, pressed together to form tennis-ball sized pebbles or fuel compacts. It should be noted that the irradiated fuel matrix also contains fission products from residual uranium impurities or defect coated particles and activation products from other impurities like lithium, chlorine etc. plus radiocarbon produced by activation of ^{14}N, ^{13}C and ^{17}O. However, this chapter will mainly concentrate on the release mechanisms from the spent fuel kernels via the coatings and the 'porous' fuel matrix.

An important issue in development of back-end options is the generation of a detailed characterization of the radionuclide distribution within the different parts of the irradiated fuel. Many characterization data have already been collected in the frame of the German HTR waste management programme and key data were made available (see additional deliverable D33.21-b). Initial U contaminations of the AVR fuel (A3) matrix materials were less than a fraction of 10^{-4} (slightly higher for THTR fuel) of the total U contents, signifying that only a very small fraction of fission products is created during irradiation outside the fuel kernel. Mechanical strength of fuel spheres slightly decreases with neutron fluence. Graphite oxidation due to water traces lead to a

weight loss of about 0.8 wt%, during operation. The typical effects in coated particle fuel from neutron irradiation are kernel swelling, pressure build-up, densification of buffer layer, gap forming layer disconnection, reduction of SiC tensile strength, all of which may increase the probability of a particle failure. Significant coating failure during irradiation was not observed. However, release by diffusion through the coating layers has to be encountered e.g. for the fission product Ag already at temperatures above 1000°C.

The integrity of the spent fuel particles during long-term disposal is impacted by geomechanical, radiolytical, geochemical and near-field processes which are specific to the choice of the repository (salt, granite, clay, etc) and the waste package.

6.2 Back-end Options

The back-end options for spent HTR fuel essentially consist of four distinct routes (see Fig. 6.1):

1) Conditioning and direct disposal of spent fuel blocks or pebbles.
2) Separation of fuel compacts from graphite blocks, followed by conditioning and disposal of compacts, and disposal or potential treatment of moderator graphite.
3) Additional separation of particles from compacts or pebble matrix, followed by conditioning and disposal of particles and compact/matrix graphite or potential treatment/conditioning of moderator material.
4) As in option 3, but additional removal of particle coatings in order to get access to kernels for reprocessing to remove fission products and potentially recycle uranium, plutonium and/or minor actinides (closed fuel cycle).

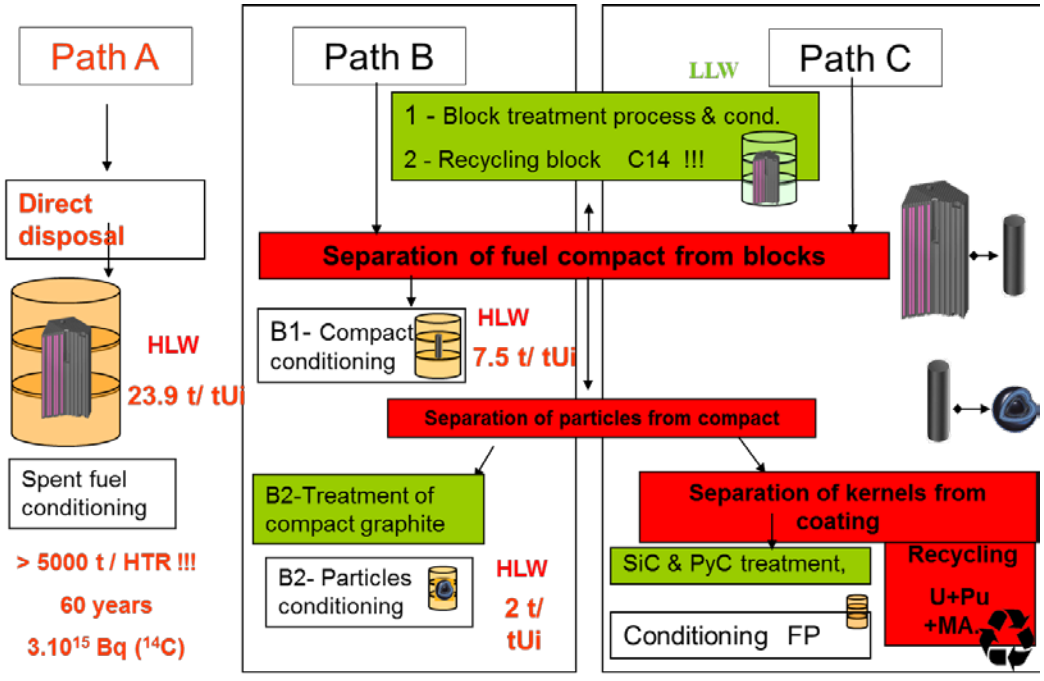

Fig. 6.1: Spent V/HTR fuel management options

Untreated spent HTR fuel from a commercial power reactor (Path A) will contain high initial concentrations of long-lived radionuclides and high levels of decay heat output and will therefore be classified as HLW. Thus, deep geological disposal will be the only waste management route available. A detailed study of the engineered barrier systems, beginning with the TRISO coating containing the fission products, and extending to the graphite matrix and the packaging container is

required, alongside with the repository design. The main phenomena to be studied for the fuel element barriers and the penetration of brines or water as well as the radionuclide mobilisation are illustrated in Figure 6.2.

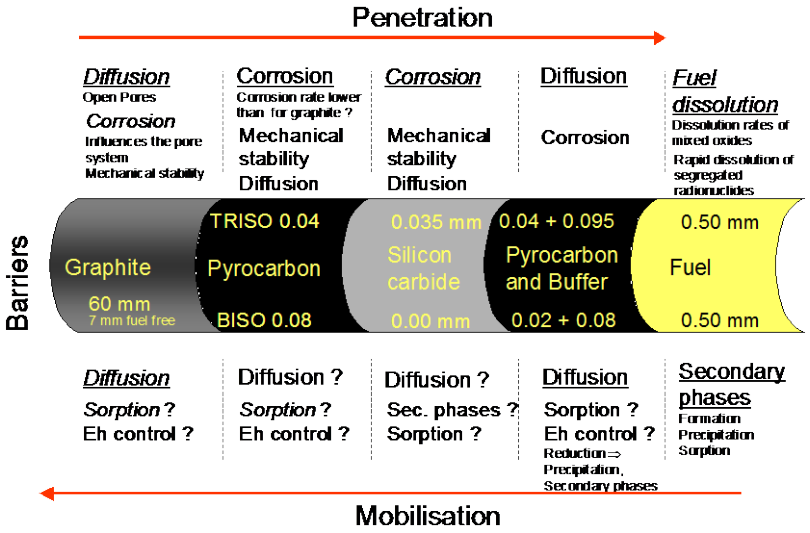

Fig. 6.2: Penetration of leachants and mobilization of radionuclides in a spent HTR fuel pebble

If HTR fuel is separated into block graphite and compacts (Path B1), or furthermore, if coated particles are separated (Path B2), the disposal strategy for the moderator graphite needs to be investigated specifically as was done in the European CARBOWASTE project. The disposal of spent compacts, fuel pebbles or separated coated particles is impacted by the radiological characteristics and thermal output of the conditioned waste which still needs to be isolated by specific waste packages, waste containers and other engineered barriers for geological disposal.

The reprocessing option (Path C) additionally requires stable confinement matrices for the separated (contaminated) coatings. The LEU kernels can be reprocessed in the existing facilities, except that the residual uranium enrichment is different from LWR spent fuel. Extracted fission products and residual actinides can be conditioned by vitrification techniques which are already industrial reality for reprocessed LWR fuel.

Container Concept

Any disposal option requires also a reasonable container concept. The container must provide absolute isolation of the waste from groundwater for thousands to ten-thousands of years. Existing container concepts were used for a concept study. A starting point is the CASTOR THTR container [Laugh, 1997] proposed by GNS to be used both for interim storage and final disposal of HTR waste (see Figure 6.3).

A single CASTOR cask contains about 2100 graphite pebbles. However, with its 37 cm container wall thickness (cast iron), this is over-engineered, e.g., as disposal container for clay formations. The reference case with a single HTR reactor of 600 MWt with 60 yrs of operation corresponds to about 9 million fuel pebbles or 4300 Castor containers = $1.2 \cdot 10^5$ metric tons of cast iron or 1.4 million m^3 hydrogen generation by slow long-term corrosion. This mass of iron is about 1/3 of the total iron used in the actual French repository concept for the whole French nuclear power generation, also for about 60 years. Thus, this option will not be viable for a larger HTR fleet.

Fig. 6.3: THTR/AVR CASTOR containers stored at Forschungszentrum Jülich, Germany

Therefore, a reduction of wall thickness would be required. The CASTOR container possesses an inner fuel canister which does not provide shielding, and may only be disposed of directly into salt formations but not in clay or granite due to radiation effects on the rock, fast corrosion, etc. It seems reasonable to put this fuel can into an overpack container of about 11 cm of iron wall thickness. Such a thickness should allow tightness for about 10,000 yrs in a repository, e.g., in clay formations. This thickness is also proposed by ANDRA in the French direct disposal concept for spent LWR fuel. The total mass of iron in such a concept would be about 4 times less than the one used with the original CASTOR.

For performance calculations, it was decided to consider a 'slimmed' version of a CASTOR type container; while the inside dimensions of the CASTOR container are kept the same, i.e. a length of 1964 mm and a diameter of 640 mm, its wall thickness is assumed to be only 110 mm. The CASTOR container can accommodate 1 THTR canister. The inside dimensions of the THTR canister are a height of about 1600 mm and a diameter of 590 mm with a volume of 437 liters.

A disposal concept considering such modified containers filled with spent fuel element pebbles still leads to an enormous amount of metallic material, even with this reduced wall thickness. A disposal concept considering a container filled with confined fuel compacts or separated TRISO particle seems to be more realistic. However, in the latter case, geometrical sintering constraints for a homogeneous final waste product have to be taken into account.

Impact on Repository

For treated or non-treated spent fuel wastes, deep geological disposal is the only waste management choice if reprocessing is discarded. Repository performance is influenced by the disposed wastes due to heat production and the consequences of disposal on hydrologic and thermo-mechanical properties. Only heat generation and hydrological constraints (resaturation times of void spaces by ground water) are subject of this chapter.

A key property of the waste is the heat generation. It is normally of primary concern for the identification of the required repository volume. This is because maximal temperatures in all repository concepts are limited. In clay, for example, the maximum temperature is 100°C.

However, the heat load of directly disposed spent HTR fuel in CASTOR containers is extremely low (~ 3°C), due to the fact that only 0.5% of the waste volume is spent LEU fuel, around 96% is fuel matrix and the rest are coatings. Thus, the disposal volume is defined by the containers, as such.

Therefore, the question was studied whether – in contrast to complete spent fuel spheres – dense packing of coated fuel particles in a suitable confinement matrix (option 3) would lead to non-acceptable heat generation. The reference spent fuel type considered for the evaluations made is an LEU UO_2 fuel with an initial enrichment of 10% ^{235}U and an average burnup of 76 GWhd/tHM. The considered disposal container for both cases is the slimmed version of a CASTOR type container, as described above. Both direct disposal of pebbles and the disposal of separated coated particles are investigated.

For the containers filled with TRISO particles, the maximum temperature increase is 67°C for a preceding 10-years cooling time and 32°C for a 50-years cooling time (see Figure 6.4). The calculations have been done with an inner diameter of the container of 590 mm; lower temperatures are expected if the diameter is 300 mm. The maximum temperature occurs 15 years after disposal. This shows that the heat production is acceptable, whatever back-end option is taken.

Directly after emplacement of the waste, the materials in a disposal gallery are strongly desatured (signifying that void spaces are filled with air or other gas and not with ground water). After sealing of the disposal gallery, infiltrating groundwater will resaturate the near field of the repository. However, the very low hydraulic conductivity of clay will strongly limit the flow rate of the infiltrating groundwater. It will thus take some time to reach complete resaturation in the near field in a repository excavated in a clay layer. Resaturation can be considered as a trigger for the start of important near field processes such as corrosion and radiolysis. Resaturation times are estimated to be in the order of 60 years up to some centuries depending on the specific site and depth.

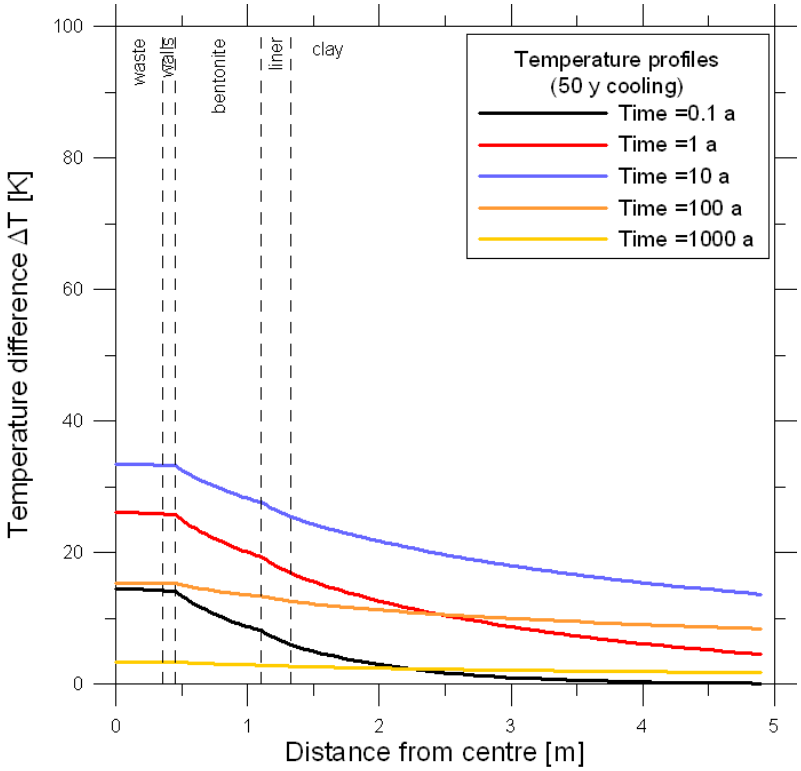

Fig. 6.4: Temperature profiles in and around the repository for containers filled with TRISO particles for a 50-years cooling time.

Confinement Matrices

Considering the feasibility of the segregation of coated particles as well as disposal of separated particles from a thermal and hydrological point of view, the confinement of the spent coated particles in a stable and largely impermeable matrix provides an interesting option to reduce the large waste volumes and shelter the fuel against access of leachants. The separation techniques

of fuel particles from the compacts of pebble matrices are subject of further studies [Guittonneau, 2008] and experimental investigations within the CARBOWASTE project. Various embedding matrices for confining spent fuel particles are proposed: glass, SiC or ZrO_2. The common objective of all confinement processes is to maintain the integrity of the TRISO coatings and fill the inter-particle space of relatively dense packed particles by an inert matrix which would provide an additional diffusion barrier against groundwater attack.

Large-scale radioactive production experience and favourable disposal properties make vitrification a primary choice for confining the particles. Embedding in sintered glass was shown to be a suitable low-temperature process. Aqueous leaching properties of sintered glass were similar to those of melted glass. No loss of integrity of coatings was observed. The glass product is capable to withstand groundwater attack for many hundreds of thousands of years. Alteration by water will lead to the transformation of the glass into a dense gel as alteration phase, which most likely will maintain the strong confinement properties of fuel particles.

Embedding in SiC is investigated as alternative method. This material would as well be capable to withstand a groundwater attack, for a long time. Conventional methods for SiC production, such as liquid sintering or hot pressing are not applicable when processing spent coated particle fuel. These methods may damage the particles and lead to a release of fission products, either due to the high processing temperatures (1800°C–2000°C) or due to the high pressures applied. Similarly, high temperatures of 1500°C were also needed for embedding fuel particles in ZrO_2 matrices produced from sintering nanoparticles of ZrO_2. In order to maintain integrity of the coated particles, a mild processing method for the production of composite reaction-bonded (RB) SiC material was developed. The samples produced show no fractures (see Figure 6.5) and the new SiC matrix is grown together with the outer pyrocarbon layer of the fuel particle.

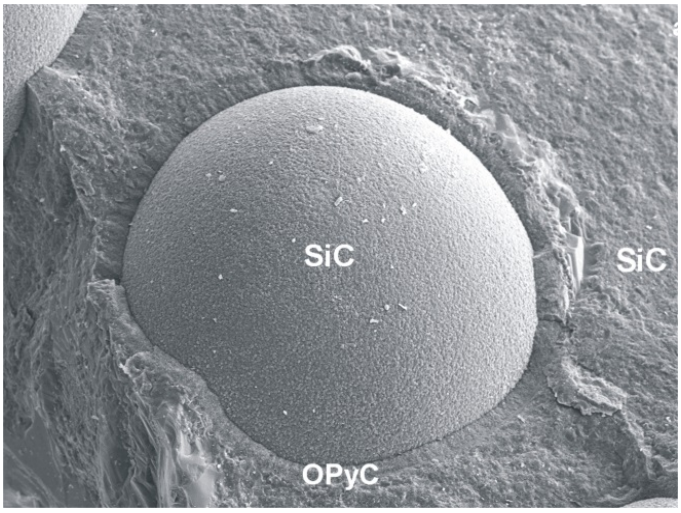

Fig. 6.5: Coated particle embedded in composite RB SiC matrix material and with a cracked-open oPyC layer and visible SiC coating layer

Waste Material Properties

A prerequisite for the assessment of back-end options is a detailed knowledge of the stability of the various fuel materials under disposal and/or interim storage conditions. If, for example, the ceramic coating of the spent fuel can be demonstrated to provide self-containment during long-term disposal in a repository, the wastes from HTR spent fuel will be considerably easier to handle than spent fuel and HLW from other reactor designs, except for the much larger volume and amount of containers per energy unit. For geological disposal, the principal stability criterion is the corrosion and leaching resistance of the various materials in groundwater or brines, since essentially only by reaction of groundwater or brines, radionuclide inventories might be mobilized from a deep repository.

Matrix Material

Water penetration in fuel pebbles with A3 matrix has been studied in the past in detail. The porosity is about 20 vol%, only slightly depending on the neutron dose. Under normal conditions, a sphere takes up about 8 ml. Under pressurized conditions and dissolution of the pore gases in the liquid raises this amount – conservatively estimated – to about 23 ml. This means that a very large fraction of the pore space becomes easily accessible to the groundwater after resaturation, container failure and pressure build-up. After water infiltration, the radionuclides bound within the fuel matrix might become released. The leaching process investigated at 90°C temperature and 13 MPa pressure within the German research program, exhibited two distinct phases: removal of surface activity on a short term (~ weeks), and removal of activity from uranium contamination and defective/failed particles on a longer term. TRISO fuel with the lower uranium contamination of the matrix showed a respectively lower long-term release.

The A3 matrix material is no real graphite due to the fact that it is not graphitized to avoid damages of the embedded coated particles during heat treatment in the manufacture process. The corrosion of A3 material has been investigated in concentrated magnesium and sodium solution as well as in water with and without presence of radiation. Tests were made in an MTR spent fuel cooling pool at 90°C with an average dose ~1 MGy per month. Significant graphite corrosion rates have been observed only in the salt brines showing a linear dose relationship. Maximal dissolution rates at 3.5 MGy were found to correspond to a rate of surface detachment of 1 nm/yr or 1 mm/1 million years.

Coating Material

Analyses have been made on the mechanisms dominating the stability and life time of the particle coating and the dissolution of the spent nuclear fuel kernels in groundwater/brines and associated release of radioactivity. Previous studies have indicated that estimated SiC life times in water may be about 10,000 years. In this study, the chemical resistance of SiC and ZrC, both as powders and in the form of coating layers, was determined under repository conditions such as salt domes, granite-based repositories and clay formations. Depending on leachant type and temperature, either ZrC or SiC is to be preferred in terms of stability under contact with the leachant. Under reducing conditions and low temperatures expected under geological disposal conditions, lower life times are expected for ZrC than for SiC.

In previous studies, potential secondary reaction products of SiC with water were not yet taken into account. The principal solid reaction product is SiO_2 forming layers on SiC, both in air and in water. In [Médout-Marère, 2003] it was shown clearly that SiC oxidizes relatively rapidly to SiO_2 even at room temperature. A detailed study of corrosion products formed in aqueous solution has been performed in [Imai, 2006]. The SiO_2/SiC interface is dense and may probably be considered as diffusion barrier. It is likely that the reactivity at the SiO_2/SiC interface might play a key role. Dissolution of SiC would then be a two-step mechanism: (1) Oxidation of SiC to SiO_2, (2) Dissolution of SiO_2. The dissolution mechanism of SiO_2 is well known. The first step is the dissolution with constant rate. Thereafter, in a second step, equilibrium with the aqueous solution is obtained and this corresponds to a discontinuation of the dissolution reaction. It is very important to study the potential equilibrium between the SiO_2 surface on SiC and the aqueous solution. If this mechanism can be validated experimentally, life time's much longer than 10,000 years may be achieved for the SiC coating. The interaction of SiC with water has also been assessed by geochemical modeling and associated thermodynamic databases. The temperature range studied was between 25°C and 200°C. The calculations have confirmed that SiO_2 is the principal solid reaction product. Under reducing aqueous redox conditions, organic reaction products are expected to be formed as well, while under oxidizing conditions CO_2 is calculated to be the principal carbon containing reaction product. The formation of SiO_2 as principal solid reaction product is confirmed by literature data. The SiO_2 seems to form a tight dense layer at the surface of SiC, which may play the role of a protective layer. If confirmed, this may imply a diffusion-controlled dissolution of SiC. The implications on the extrapolated life time of the SiC coating are very important: since diffusion rates will decrease typically (not always) with the square root of

time, life times of the SiC layer would extend well beyond millions of years, much longer than considered in the present project. This means that the use of a linear rate law is conservative, but more research would be necessary if benefit is to be taken of the diffusion-based rate law.

Kernel Material

The fuel kernel is the source of any potential radionuclides release from a spent fuel waste package in case of groundwater access, during final disposal. It is well known that uranium dioxide stability in groundwater is strongly degraded in presence of oxidants. In deep repositories as planned in Europe, conditions are favourable for UO_2 stability, since only few oxidants are present and reducing conditions prevail. Nevertheless, both oxidants and reductants are produced by the decomposition of water by ionizing radiation. This process is called radiolysis. Three experiments were conducted with uranium dioxide, Th/U and uranium carbide kernels.

(1) The dissolution of TRISO UO_2 spheres under α irradiation was studied using the beam of a cyclotron (CERI/CNRS, Orléans/France). 30 UO_2 kernels were put in 30 ml of aqueous solution ([NaCl] = 10^{-3} mol/l and [$NaHCO_3$] = $4 \cdot 10^{-2}$ mol/l). In two irradiation experiments, the beam hit the solution but not the particles. In a last experiment, the particles were positioned in the alpha beam (7 MeV) with a dose rate of 100 Gy/min for two hours. Ar and Ar/H_2 gas were used to remove traces of oxygen and to study H_2 gas effect on UO_2 dissolution. In fact, H_2 gas will also be produced for thousands of years by container corrosion, under the expected repository conditions.

Under Ar gas, U concentration increases from 0 to 1.86 10^{-8} mol/l) at the same time as H_2O_2 concentration decreased from $1.01 \cdot 10^{-5}$ in absence of UO_2 to $0.86 \cdot 10^{-5}$ mol/l in presence of UO_2 due to UO_2 oxidation and U(VI) dissolution. In presence of H_2, H_2O_2 production increased by about a factor of 2 leading as well to an about two fold larger extent of UO_2 dissolution. Thus, in contrast to the frequently observed diminishing effect of H_2 gas on UO_2 dissolution rates, there is a non-negligible enhancing influence of H_2 on UO_2 dissolution, under the specific conditions.

Both in presence and absence of H_2, less than 1% of the consumed H_2O_2 is used for UO_2 dissolution, the rest probably for UO_2 oxidation. In contrast, when the UO_2 kernels are under the direct influence of an alpha beam, the effect on UO_2 dissolution is very important while H_2O_2 consumption is the same (about $7 \cdot 10^{-6}$ mol/l). Here, about 50% of consumed H_2O_2 is used for dissolution. Under the direct alpha beam, local H_2O_2 concentrations at the UO_2 surface are much higher than measured by the Ghormley method. Scanning Electron Microscopy (SEM) results show that UO_2 dissolution occurs locally, in particular, in grain boundaries.

(2) The dynamic leaching of an irradiated HTR fuel kernel (U/Th MOX) has been conducted for four years including H_2 saturated conditions. The irradiated kernel is fixed in a stainless steel flow-through reactor equipped with a filter. Synthetic groundwater kept at reducing potential passed through at flow rates between 1 and 4 ml/h.

Cesium was analyzed by gamma spectrometer, strontium and plutonium by liquid scintillation, and uranium by ICP-MS. The main radioactivity in this material is due to the fission product nuclides cesium-137 and strontium-90. The analyses results of these two isotopes have been normalized to their inventory in the fuel to be compared with those of plutonium 238.

For ^{137}Cs and ^{90}Sr, there is a high instantaneous release at very short time corresponding to the labile fraction from the fuel surface and the accessible grain boundaries. The nature of the present gases seems to play a role: in Ar/H_2 atmosphere corrosion rates are about 2 times slower than in Ar, in contrary to what was observed for irradiation in the alpha beam. Results show relatively weak and congruent leaching from Sr and Pu indicating fuel matrix dissolution rates of about 0.23 mg m^{-2} d^{-1}.

(3) Two different types of uranium carbide kernels used for the leaching experiments were formerly manufactured by the company HOBEG. About 20 mg of unirradiated isolated

kernels were filled into a stainless steel tube and immersed with 7 ml aqueous phase (brine 2 or Mont Terry water). The whole filling procedure was performed under an argon atmosphere. Totally 60 irradiation tubes have been prepared by this procedure. The gamma irradiation was performed at different locations in the spent fuel cooling pool of the FRJ-2 research reactor.

Leaching experiments with irradiated kernels were performed by placing one particle of each of the two different types of coated particle inside a glass vial. All of the filled glass vials were individually inserted into a lead container and then placed inside a drying cabinet with a temperature of 90°C. This experiment was performed over a total period of four weeks. The liquid samples that were removed from the vials were analyzed by gamma spectroscopy.

The scanning electron microscope examinations performed on the solid residues that remained on completion of the leaching tests conducted with salt brine with unirradiated kernels and without radiolysis show that there are no longer any intact kernels following the completion of these tests (see Figure 6.6). This indicates that the aggressive salt brine destroyed the kernel structure.

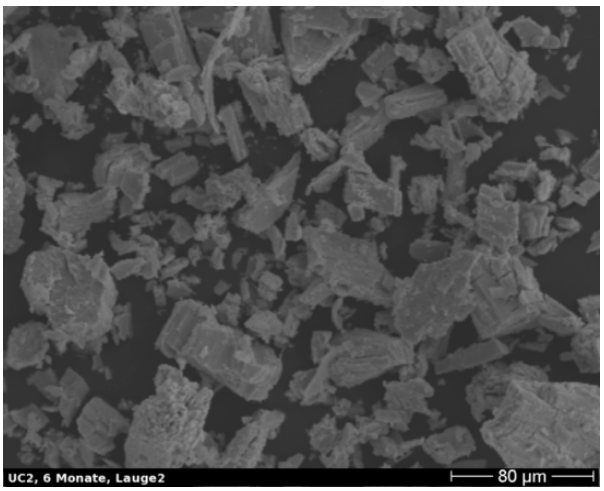

Fig. 6.6: UC_2 kernel after 6 months in salt brine

Fig. 6.7: UC_2 kernel after 6 months in Mont Terry Water

On completion of the leaching tests conducted with Mont Terry Water, there are still intact kernels (see Figure 6.7). Although these kernel surfaces exhibit damage caused by the leaching process, they still retained their original shape. As leaching time increased, it was possible to observe at higher magnifications that cracks were

progressively forming on the surface and also that a new phase arose on these kernel surfaces.

In summary, the stability of the UC_2 fuel in water is drastically reduced relative to UOX fuel. Therefore, a reduced stability of UCO must be assumed and needs to be investigated, in more detail.

6.3 Modeling

A simplified geochemical model was developed combining geochemical/transport and radiolytic assessment of reactions relevant to pore water chemistry and spent fuel kernel dissolution. A radiolytic graphite dissolution model with H_2O_2 as principal reactant has been established. The model has partly been validated against published experimental data, but a more extensive comparative study for a variety of environmental conditions (presence or absence of gas phase, pH, varying surface to volume ratio etc.) would still be necessary.

The model predicts first H_2O_2 concentrations as a function of radiolytic decomposition of water and is able to predict the effect of radiolytic H_2O_2 concentrations on dissolution rates of graphite pore wall surfaces. It allows calculating the effect of graphite dissolution on the pore water concentration of dissolved organic and inorganic carbon species.

The graphite corrosion model confirms graphite life times of about 10 million years. Graphite corrosion rates even at low dose rates of 0.1 Gy/h are predicted to be as high as $1.3 \cdot 10^{-7}$/yr. The results show that graphite is essentially stable, but porous. The current model seems even to overestimate graphite dissolution rates under gamma irradiation conditions.

A model for the spatial evolution of alpha, beta and gamma dose rates in HTR fuel pebbles has been set up using the Bethe Bloch equation and dose profiles have been coupled to a pore water transport model for radiolytic species using the TraRaMo code. Alpha dose profiles extend to about 40 µm into the PyC buffer layer of the coated particles. Beta and gamma dose profiles extend much longer. The beta and gamma dose rates between the different fuel particles are governed by all adjacent fuel particles in all 3 dimensions. Shielding effects by other fuel particles were ignored. Radionuclide inventories and gamma emission as a function of time were calculated with the code Origen-ARP 2.00 (2002). Two cases were distinguished: early container failure and container failure after 1000 yr.

The initial dose rate in a fuel pebble, after an unexpectedly early container failure, is governed by the beta radiation of ^{90}Y and the gamma dose rate. It has been shown that the dose rate decreases only weakly between the fuel particles. This is due to the high energy of the beta radiation of this nuclide. The dose rate at the fuel surface is more than 10 times higher than in the case of a single particle. After about 300 yrs, due to the decay of $^{90}Sr/^{90}Y$, this dose rate will be about 1000 times lower and it will be entirely negligible after 500 yrs.

The dose after expected container failure after 1000 yr but before SiC failure is governed by beta decay of ^{126m}Sb. The specific activity in the fuel is 38 kBq/g. The dose rate stays constant for up to 100,000 yrs and decreases thereafter. The dose rate decreases more strongly with distance from the fuel kernel surface than in case of ^{90}Y since the energy is lower.

The overall evolution of dose rates with disposal time is given for the 3 cases in Figure 6.8. In no case, gamma radiation becomes dominant. The dose rates (beta) are by far highest in the first unexpected case of initial container failure. They decrease by about 5 orders of magnitude for case 2. An increase of dose rates is only expected for case 3 (there was only mention of 2 cases before) once the SiC coating has failed. After SiC failure, alpha irradiation is the principal dose contributor at the fuel kernel surface.

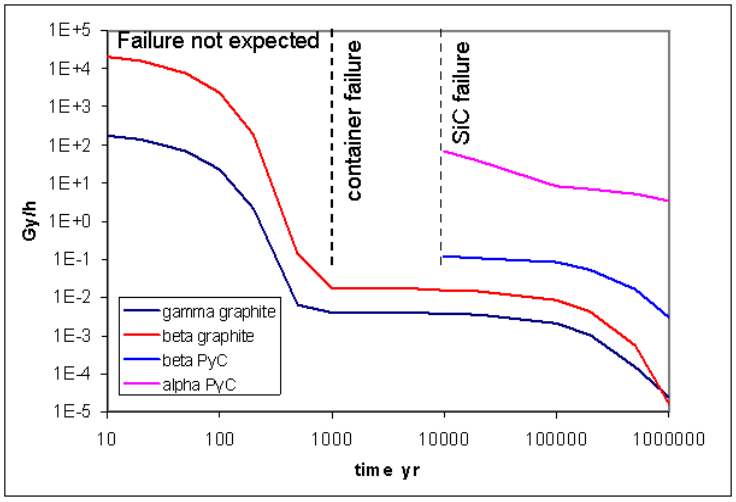

Fig. 6.8: Evolution of alpha, beta and gamma dose rates in a fuel pebble (graphite and IPyC) as a function of time

For the geochemical modeling of graphite pore water composition, two calculation cases are distinguished: Case (1) is based on the conservative assumption of an early accidental container failure. Here, gamma dose rates can be up to 3000 Gy/h. High oxalate concentrations and low pH values are expected in the pore water with concentration gradients from the centre to the periphery of the pebble. Steady state profiles will be achieved after 60 years. These high organic acid concentrations and low pH values will lead to a situation where radionuclide mobility in graphite pore water is high and a significant retention cannot be assumed within the fuel pebble. The principal barrier would then be the mass transfer resistance in the fuel pebble and the stability of the SiC coating. Case (2) considers the expected evolution scenario where container failure will only occur after significant corrosion after about 10,000 years. In this case, alpha radiolysis is dominant. Pore water solution composition in the graphite matrix will be similar to groundwater composition. In contrast to case (1), significant radionuclide retention can be expected in the fuel pebble, even if dissolution of fuel kernels may occur after very long times.

As a modeling platform for spent fuel dissolution, the code TraRaMo is used together with the code MAKSIMA for radiolytic reactions. Rate equations were already calibrated in the NF-PRO project of the European Commission using experimental data for a wide range of experimental conditions. Most calibration data stem from unirradiated UO_2 while application cases include real spent fuel.

The coupled radiolysis/transport code TraRaMo has been developed by Lundström [2003] in collaboration with SUBATECH, partly in the context of the European SFS project. Transport of radiolytic and other dissolved species in aqueous solution is considered in a one-dimensional way by a finite-difference representation of an advection-diffusion equation, dividing the transport path into a selected number of equally sized cells. Radiolysis reaction schemes are calculated for each cell as a function of dose and time step using the well-known MAKSIMA code as module in TraRaMo. The database of MAKSIMA needs to be adapted to the problem studied: different databases are used for high saline and low saline solutions. In the case of application of fuel dissolution in a graphite environment, additionally to the “simple” spent fuel water system, also the competition of graphite for the generation of radiogenic hydrogen peroxide must be considered.

Some preliminary modelling results for fuel dissolution rates are obtained for a reference surface dose rate of 8 Gy/h for alpha radiation. This corresponds to the long term dose indicated in figure 6.8 due to SiC failure after about 100,000 years. Dose contributions for beta and gamma radiation were found to be insignificant. External carbonate concentrations were set at 0.01 M simulating typical carbonate controls by groundwater. Only little variation of carbonate concentrations in the fuel pebble was predicted. The corrosion rates of the fuel kernels are calculated to be lower than 0.001 $mg \cdot m^{-2} d^{-1}$ due to the consumption of H_2O_2 by the graphite.

The analytical results of the performance of the various near field components have been used in a total system performance assessment. The proposed very stable confinement matrices complemented with the resistance provided by the different layers of the intact TRISO fuel particles result in no release of radionuclides into the biosphere during the first 200,000 years. The calculated dose rates are a few orders of magnitude lower than the dose constraints that are in use in European countries for geological repositories.

6.4 Conclusions on Direct Disposal of Spent V/HTR Fuel

The principal ways of how to assess the performance of treated and non-treated waste packages are now better understood after having developed models for pore water evolution, coating corrosion, and fuel dissolution both for early container failure and for the expected container failure after 1000 years. It is now possible to couple the different models into an overall view.

For direct disposal of spent fuel pebbles, a corrosion model has been developed to describe fuel kernel dissolution within the fuel pebble considering the following scenario: diffusion of water into the pebble within a few years, failure of the SiC coating within about 10,000 years, dissolution of kernels after about 10^6 years under reducing and 10^4 years under oxidizing conditions. Solubility limits in the fuel pebble pore space are considered as well. The results show that the instantaneous release is of minor importance for HTR fuel. This is due to the stability of the SiC coating.

Chemical and mass transfer modelling of the reaction network governing radionuclide release from spent V/HTR fuel under geological disposal conditions show for direct disposal of pebbles (or prismatic blocks) that radiolysis of pore water in the graphite matrix is an important process influencing pore water chemistry which may control both the dissolution rates of the graphite and the pore water chemistry. High carbonate and oxalate concentrations in pore water produced by gamma radiolysis may lead to high radionuclide mobility in graphite pore water. However, this is only valid for the case of the early container failure scenario. Radiolysis is the main source for graphite oxidation/dissolution.

Significant elements for a prediction of the radionuclide release properties from embedding matrices have been gathered: Basic matrix corrosion rates are available for glass, SiC and ZrO_2. This will provide a lower bound for the rates, by which embedded fuel particles may come in contact with diffusion groundwater. It is expected that the corrosion rates of the embedding matrix will be in the order of 10^{-5}/yr for glass or lower for other matrices. Hence, the rate by which water comes on average in contact with the fuel particles will be slower than the rate by which the SiC coating may disintegrate (10,000 yrs). Hence, the SiC coating will probably provide no additional barrier function when comparing the relative importance with that of the matrix. Important reduction of release rates is also expected from the fuel particles. The above described experimental results show that hydrogen from container corrosion strongly reduces corrosion rates of irradiated fuel. Moreover, data from light water reactors show that there is a threshold for specific alpha activity in the fuel, below which increase of fuel dissolution rates by radiolysis do not need to be considered. Considering that the embedding matrix will strongly delay groundwater contact with the fuel particles, radiolysis is most likely not a factor, which needs to be considered with regards to fuel dissolution rates. Indeed, the above mentioned model prediction of rates below 0.001 $mg \cdot m^{-2} d^{-1}$ corresponding to a propagation of the corrosion depth into the UO_2 matrix of the fuel kernel of 1 µm in 300,000 years are consistent with UO_2 dissolution rates under reducing conditions, in absence of radiolysis. The stability of the UC_2 (or UCO) fuel in water or brine is drastically reduced relative to UO_2 fuel.

Further detailed investigations on the corrosion mechanisms of irradiated kernels as well as of pyrocarbon and SiC layers are still necessary. Geological forces due to the convergence of the disposal drafts and subsequent mechanical impacts on the fuel elements also need to be taken into account.

7 Sustainability Evaluation of the V/HTR System

Sustainability, as e.g. defined in the Brundtland report and the Rio Declarations, is linked with ecological, social, economical, cultural and institutional development aspects of the increasing world's societies and their standards of living. Viewing these different sections of sustainable development from a normative perspective, the term 'dimensions' or 'pillars' has been widely established for the assessment of different energy supply systems.

An isolated 'nuclear interpretation' of 'sustainability' should be avoided to enhance the general acceptance of the results. Appendix 1 shows the general indicators being valid for all energy supply systems and those which are additionally specific to nuclear energy. Although this paper concentrates on nuclear fuel cycle issues, it must be kept in mind that an evaluation of sustainability has to integrate all aspects. Thus, a Multi-Criteria Decision Analysis (MCDA) would finally be necessary to elaborate a balanced result on a specific system, such as the V/HTR. In this chapter, only some of the most important sustainability indicators related to the back-end of V/HTR are qualitatively highlighted.

Sustainability of nuclear energy has to be regarded in a larger context and not for a specific nuclear system alone. Different system features have to be combined in a mix of diverse energy carriers and technical concepts to respond to different economic, ecological and social requirements and to the different application fields (e.g. Heat and Power). Finally, these boundary conditions determine the choice and the combinations of systems within an energy supply network. In principle, sustainability evaluations of selected technologies should start from the market needs, the technological availability/maturity and the political environment to define compatible energy supply systems which may be different from country to country (e.g. emphasis on proliferation resistance, closed cycles with reprocessing, open cycles etc.). The 'trilemma' of technology choices is illustrated in Figure 7.1 and shows the interdependence with economics and politics which include environmental and social aspects.

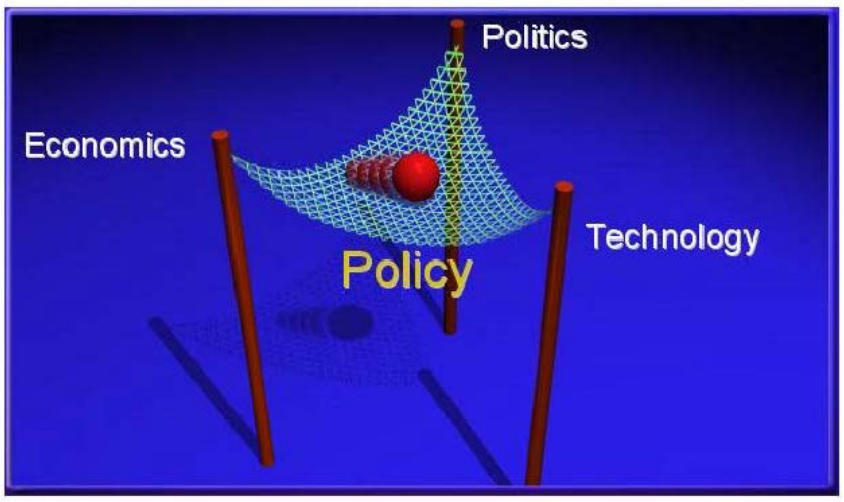

Figure 7.1: Trilemma of technology choices

Nuclear energy systems have not only to coexist with other renewable and non-renewable energy carriers but also with different nuclear systems. Symbiosis of LWR and Fast Reactor, e.g., has been discussed in the past to address fuel resources and waste issues in a complementary way to achieve a better sustainability ranking together. Such combinations may also be essential by including the specific features of V/HTR such as deep-burn capabilities, e.g. in combination with LWR. Finally, it needs to be kept in mind that the competing conventional, renewable and nuclear energy supply systems maintain their own innovation efforts and that technologies are not frozen.

The provision of adequate and reliable energy services at an affordable cost, in a secure and environmentally benign manner and in conformity with social and economic development needs, is

an essential element of sustainable development. Energy is vital for eradicating poverty, improving human welfare and raising living standards. However, most current patterns of energy supply and use are considered unsustainable [Vera 2005].

This can be illustrated by the fact that about two thirds of the primary energy is lost e.g. for electricity production (see Figure 7.2 [IEA 2008]). Thus, better use of primary energy by increases in conversion efficiencies and the use of waste heat are essential targets for a more sustainable use of energy.

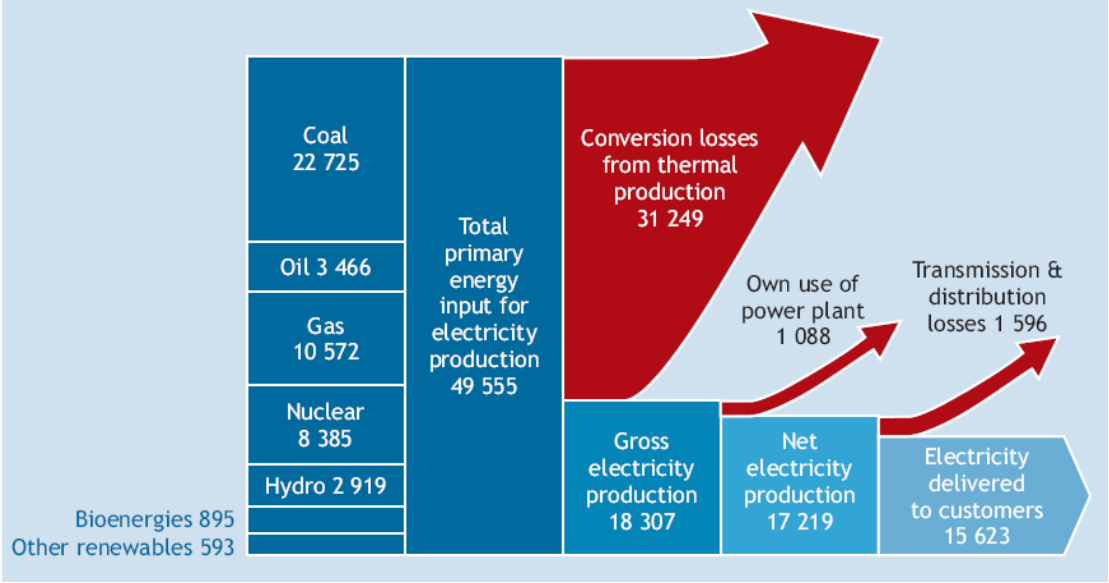

Figure 7.2: Energy flows in the global electricity system [IEA 2008]

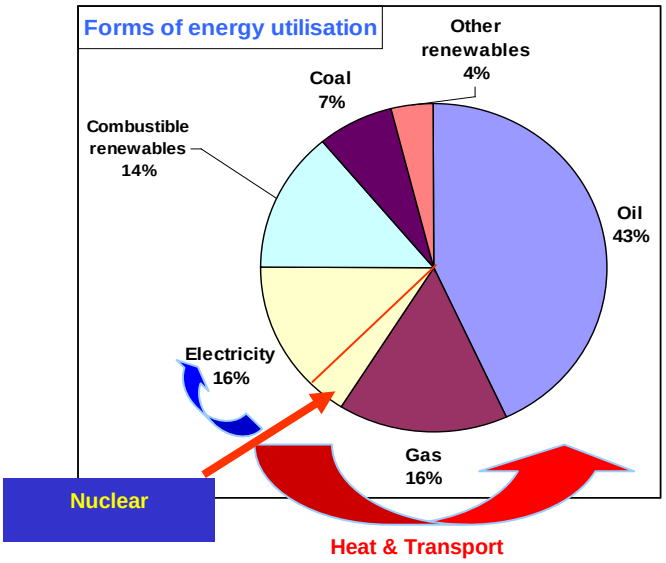

Figure 7.3: World energy utilization

Today, nuclear energy is mainly deployed for dedicated electricity generation which only represents about 16% of total energy consumption, worldwide. Nuclear energy is providing only about 15% of this share. However, most of energy needs are directed to heat and transport. All these energy requirements need to be fulfilled in a sustainable manner and nuclear energy may contribute by not only penetrating a bit further into the electricity market but also by providing district heat, process heat and hydrogen for different purposes (fuel, fertilizers, chemicals etc.). Such applications could open new markets for nuclear energy, especially for V/HTR.

Sustainable Design Principles

Only a selected number of energy-related priority areas from the Indicators for Sustainable Energy Development (ISED) have been chosen to assess the ranking of V/HTR in this context.
Efficiency of energy conversion (ECO 3)

- High thermal efficiencies having an impact on
  - *Better fuel utilization, less uranium demands (ECO 4/5)*
  - *Less specific waste generation (ENV 7-10)*
- Diversification of fuel mix (ECO 11/12)
  - *Substitution of limited fossil energy resources (ECO 15 / ENV 1)*
  - *Combined Heat, Power and Hydrogen Production (ECO 16)*
- Waste generation (ENV 8-10)
  - *Minimisation & safe management of waste streams*
  - *Symbiotical fuel cycles*
- Safety (SOC 4)
  - *Exclusion of significant radiological impacts*
- Strategic fuel stocks (ECO 16)
  - *Uranium reserves*
  - *Use of thorium*

High Thermal Efficiencies

The economics of nuclear power is challenged by an ongoing improvement in the efficiencies of conventional power plants. The following figure illustrates the development within the last century. It can be seen that efficiencies of LWR (~35%) were the state-of-the-art in the 1940s when ending the era of the steam engines. Efficiencies above 50% have now been reached by combined gas and steam turbine cycles and by super- or even ultra-supercritical steam conditions. This increase in efficiencies compensated for the cost escalations of fossil energy to a great extent.

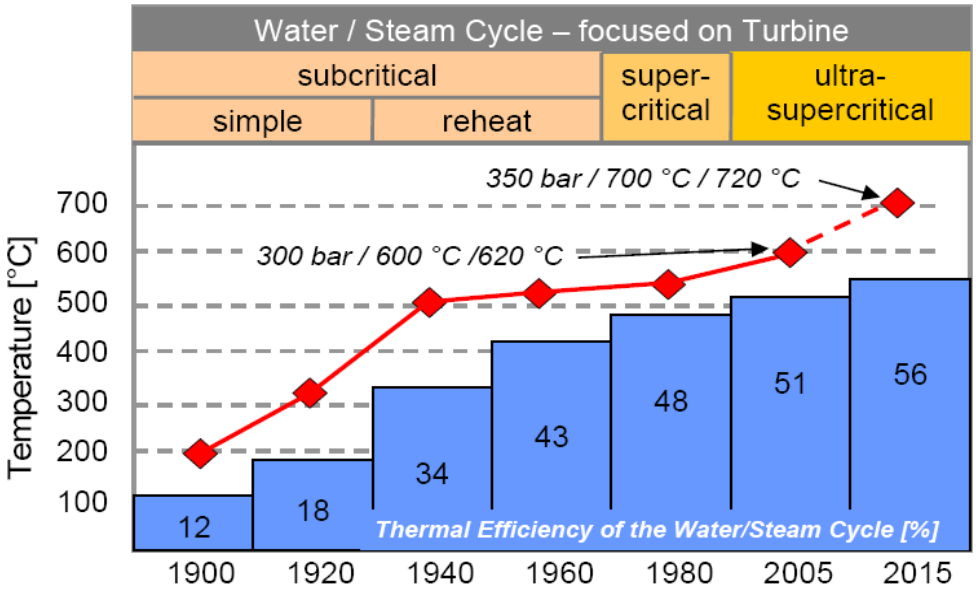

Figure 7.4: Evolution of efficiencies in conventional power plants

The low efficiency of current nuclear plants results in high uranium consumption and large amounts of radioactive waste. Therefore, an elementary design principle for all kinds of future nuclear systems must be an enhanced thermal efficiency, which can only be achieved by higher temperatures.

Higher temperature allows power conversion to electricity with higher efficiency. Of the many possible power conversion cycles existing, Rankine cycles today achieve individually the highest efficiency, they can be dimensioned reliably with minimum development risk, with materials and components available off-the-shelf.

Typical PWR designs produce saturated steam at 7 MPa and approx. 300°C and achieve an electric efficiency of 32%. Several nuclear power plants (e.g. in Finland and Switzerland) produce low-temperature steam in co-generation mode for district heating or paper production.

Modern coal-fired power plants produce supercritical steam at up to 25 MPa and approx. 550°C and achieve an electric efficiency in excess of 50%.

Combined cycle power plants (CCPP) combine an open gas turbine cycle with a closed steam cycle yielding overall efficiencies of 60%.

V/HTRs allow to follow the trends towards higher efficiencies within the conventional power conversion technologies and can be operated with direct or indirect Brayton and Rankine cycles or a combination of both. V/HTR can also apply supercritical steam or CO_2 cycles for further enhancement of the performance and of the waste generation.

Combined Heat, Power and Hydrogen Generation

Combined Heat & Power (CHP) is spreading in the conventional energy sector allowing for much better fuel utilization whereas nuclear energy is mainly operated without making any use of the waste heat. Thus, a second design principle towards improved sustainability should be to optimise nuclear plants for CHP applications, i.e. adaptation of the power size to the heat demands which are mainly in the range of several hundreds up to few thousands of thermal MW. By CHP, the uranium utilization can be more than doubled and specific waste production accordingly be reduced. Nuclear CHP can directly substitute fossil energy and thus reduce CO_2 emissions. However, siting conditions and safety requirements may be different compared to remote dedicated electricity generation.

A comparison of dedicated power generation in comparison to cogeneration of high temperature process heat and electricity with a bottoming Rankine cycle is illustrated in the next figure. It shows that efficiencies of about 70% can be achieved, which is more than double the efficiencies of LWRs. This comparison also shows the high exergetic value of the high-temperature heat. The use of the residual heat in the low temperature range can further improve the efficiency, in both cases.

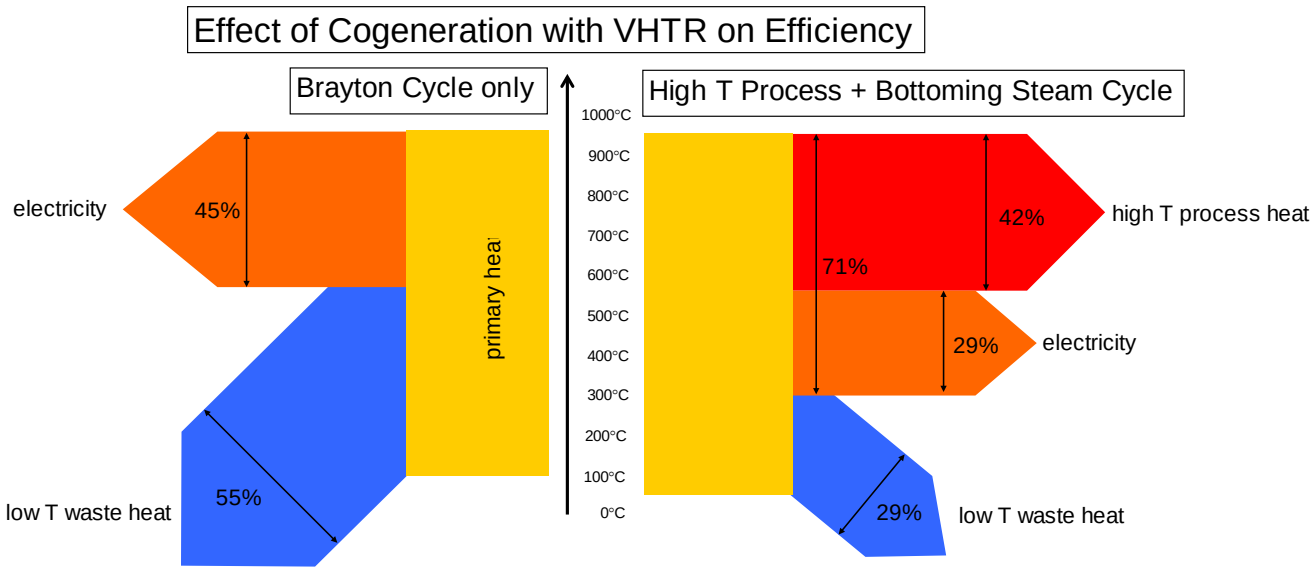

Figure 7.5: Comparison of dedicated electricity generation against CHP

Reduced Waste generation

The coated particle fuel concept differentiates V/HTRs from all other reactor systems. This fuel allows very high burnup, which can, e.g., also be used to burn long-lived waste (deep-burn).

A comparison with LWR shows only a slightly lower consumption of natural uranium but the production of waste is considerably lower e.g. a factor of 3 with regards to the generation of Transuranium (TRU) elements. However, the different uranium enrichment and thermal efficiencies of V/HTR vs. LWR needs to be taken into account when comparing the waste streams.

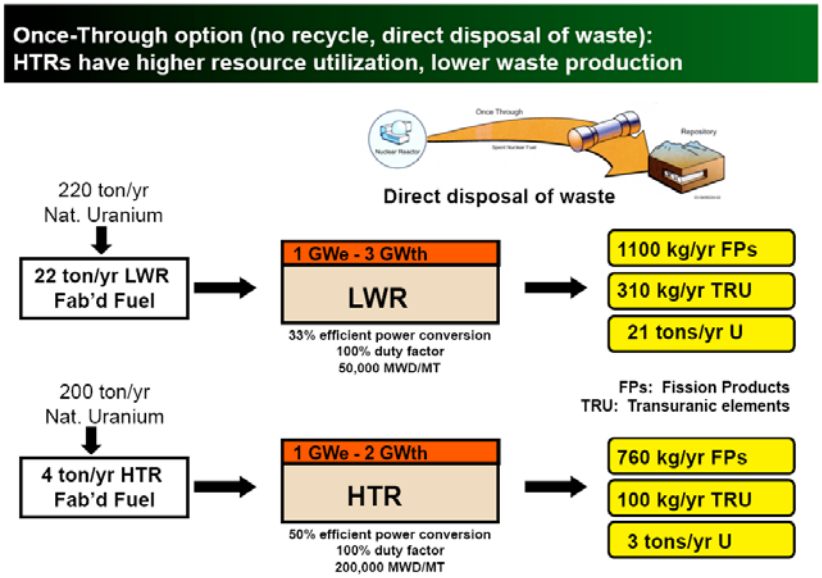

Figure 7.6: Comparison of waste generation of LWR and HTR in the Once-Through mode

Further waste reduction can be achieved if HTR are feed with reprocessed spent LWR fuel in the deep burn mode. Contrary to LWR, HTR can be safely operated with a pure Pu core.

If only HTR are applied with one intermediate reprocessing step, the amount of TRU can be reduced to one tenth of the original TRU waste of an LWR.

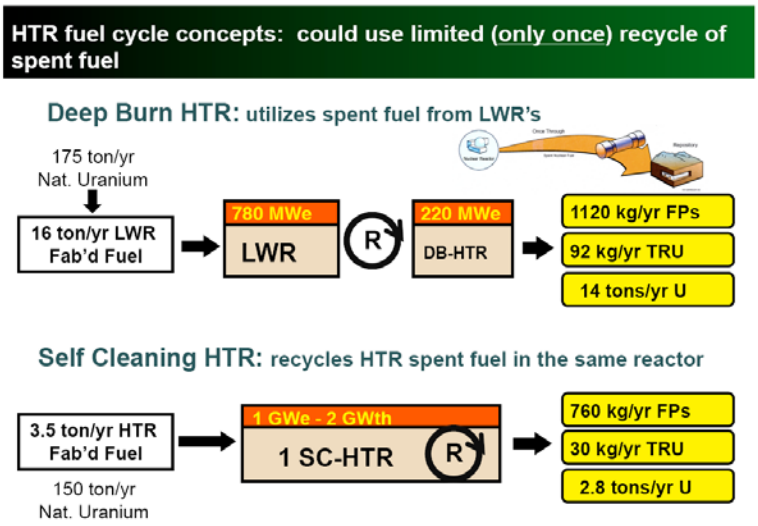

Figure 7.7: Waste generation of LWR and HTR in the Recycle mode

Another important difference of coated particle fuel as compared to other spent fuel is the fact that also long-lived fission products are retained in the coated particles for long disposal periods. The next figure shows that radioactive releases of directly disposed spent LWR fuel are strongly dominated by the volatile long lived fission and activation products. Spent HTR fuel does not show such instantaneous release fractions. However, the potential release of long-lived activation products like ^{14}C and ^{36}Cl from the graphite fuel matrix needs to be taken into account for V/HTRs.

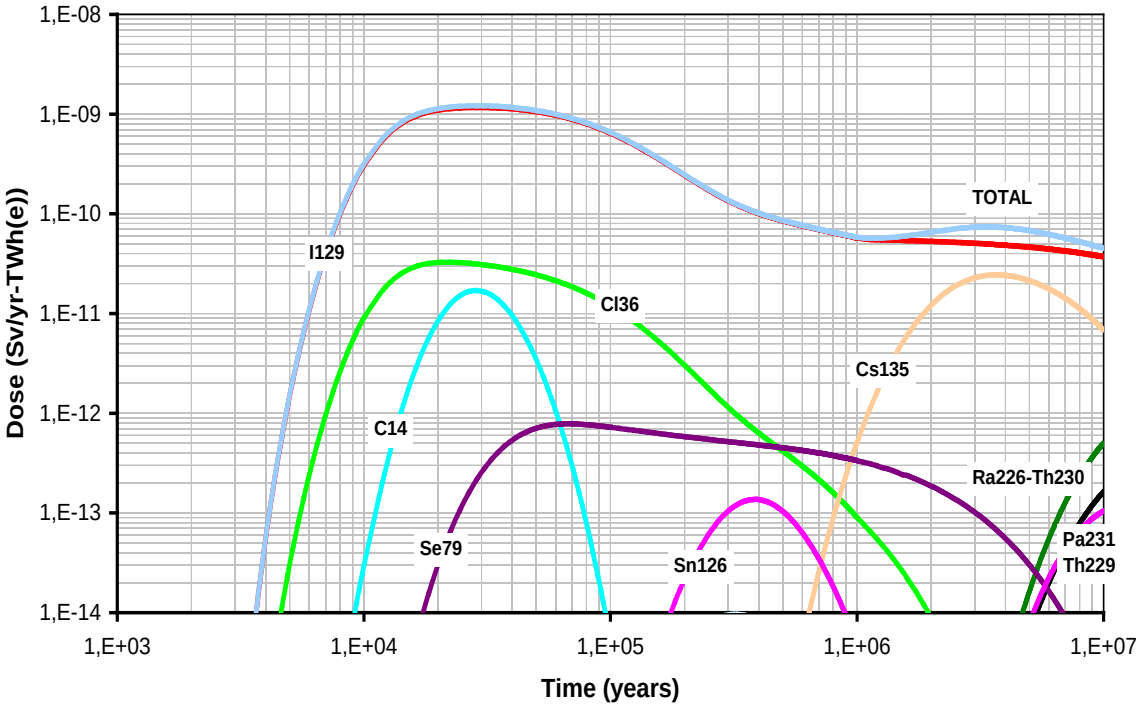

Figure 7.8: Doses related to LWR spent fuel disposed in granite

The high burnup potential of V/HTR fuel reduces the number of reprocessing steps in the case of closed V/HTR fuel cycles. This reduces the effluence of volatile radionuclides during reprocessing significantly.

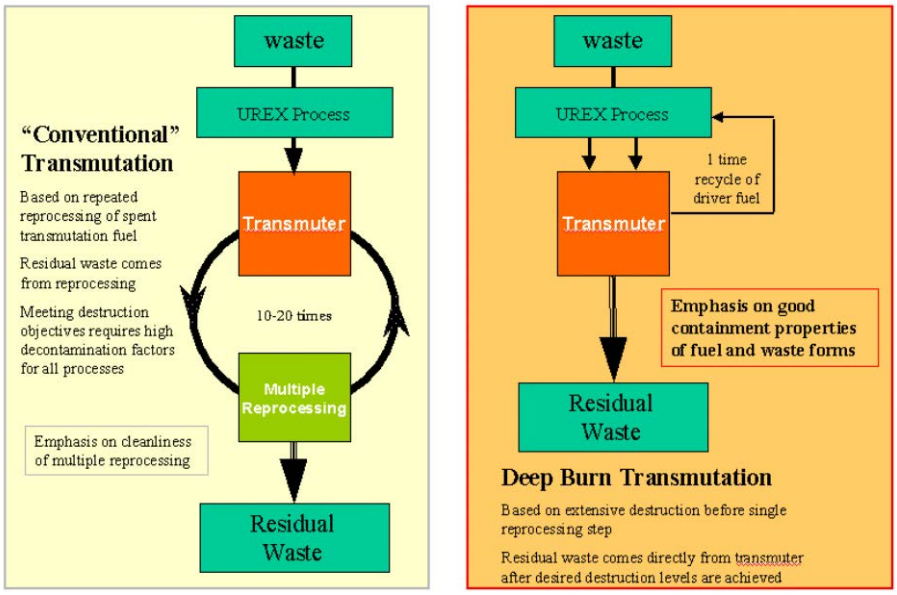

Figure 7.9: Approaches on transmutation

In consequence, it can be stated that V/HTRs in an open cycle and especially in a closed (self-cleaning) cycle can significantly reduce the TRU discharges and contribute to the transmutation of TRU by deep-burn.

It is obvious that closed fuel cycles are superior in terms of fuel utilization. Finally, it is an economic decision whether the cost for recovering residual fissile material compensates for (multiple) reprocessing, re-fabrication and potential benefits in the waste management routes.

Another route to waste minimization is the use of thorium. This cycle significantly reduces the generation of TRU and exhibits much smaller radiotoxicity as shown in Figure 7.10. After decay of most fission products, the thorium cycle achieves the same radiotoxicity level as other transmutation processes after multiple recycling. In addition, the thorium matrix is much more leach resistant and stable due to the chemical monovalence of thorium oxide.

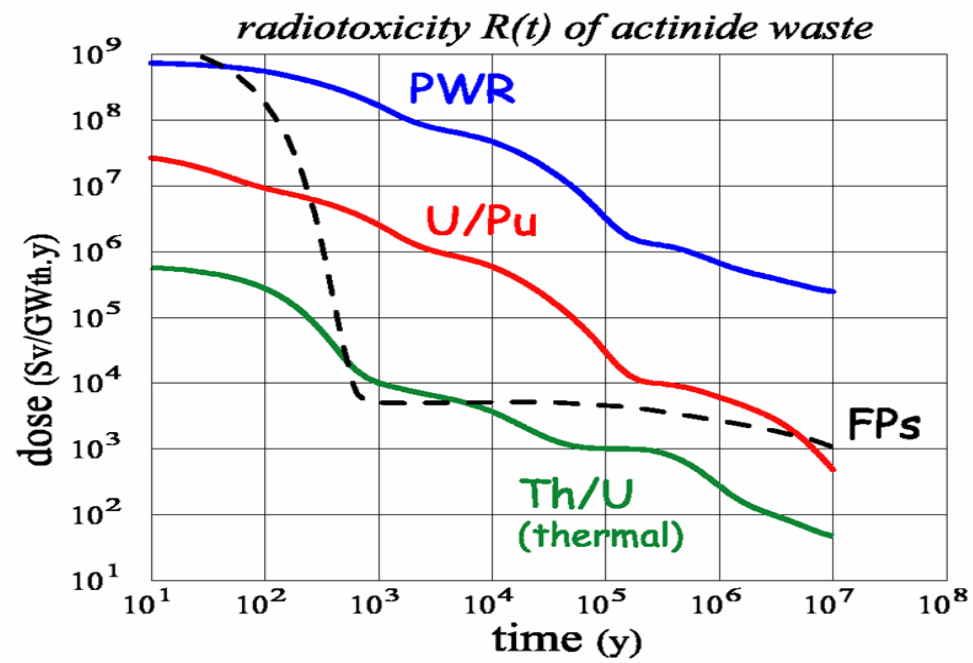

Figure 7.10: Radiotoxicity of the thorium cycle [Source: CNRS]

A further fundamental principle is concerned with '<u>neutron economy</u>'. Present reactors (except CANDU) need to compensate excess reactivity of the fresh fuel by neutron poisons and tolerate parasitic neutron capture, e.g., in the light water coolant. This also leads to additional uranium consumption, waste generation, and reduced breeding capabilities. Therefore, neutron economy needs to be improved for future systems by applying low absorbing coolants and core structures.

Safety Issues

An important design principle is the <u>exclusion of large radioactive emissions</u> even in hypothetical accidents such as reactivity excursions and loss-of-coolant accidents. Dependence on active systems needs to be reduced to cope with station black-out conditions. Loss-of-coolant accidents can mainly be avoided by integrated modular plant designs. Alternatively or complementary, inherent or passive decay heat removal systems can avoid core damage and release of fission products from the primary circuit. These measures should also allow covering nuclear risks by commercial insurance conditions and contributing to enhanced social acceptability.

Strategic Nuclear Fuel Stocks

In recent years, the media have recurrently published the view that availability of uranium ore would set tight limits to the expansion of thermal nuclear reactors, deducing that it would make no sense investing in this technology any longer. Depending on nuclear energy forecasts, allowable periods between 35 and 120 years are quoted, corresponding to only 1 or 2 reactor lifetimes. The same argument of uranium shortage is used by proponents of fast breeder reactor systems who expect these reactors to stretch uranium resources by a factor 50. While this advantage cannot be denied, it does not prohibit the parallel expansion of thermal reactors.

The OECD/NEA and IAEA “Red Book” of 2011 indicates Reasonably Assured Resources (RAR) plus Inferred Resources (IR) available at 5 327 200 tU (US$ <130/kgU, or 50/lbU3O8); -1.4% from 2009 and 7 096 600 tU (US$ <260/kgU, or 100/lbU3O8); +12.5% from 2009. Undiscovered Resources (Prognosticated and Speculative) are estimated to be in the range of 10 429 100 tU; <1% up from 2009 (not reported by all, e.g. Australia, Namibia…).

With regard to the nuclear fuel resources, between 1994 and 2010, world uranium production was increasing from ~31 kt to ~54 kt/a mainly due to higher production volumes in Kazakhstan. Compared to the 2005 production level, however, the 2010 production level was 40% lower. The total identified uranium resource base is estimated at around 7.1 Mt U, with an increase of over 12% since 2009. The IAEA 2011 Red Book on “Uranium: Resources, Production and Demand” concludes that the “identified resources are sufficient for over 100 years of supply for the global nuclear power fleet”, at 2010 global reactor requirements. Of the 31 countries with nuclear power plants, there are three which are are self-sufficient in their uranium needs: Canada, Russia, and South Africa .

A significant increase in extraction costs has been observed, which has caused a reduction in the lower cost range. Still, the uranium production supplied at cost of < 130 $/kg may perhaps meet the fuel demand in the IAEA low-case scenario for the next 10 to 20 years.

During the two decades before 2005, very little uranium exploration has taken place. The recent increase in exploration effort raised known uranium resources by 15% (17% in the cost category to $80/kgU). In analogy with other ores, a doubling of price from present levels could be expected to create about a tenfold increase in measured resources. Knowing that fuel contributes only a few percent to the kWhe generation cost, such a price doubling does not seem prohibitive, even higher prices may be acceptable one day. This would lead to more than 800 years of uranium availability at current consumption in today’s thermal reactors, not even considering the beneficial effects of higher efficiencies, e.g. through co-generation (factor 2).

Thorium is another fuel type which can be employed in thermal reactors such as CANDU or V/HTR. Thorium is reported to be about three times as abundant in the earth's crust as uranium. The 2005 "Red Book" indicates a figure of 4.5 Mt of reserves, excluding data from much of the world. The fuel cycle needs to be started using a fissile material such as U-235 or Pu-239. The thorium fuel cycle has several attractive features although specific technology requirements make it somewhat more expensive than the uranium fuel cycle.

More recent figures [WNA] estimate 6.2 million tonnes of thorium, of which reasonably assured resources are 0.8 to 1.5 million tonnes. Some of the figures are based on assumptions and surrogate data for mineral sands (monazite x assumed Th content) and not on direct geological data in the same way as most mineral resources.

These resource predictions indicate that fuel availability is no serious argument which would curtail the expansion of thermal nuclear reactors such as the V/HTR. The real limit of fuel resources is probably given less by economic than by energetic considerations: the fuel cycle must require significantly less energy for a given quantity of fuel than this same fuel can produce in a reactor. If in addition, waste streams can be reduced and the remaining waste be disposed of safely, safe nuclear fission reactors do have the potential to displace large amounts of fossil fuel in the next centuries.

Sustainability of V/HTR

For systematic reasons, the sustainability indicators and criteria as shown in the Appendix 1 are taken for the assessment of V/HTR. It is expected that V/HTRs will not only be deployed for dedicated electricity generation but also open new markets for nuclear energy by CHP and (co-) production of hydrogen. In addition, V/HTRs are assumed to be compatible with existing U-Pu fuel cycles as well as with new thorium-based fuel cycles. V/HTRs can also be used for waste management purposes by deep-burn capabilities.

Advanced fuel cycles with the main objective of sustainability have demonstrated that opting for the pure reactor grade plutonium cycle can clearly be recommended, whilst in addition, it addresses the increasing issue of the world's growing reactor grade plutonium stockpiles. This option effectively incinerates the reactor grade plutonium generated in LWRs. This cycle uses existing reprocessing facilities to provide the necessary feedstock of fissile material from the LWR spent fuel.

A practical cycle which has already been deployed in commercial size is the Th/HEU cycle as proven in the THTR and FSV. Even though this cycle promotes sustainability, it does require proliferation resistance considerations. However, the Th/HEU cycle can also be established without these issues by breeding ^{233}U in-situ. It is possible to design a V/HTR core as a near-breeder, e.g. by inserting pure thorium fuel elements and extracting them at low burn-up. After decay of the Pa, this fuel can be used for another V/HTR only being fed by ^{233}U/Th MOX. This will drastically reduce the generation of MA. As shown in Figure 7.10. Subsequently, the spent fuel could be reprocessed whereby only a part of the Th and ^{233}U is separated allowing for a mixture of Th/^{233}U that would guarantee a proliferation resistant case. This option offers numerous possibilities, including a dramatic improvement in the conversion ratio, which would lead to drastic savings of the uranium resources.

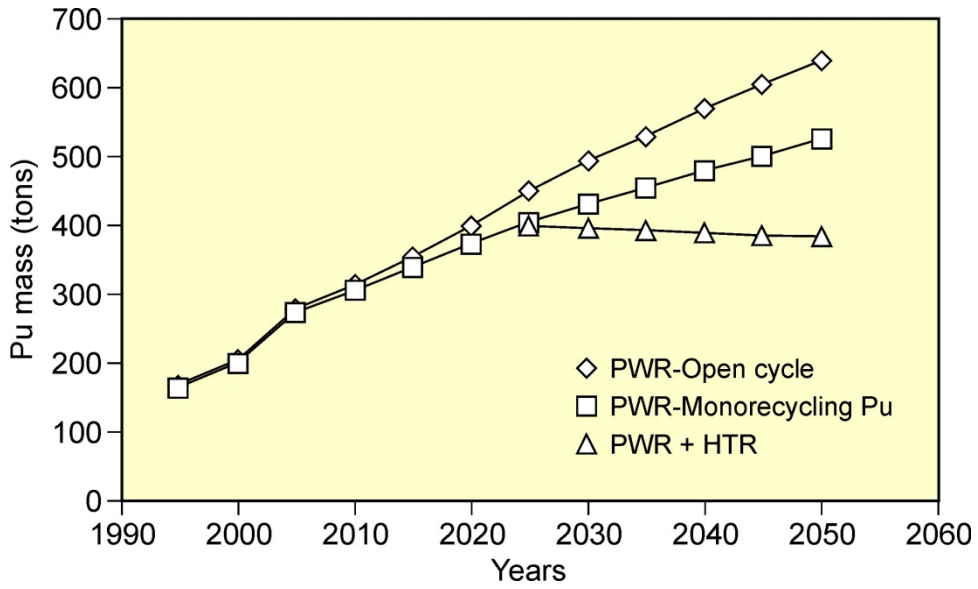

Figure 7.11: Stabilization of Pu stockpiles assuming 1 modular HTR per 3 LWRs [Source: AREVA NC]

Economics

Modular construction of V/HTR may benefit from series and standardization effects. Similar specific overnight construction cost can be expected from mass production as compared to large units. Simplification of safety systems is a further source for reducing cost.

Increased fuel cycle cost will be offset by the enhanced safety features mainly achieved by providing heat capacity via large amounts of graphite moderator as integral part of the fuel element. Simplification of the fuel cycle – Once through (OTTO) *versus* Multi-pass (MEDUL) – might also contribute to considerable savings.

Use of waste heat (CHP) will provide additional financial benefit especially when taking CO_2 taxes into account. For a V/HTR of 100 MW_{el} a figure of about 1,500,000 tons of CO_2 is replaced annually.

There are no established procedures yet for calculating external cost. Nevertheless, it can be assumed that the absence of significant radiological risks due to inherent safety features will have some significant positive impact.

Social Aspects

Due to higher investment cost, work opportunities are positively affected because nuclear energy provides extended direct employment and additional income in the workshops and at the construction site. The same reasoning applies to some renewable energies, too. Lower investment technologies are normally dependent on higher cost for fossil energy imports.

In this context, the developing world nuclear technology provides a basis for the uplift of a nation's economy. In 1963, the economy of the Republic of Korea was comparable to that of actual African countries with a GDP per capita of about $100. In the late 1970's, South Korea opted for the nuclear route through a technology transfer program with the USA. By 1995, the country had a GDP per capita of $10,000 and in 2007 it grew to $25,000 strongly supported by its secure supply of energy by nuclear power.

Modular V/HTRs could also be a good choice for developing countries to support their electricity grids and to provide process heat, e.g., for desalination in arid regions.

Competitive energy supply is a booster for growing economies and favours nuclear energy as a secure and economic energy source. National economies are dependent on the availability of affordable energy under the constraints of climate change auspices. Due to lower cost, nuclear energy also allows more CO_2-free technologies to enter into the energy mix.

V/HTR-specific aspects can be seen in the potential for CHP by providing low-cost process heat to a variety of industrial applications. Hydrogen production opens the market for transport fuel synthesis. The use of CO_2 together with nuclear hydrogen (see Figure 7.12) allows the production of liquid fuels which are compatible with the existing infrastructures and car parks. Nuclear process heat & hydrogen provides nearly double yield from conversion of fossil energy. This also reduces dependencies on energy carrier imports.

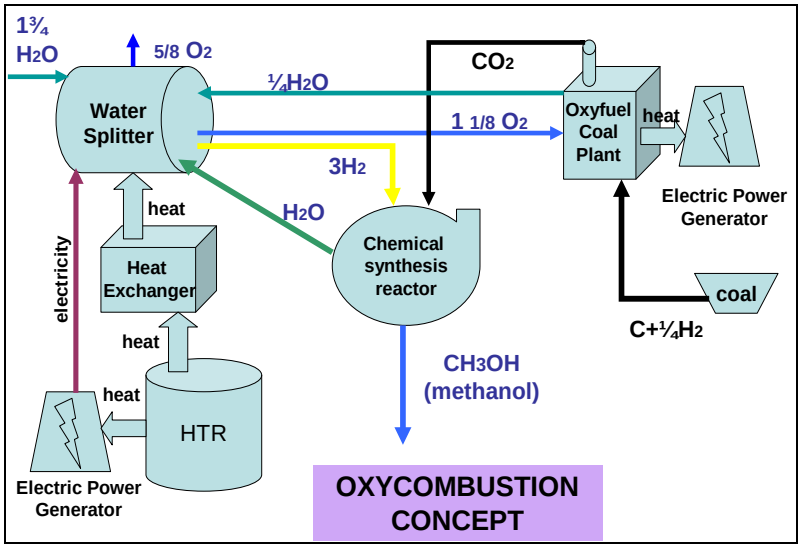

Figure 7.12: CO_2-based fuel production

Higher thermal efficiencies and a good neutron economy of the V/HTR lead to reduced uranium requirements and accordingly extended availability of this resource. This is true for open and closed fuel cycles. Closing the LEU fuel cycle for V/HTR only needs a specific head-end process to separate the kernel from the graphite and the coating. Such processes are subject of the V/HTR System Research Plan and have already shown their feasibility. As is the case for all other Gen IV systems, V/HTR can apply open and closed fuel cycles based on uranium and/or thorium..

The case for thorium in a V/HTR is based on its superior neutron characteristics. In detailed fuel cycle investigations performed in this regard, it was shown that in the order of more than 4 times the fissile material is produced in a thorium/uranium fuel cycle compared to a uranium/plutonium cycle. The choice is mainly dependent on economic and resource parameters.

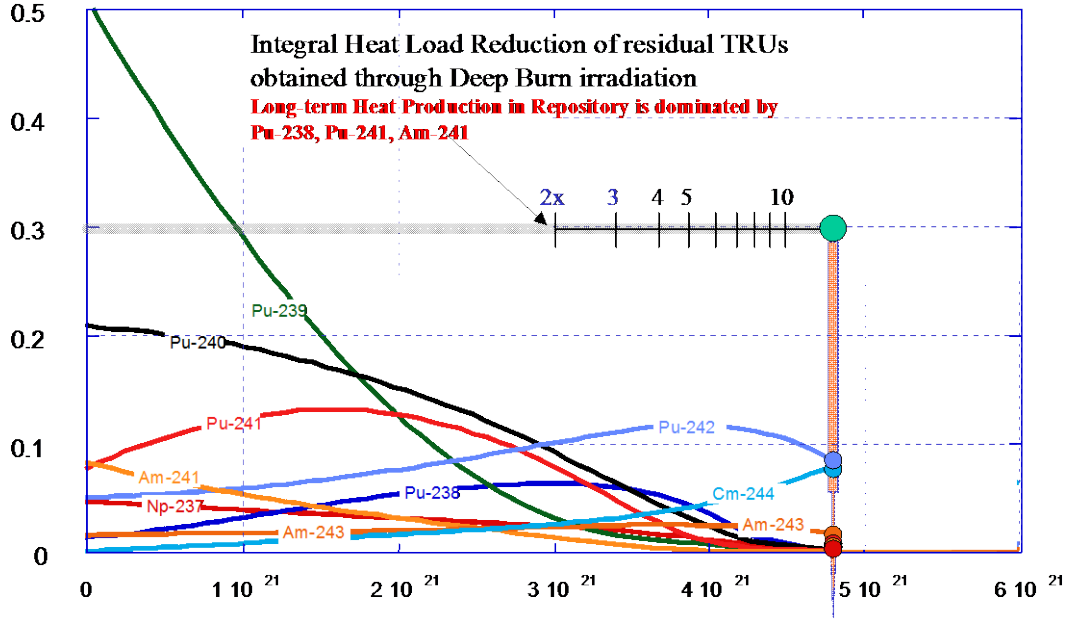

Figure 7.13: Proliferation resistance by deep-burn

Concerning intrinsic proliferation resistance of spent V/HTR fuel, the deep-burn capability of coated particle fuel opens an alternate route to reduce fissile isotopes in the discharged fuel in a single once-through pass (see Figure 7.13). This avoids any diversion possibility because multiple reprocessing and refabrication of the fuel are not necessary. In general, V/HTRs show higher burnup as compared to most other Gen IV systems. This is already equivalent to a multiple recycling of present LWR fuel. Extrinsic proliferation resistance measures can be applied as with any other reactor system.

Ecology and Public Health

Nuclear energy in general has favourable characteristics in terms of specific non-energetic resource consumptions (e.g. steel, concrete, copper etc.) especially when compared to most renewable energies. However, the uranium reserves are limited and strongly dependent on the cost assumptions. The time range is also influenced by the specific consumptions of the different reactor types. V/HTR show high thermal efficiencies and excellent neutron economy due to low parasitic neutron capture of the coolant and moderator. Especially in the CHP mode, specific uranium consumption can be less than half of current LWR needs. The use of thorium broadens the resource basis and provides a further improvement in neutron economy by higher neutron yield and less absorption cross sections. Thorium fuels have already been tested in former HTR in large scale. The feasibility of reprocessing (THOREX) has been demonstrated in pilot plants.

Land use of modular V/HTR is often perceived to be much higher as compared to actual large LWR units. Due to the higher thermal efficiencies, multi-modular V/HTR arrangements of same power do not require a larger footprint. The simplification of safety systems also lead to a lower specific material use. Resettlement necessities in case of accidents are restricted to the plant site as a result of the fission product retention capabilities of the coated particle fuel.

Global warming potential is mainly governed by 'greenhouse' gases like CO_2, CH_4, N_2O, SF_6 and fluorocarbons. Nuclear energy only contributes insignificantly to these emissions when considering the whole life & fuel cycle of the plants. V/HTR can offer additional benefits to CO_2 reduction when substituting process or district heat in the CHP mode. Nuclear hydrogen production and process heat can be used for transport fuel synthesis. Principally, crude oil, tar sands, heavy oils, biomass etc. can be converted without any CO_2 emission by up to double yields of hydrocarbon products. Thus, V/HTRs enlarge the application of nuclear energy beyond dedicated electricity generation and contribute to reduced CO_2 emissions and a stretching of fossil resources simultaneously.

High operational temperatures allow to use the exergy at the top e.g. for hydrogen production and the rest of the temperature range for cogeneration of electricity and low-temperature process heat.

As all other NPPs, V/HTR does not contribute to other noxious, eutrophication, ozone, and photo-oxidant emissions, but displays significant advantages in terms of thermal discharges (see figure 18), which lead already to restricted operation of existing LWR during summer or water shortage. Cooling water requirements are reduced and air cooling has already been demonstrated at the THTR.

Concerning radioactive waste streams, the high thermal efficiency, good neutron economy and high burnup lead to a lower amount of radioactive waste for open and especially for closed or symbiotical cycles together with other reactor types (e.g., burning Pu and MA from spent LWR fuel). However, the large amount of contaminated graphite, which is about 92% of the fuel element mass, needs further attention due to mobile ^{14}C and ^{129}I contents. It will be necessary not only to close the fuel cycle but also to close the 'graphite cycle' for reasons of sustainability. The confinement time assured by the coated-particles under corrosion and radiolytical attack in a final repository still needs to be quantified. First results indicate very low release fractions and long-term stability of the coatings.

Health impacts on workers and public are considered to be very low during operation as shown by the former HTR plants. Potential damage during hypothetical accidents will be limited to the site with no resettlement necessities around the plant. This is of special importance for realizing CHP applications in more populated areas.

As a consequence of the safety behaviour of a well-designed modular V/HTR, it is possible to construct nuclear reactors, which cannot cause catastrophic damages outside the nuclear power plant. In a study performed in Germany, the risk *versus* cost scenario in the event of a catastrophic failure of typical, so-called Generation III+ reactors clearly indicates that in the case of V/HTR civil liability is reduced to a minimum with no credible upset event shown to lead to a financial risk beyond what can be carried by the plant owner. In the event that owners feel that further risk mitigation is preferable, the risk is insurable due to the reduced value of the loss, in the unlikely event that it should occur. This has previously not been possible with the high cost of familiar technologies, related designs and the probabilities of these events occurring.

However

As already stated for the problems of final repository implementation in Chapter 3, the principal advantages of the V/HTR system are not yet generally accepted either by the public or by the relevant industrial, political and scientific stakeholders. The differences between the nuclear systems (LWR vs. V/HTR) as well as their further innovation potentials (e.g. Gen II vs. Gen III or Gen-IV) are practically not reflected, when considering future deployment of nuclear energy. Even a 'rational' evaluation of sustainability indicators 'collides' with the 'intuitive' perception of associated risks of nuclear energy and with the perceived potential of 'risk-free' alternatives, such as renewable energy.

The nuclear community needs to accept the human risk perception mechanisms and find new ways of public communication and adaptations of technology choices to respond to this dilemma.

8 Conclusions & Recommendations

For V/HTR spent fuel management, specific additional considerations have to be undertaken due to the fact that the graphite moderator is an integral part of the fuel element and because the coated particle fuel is directly embedded into the graphite matrix. Figure 8.1 shows the volume ratios of graphite matrix, coatings and UO_2 kernel, for a pebble fuel element. The situation for block type fuel is not much different.

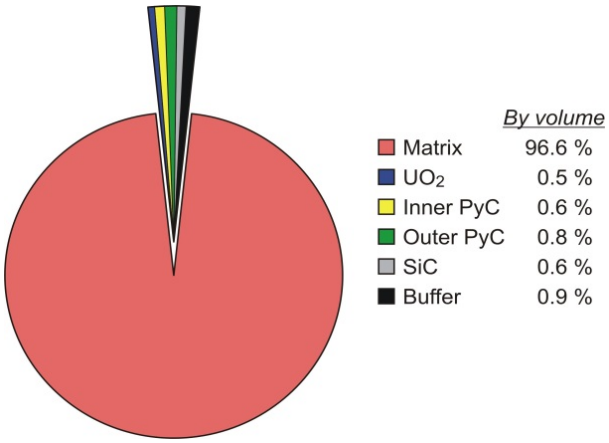

Figure 8.1: Volume ratio for HTR Pebble fuel element

The large graphite fraction of the V/HTR fuel is the 'key' to the inherent safety features of V/HTR and the 'price' to be paid for achieving a large heat capacity of the core with slow temperature transients and a self-stabilization of the maximum accident temperatures not exceeding the fission product retention capabilities of the coated particles, even in absence of active cooling measures. The graphite core structures are part of the overall core design and contribute to the lifetime of i-graphite discharges as quantified in chapter 4.

The safety advantages therefore result in a challenge for managing about 5000–10,000 tons of irradiated-graphite waste per (600 MWth) V/HTR module over an anticipated operational lifetime of 40-60 years. The expected ^{14}C activity of the i-graphite discharge is in the range of 3×10^{15} Bq, which is e.g. nearly 10 times more than the allowable total ^{14}C activity of the German Konrad repository (4×10^{14} Bq) !

It is evident that steps need to be undertaken to reduce the ^{14}C generation in V/HTR by limiting the nitrogen content within the graphite and the primary system as far as possible.

In case of V/HTR, the fuel kernel represents only 0.5% of the spent fuel volume. This shows the principal difference to spent LWR fuel where the bulk of the High-Level Waste (HLW) for direct disposal are the UO_2 pellets themselves. Therefore, different disposal and treatment strategies need to be analyzed for V/HTRs and cannot simply be taken over from waste management technologies established for LWRs.

While direct disposal of one spent LWR fuel element would consume only 0.7 m^3 in the final repository, the direct disposal of spent HTR fuel elements, equivalent to the same energy generation of the LWR fuel element, would require 30 m^3 (see Figure 8.2). This ratio is also compatible with the difference in the power densities of V/HTR vs. LWR cores. If the coated particles will be separated from the fuel elements, they would require a volume of 0.17 m^3 after embedding them in a stable matrix (e.g. sintered glass). This would correspond to 0.11 m^3 of vitrified HLW after the reprocessing of one spent LWR fuel element.

As was discussed in chapter 6, the direct disposal of spent V/HTR fuel would possibly not be acceptable in case of a larger V/HTR fleet, because of the large associated volumes and large amounts of steel for the containers. As the heat generation of the spent fuel is an important factor in a geological repository, the effective use of disposal volume still needs to be quantified in more detail.

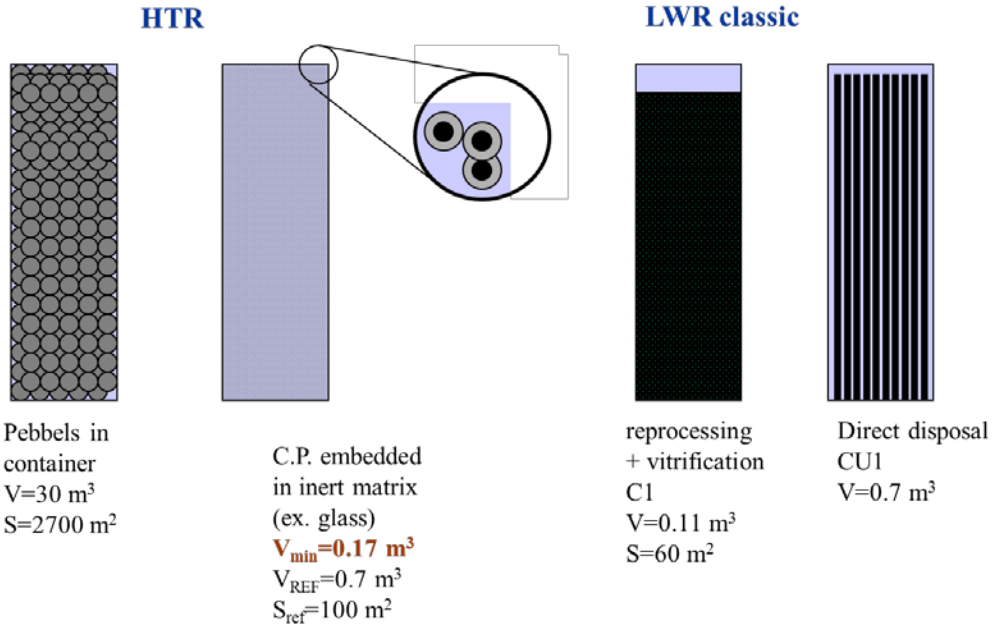

Figure 8.2: Volume of spent fuel from LWRs (normalized to same power generation)

It appears reasonable to further develop and test processes for the separation of coated particles and/or fuel compacts from the graphite matrix as well as the conditioning methods for the embedding of the extracted spent fuel particles.

JRC and SelFrag have performed tests with surrogate **pebble fuel** elements and coated particles in commercially available high voltage (HV) discharge installations [Fütterer 2010]. The product to be fragmented is poured into a vessel, which is filled with water. An electrode delivering repetitive HV discharges under water is plunged into the vessel creating shock waves and the coated particles were liberated intact from their matrix (see Figure 8.3).

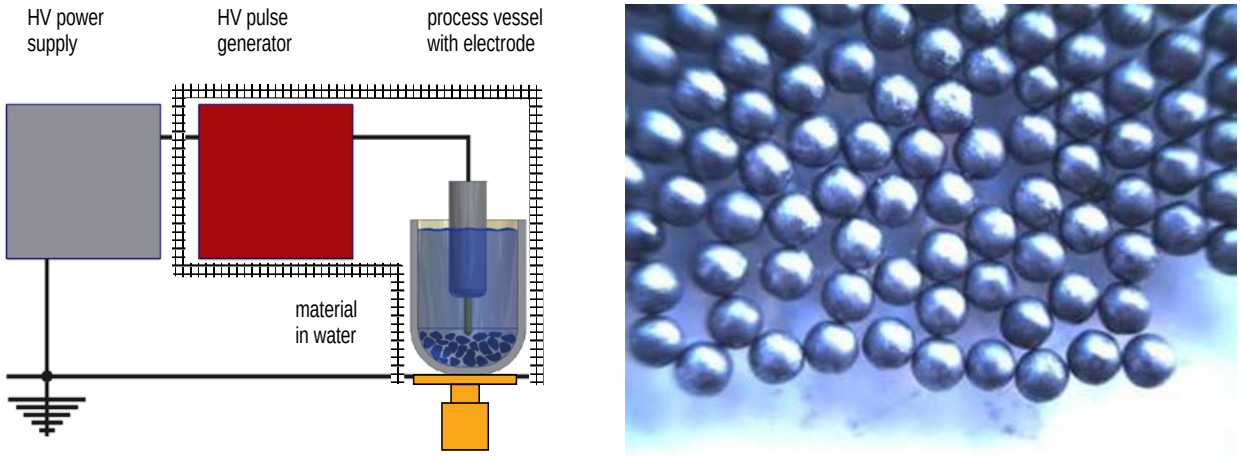

Figure 8.3: Pulsed power defragmentation [source: www.selfrag.com]

The energy consumption of the process is low and the installation is suitable for a hot cell environment and industrially relevant material streams. After further fragmentation of the coated particles, the fragmentation product can directly be fed into classical aqueous reprocessing.

Complementary tests were made by Subatech using HTR-type **compact fuel** with surrogate ZrO_2 TRISO particles to test two separation methods: low ($H_2SO_4 + H_2O_2$) and high ($H_2SO_4 + HNO_3$) temperature acid treatments applying microwaves. In both cases, the TRISO separation was complete but some TRISO layers oxidized at high temperature. At low temperature, the segregation of graphite grains is facilitated by intercalation of sulphuric acid between the graphene layers. The obtained acid graphite intercalation compound (GIC) consists of pure phases of high quality suggesting their potential for industrial recycling. Figure 8.4 shows some GICs after treatment and extraction of the coated particles.

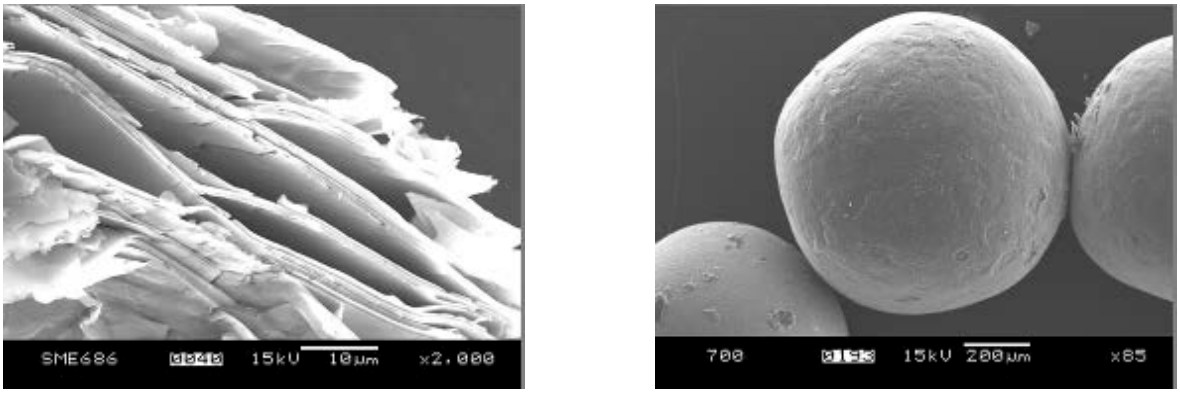

Figure 8.4: Graphite intercalation compounds (GICs) and extracted coated particles from compacts

However, it needs to be noted that these defragmentation processes have only been tested with unirradiated fuel and that they still need to be demonstrated with irradiated and especially with high-burnup fuel to be sure that the surrounding graphite will not be cross-contaminated by defective particles.

The same attention must be paid to the recycling of the irradiated graphite to reduce the accumulation of unacceptable amounts of A3 matrix from the spent fuel and of i-graphite from the core structures. It should also be studied whether graphite can be re-manufactured from i-graphite with sufficient quality and whether residual ^{14}C left after i-graphite treatment contents may be accepted during the manufacture process, if remote handling and shielding are applied. The reuse of purified A3-matrix material also needs to be investigated and remote fuel manufacture processes to be established, for closing the 'i-graphite cycle'.

In case of direct disposal of V/HTR spent fuel, it was shown in chapter 6 that the fission product release characteristics are mainly governed by the corrosion resistance of the pyrocarbon and the SiC coating layers (see Figure 8.5), because the porous fuel matrix does not represent an efficient barrier. In addition, engineered barriers, containers for the disposal of spent V/HTR fuel, the potential embedding of the coated particles in glass or SiC and the choice of the fuel kernel (UO_2 vs. UCO) have a major impact on the release mechanisms in a final repository.

Care must also be taken for the adaptation of engineered barriers to the specific features of spent coated-particle fuel. Due to the large specific volume of the spent V/HTR fuel, one needs to avoid using thick-walled containers and the associated huge mass of steel and the successive generation of gas due to the corrosion of the containers. Fully ceramic containers (e.g. SiC) made of the tails from irradiated graphite may be an adequate approach.

Based on the results obtained in the HTR-N and RAPHAEL projects and assuming a constant and uniform corrosion, the lifetimes of the graphite layer free of fuel pebbles can be estimated. The calculated lifetimes are in the range of 10^7–10^6 years without irradiation and 10^5–10^5 years with the high-dose gamma irradiation. A highly conservative lifetime can be calculated if assuming that

each graphite grain be surrounded by sufficient water. This will reduce the lifetime of the fuel-free graphite layer by 3 orders of magnitude. The real lifetime will be somewhere in-between this border calculations.

For more reliable calculations, further information about the inner surface area of the A3 matrix and the penetration behavior of diverse leachants as well as leach and corrosion tests with irradiated fuel elements are still necessary.

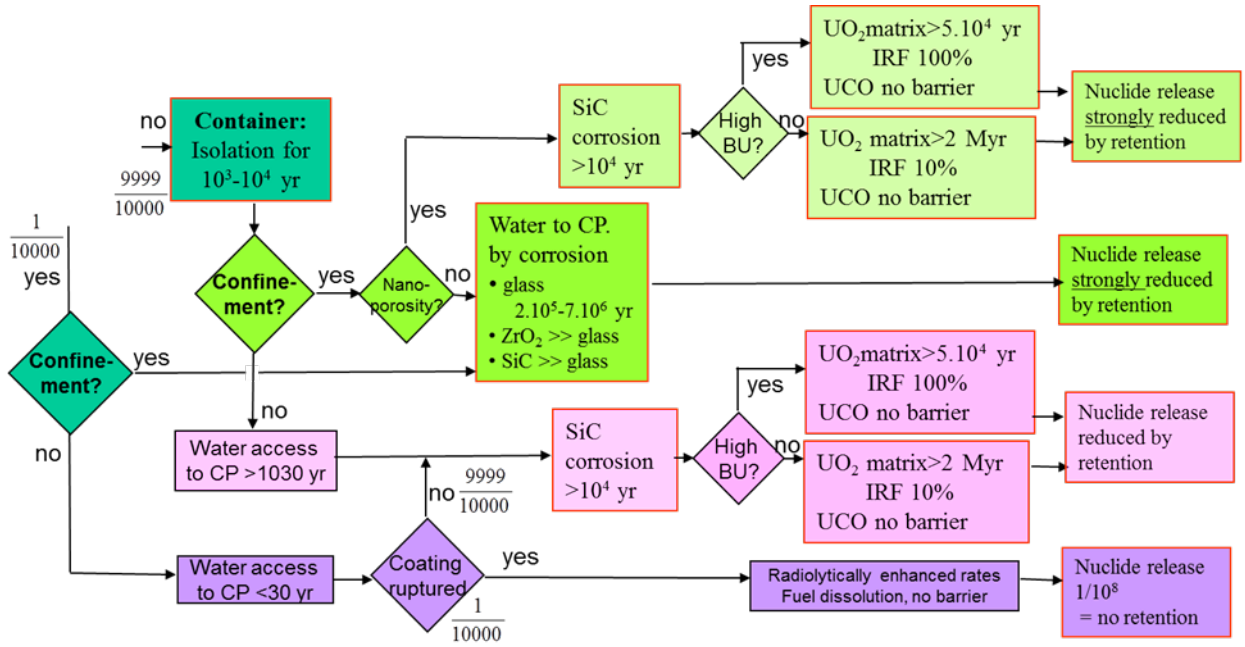

Figure 8.5: Assessment of disposal behavior for coated particle fuel

Analyses have been made on the mechanisms dominating the stability and life time of the particle coating and the dissolution of the spent nuclear fuel kernels in groundwater/brines and associated release of radioactivity. Previous studies have indicated that estimated SiC life times in water will be about 10,000 years. The chemical resistance of SiC and ZrC, both as powders and in the form of coating layers, was determined under repository conditions such as salt domes, granite-based repositories and clay formations (see Figure 8.6).

The results show that the CVD-deposited SiC layers of the unirradiated dummy coated particles, which did not yet possess an outer PyC layer, have a lifetime between 10^3-10^4 years and show a visible dependence on temperature. The behaviour of irradiated coated particles still needs to be investigated to provide reliable evidence for the potential benefits with regards to final disposal of coated particle fuel.

In previous studies, potential secondary reaction products of SiC with water were not yet taken into account. The principal solid reaction product is the formation of SiO_2 layers on SiC, both in air and in water. In [Exter 2007] it was shown clearly that SiC oxidizes relatively rapidly to SiO_2 even at room temperature. The SiO_2/SiC interface is dense and may probably be considered as diffusion barrier. It is likely that the reactivity at the SiO_2/SiC interface might play a key role. Dissolution of SiC would then be a two-step mechanism:

(1) oxidation of SiC to SiO_2,

(2) dissolution of SiO_2.

The dissolution mechanism of SiO_2 is well known. The first step is the dissolution at a constant rate. Thereafter in a second step, equilibrium with the aqueous solution is obtained and this corresponds to a discontinuation of the dissolution reaction.

In recent studies [Chaou 2014], the alteration of BISO (with pyrolytic carbon) and TRISO (with SiC) particles under geological conditions were simulated at temperatures of 50 and 90°C and in the presence of synthetic groundwater. Solid state (scanning electron microscopy (SEM), micro-Raman spectroscopy, electron probe microanalyses (EPMA) and X-ray photoelectron spectroscopy (XPS)) and solution analyses (ICP-MS, ionique chromatography (IC)) showed oxidation of both pyrolytic carbon and SiC at 90°C. Under air this led to the formation of SiO_2 and a clay-like Mg–silicate, while under reducing conditions (H_2/N_2 atmosphere) SiC and pyrolytic carbon were highly stable after a few months of alteration. At 50°C, in the presence and absence of air, the alteration of the coatings was minor. In conclusion, due to their high stability in reducing conditions, HTR fuel disposal in reducing deep geological environments may constitute a viable solution for their long-term management

It is very important to study the potential equilibrium between the SiO_2 surface on SiC and the aqueous solution. If this mechanism can be validated experimentally, life times much longer than 10,000 years may be achieved for the SiC coating. But this still needs to be fixed, before long-term retention of fission products within the coated particles can be claimed in a credible way. In this context, also the barrier function of the PyC layers needs to be quantified in addition and in correlation to the SiC layer. It is proposed to perform leach experiments with fresh irradiated coated particles to study the corrosion mechanisms in a realistic and accelerated way under different geochemical conditions.

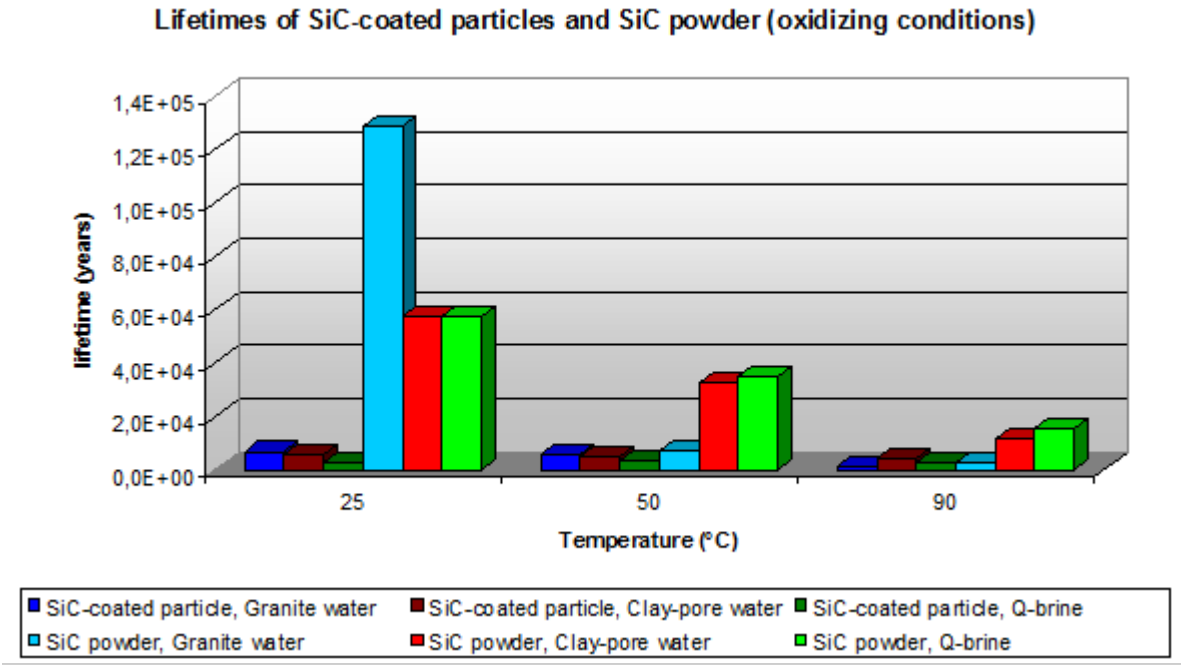

Figure 8.6: Temperature dependence of lifetime for different SiC grades in Q-brine [Exter 2007]

It also should be pointed out that the leaching of long-lived activation products, such as ^{14}C and ^{36}Cl, from the spent fuel matrix also needs to be considered, because these activation products play a major role in performance assessments of final repositories. As already mentioned, the spent fuel matrix of V/HTRs contains significant amounts of ^{14}C stemming from activation of ^{13}C and ^{14}N. Figure 8.7 shows that the ^{14}C concentration is quite high at the surface of the fuel element, which means that this part of the ^{14}C will be leached, in a prominent way.

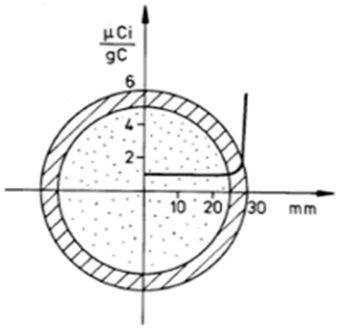

Figure 8.7: Radiocarbon concentration profile in a spent V/HTR fuel pebble

Therefore, it is quite essential to reduce any nitrogen ingress into the primary circuit during the operation of the V/HTR plant and possibly not to expose the fuel elements to air before inserting them into the reactor. During the manufacture of nuclear graphite, the nitrogen content should also be reduced (e.g. by using inert gas during graphitisation).

Returning finally back to the question of potential socio-technological benefits of the safety and waste management features of the V/HTR system, recent experiences in Germany may highlight the future expectations:

Although the AVR and the THTR have both been permanently shut down since the end of the 1980s, they are still subject of the public debate, even after more than 30 years. The postulated safety advantages and the management of the specific V/HTR waste are still discussed in a rather controversial way.

It needs to be kept in mind that the V/HTR development in Germany was strongly defended by the 'catastrophe-free' safety features and the expected advantages of the coated particle fuel during final disposal. The postulation of superior safety as compared to other nuclear reactor systems was questioned by the AVR water ingress accident in 1978 and the observation of excess core temperatures revealed by the so-called melt-wire experiment [see http://www.fz-juelich.de/portal/DE/UeberUns/selbstverstaendnis/verantwortung/avr/Aktuelles/_node.html]. This discussion had a quite fatal impact on the credibility of the whole nuclear community, because AVR was presented as an example for superior passive safety features, but was finally also experiencing technical deficiencies which can especially be expected for any new type of innovative technology. In addition, disputes are still going on about potential increase of thyroid cancer in the vicinity of AVR and THTR, despite the fact that the radioactive releases of these plants have been insignificant following expert judgements.

It is questionable whether new designs of V/HTR can only be based on the fission product retention characteristics of the coated particle fuel. Advanced containment and filter designs as well as catalytic recombiners to cope with flammable gases in case of air or water ingress need to be developed to achieve enough confidence in the safety assessment organizations and in the public. Much more R&D is necessary to underline the benefits of the V/HTR safety features / advantages.

The decommissioning of the AVR generated new challenges due to the highly contaminated primary circuit, the presence of graphite dust and the open issues of irradiated graphite retrieval, conditioning and disposal. More than 650 M€ are being spent to fill the primary circuit with light-concrete, to extract the reactor pressure vessel (including the graphite internals) from the containment and to dispose it nearby for several decades, until the disposal and the conditioning methods are available. Special attention is advised to the waste characteristics of the carbon brick surrounding the graphite reflector, due to its very high content of impurities and successive activation products. Little is yet known about this special kind of radioactive waste which should be avoided for any future V/HTR concept.

The spent fuel from THTR has been stored in the Ahaus interim storage facility since 1992, whereas the irradiated AVR fuel is still stored in Juelich within 152 CASTOR containers. The

transport of the AVR pebbles to the Ahaus storage is heavily opposed by the Ahaus and Juelich public initiatives organizing demonstrations in Juelich and Ahaus against a transport of these CASTOR containers. It is even still subject of debates in the State and Federal parliaments how to proceed with the spent fuel of AVR. As an alternative, it is investigated to ship this spent fuel back to the USA for reprocessing, as the uranium enrichment is raising proliferation concerns.

The enormous effort for the decommissioning of the small experimental AVR reactor has to be avoided for any future V/HTR project. This needs a different engineering approach by reflecting the decommissioning strategy already from the beginning of the future V/HTR reactor designs. The management, transport and storage of V/HTR fuel is not perceived in a different way, as compared to other radioactive waste from other reactor systems. The mere volume of the spent fuel from V/HTR and the relative high uranium enrichment raises additional concerns, in the general and political public.

Technical maturity may establish at the end of a consequent, long-term R&D strategy, whilst steadily pinpointing to the challenging R&D goals not only for the nuclear island, but also for the associated waste issues. Pilot plants and prototypes, such as AVR, THTR and possibly HTR-PM, are part of an R&D program and not 'simply' the end-points of a nuclear system development. It also needs to be mentioned that competing conventional and nuclear technologies are subject of permanent technical and economical improvements. Year by year, they are compiling additional operational experiences which can hardly be compensated by the introduction of new nuclear system with limited operational results. V/HTR would have the advantage to make use of the most developed, efficient conventional energy conversion systems, such as super-critical steam turbines, as the V/HTR gas temperatures are sufficient for this purpose. Hopefully, the V/HTR community will take the chance of the Chinese HTR-PM prototype to focus on a common 'harvesting' of operational results and a world-wide synergetic development of the nuclear system and associated waste management issues which are equally important for public acceptance.

9 Appendix

Sustainability Criteria & Indicators

Dimension	Criteria	Sustainable Development Indicators for Electricity Generating Technologies	
		General	**Additional for Nuclear Systems**
Economy	Private cost	1. Production cost (total private life cycle cost)	
	Public cost	2. External cost	
	Work opportunity	3. Direct employment	
	Income	4. Added value	
	Security of supply	5. Fuel import dependency 6. Reserves / Production ratio	
Social	Proliferation risk		7. Intrinsic proliferation resistance features 8. Extrinsic proliferation resistance measures
	Public acceptance	9. NIMBY (Not in my backyard)/BANANA (Build absolutely nothing and nowhere)	
	Risk aversion		10. Kind of risk constraints 11. Nature of risk Source 12. Dimension of risk consequence
	Public security		13. Measures necessary against terrorist attacks
Ecology & Public Health	Non-renewable resource use	14. Use of energetic resources 15. Use of aluminium 16. Use of iron 17. Use of copper	
	Land use & occupation	18. Area for life cycle 19. Resettlement necessities 20. Noise pollution 21. Change of landscape	
	Climate change / air & water quality	22. Greenhouse gas potential 23. Acidification potential 24. Eutrophication potential 25. Ozone 26. Toxic emissions 27. Radiological emissions 28. Discharged waste heat (e.g. cooling water)	
	Waste generation & management	29. Radioactive waste 30. Non-radioactive, toxic waste 31. Non-radioactive, non-toxic waste 32. Confinement times	
	Safety & health	33. Health impact of normal operation on workers 34. Health impact of accidents on workers 35. Health impact of normal operation on public 36. Health impact of accidents on public 37. Potential damage of severe accidents (range)	
	Ecosystem	38. Biodiversity/species	

Source: European RED-IMPACT Project

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