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WP4
HTR Specific Wastes – Executive Summary

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1 Introduction

HTR-N is one of a family of HTR projects being studied under the EC's 5th Framework programme. HTR-N itself, being compiled in response to EC contract FIKI-CT-2000-00020, has five primary work packages, dealing respectively with nuclear physics tools and data (separate packages for block type of reactors and pebble bed type reactors), fuel cycle, HTR specific wastes and the long term behaviour of disposed spent fuel. The work and findings of work package 4, the specific wastes arising from HTRs, are detailed in a WP4 Final Report. A summary of this main WP4 report is presented in the sections below.

2 Description of Work Areas, Tasks and Deliverables

This report is concerned with HTR-N Work Package 4 only, the specific wastes arising from HTRs. The objective of the work was to examine all waste arising from the lifetime operations of HTRs. To achieve this objective the following areas of work have been undertaken:

- **Activation analysis** has been performed to quantify the nature and volumes of spent fuel and of irradiated graphite from HTR operations and decommissioning.

- **A comparison between HTR and LWR waste volumes** has been undertaken, on an equivalent generated output basis. Differences in generated volumes of High Level Waste (HLW) and Intermediate Level Waste (ILW) are determined by comparison of HTR waste streams with the waste streams of Light Water Reactors (LWRs).

- **A HTR literature archive** has been compiled on HTRs in the world that have shutdown. From this archive, information on the quantities of waste arising from operational and decommissioning has been extracted. The information has been used to compare operational and decommissioning wastes quantities for shutdown HTRs. This has enabled trends in generated wastes and HTR design to be identified that might be applied to future HTRs.

- **Waste minimisation** is considered in the context of mitigating the routes to waste generation identified from the above work. For example, the potential for power circuit contamination is considered by analogy with BWRs, the only other common direct cycle design, and from consideration of the contamination of the gas side of gas reactor boilers in the UK.

- **Wigner energy** accumulation in irradiated graphite has been considered by comparison with existing reactor designs.

These areas of work are reported in the main part of this report. The completion of this work addresses the following HTR-N, Work Package 4 specific tasks:

- Task 4.1, Operational Waste.
- Task 4.2, Decommissioning Waste.
- Task 4.4, Comparison of Wastes.
- Task 4.4, Waste minimisation.

The deliverables in the form of the HTR literature archive (comprising both operational and decommissioning waste data), a comparison of HTR and LWR wastes and a proposal for HTR waste minimisation are presented in the full WP4 report. This is a summary of that report.

3 Work Description and Results

The work undertaken is categorised into the work areas identified in Section 1 and is summarised, along with the results, in sections 3.1 to 3.5 below.

3.1 Activation Analysis

Activation analysis was performed by CEA to characterise the spent fuel and graphite arising from HTR operations and decommissioning. This was based on the block type of HTR, GT-MHR. The fuel was assumed weapons grade Pu in the form of PuO_2 kernels of 200μm diameter, grouped within graphite compacts that are inserted into graphite fuel assemblies. The burn-up assumed is 640 GWd/tIHM, which equates to 770 irradiation days. The graphite and spent fuel analyses are considered separately below.

Graphite Analysis

With the GT-MHR design of HTR a proportion of the graphite reflector blocks are replaceable and the activation analysis results are presented on the basis of a proposed period-of-replacement.

The assumed impurities of the graphite were a representative average of levels found in actual and former nuclear graphite grades. Activation analysis was performed at different locations within the reactor, for example, within the fuel assemblies, within the permanent side reflector and within the central and outer replaceable reflectors. The activity levels of each radionuclide after irradiation were compared against the corresponding French ANDRA limits for surface disposal.

From analysis, the key nuclide contributors to graphite activity were identified as H3, C14 and Cl36 (illustrated in Figure. 1). H3 and Cl36 arise from activation of lithium and chlorine impurities, respectively, whereas C14 arises from activation of C13 as part of the graphite itself and from N14 trapped in porosities of the graphite. The predicted activities of these radionuclides for zero decay time at various graphite locations are plotted in Figure 2 as a percentage of the applicable ANDRA limit. An irradiation of three years was chosen since this is close to the residence time (840 days) of the graphite fuel assembly blocks in the reactor. It

was determined that the activity is highest for the inner replaceable reflector. For all graphite locations, tritium initially has the highest activity compared to the ANDRA limit. In fact, the limit is exceeded by up to 15% for some graphite locations but the nuclide is short lived (a 12 year half-life) and the levels of tritium would not prevent surface disposal in the longer term. Of the long-lived radionuclides the percentage margins to upper activity limits are lowest for C14 and Cl36. Other nuclides also arise from irradiation of impurities, most of them metallic in nature, but the predicted activity levels are only the order of 1% or less of the corresponding ANDRA limit. The ANDRA package acceptance limits are listed in the main report.

The activity levels for the inner replaceable reflector graphite were calculated for times of irradiation ranging from 2 to 60 years and are plotted for the H3, C14 and Cl36 radionuclides (the limiting ones from the ANDRA perspective) in Figure 3. From this it is determined that the increase in specific activity due to C14 is approximately proportional to the time of irradiation. By linear interpolation the highest C14 activity in the replaceable reflector graphite approaches the ANDRA limit after approximately six years of irradiation. Based on the incentive to minimise the volumes of higher activity waste, whilst retaining an awareness of any potential increase in total graphite waste, it might be considered beneficial to replace the replaceable reflectors every six years.

It should be noted that there are other mechanisms for radionuclide activity within graphite that were not assessed as part of the activation analysis. For example:

In the case of tritium:

- H3 arising from the reaction of He3 in the primary coolant.
- The production of H3 in the fuel material, by ternary fissions.

In the case of C14:

- C14 formed on the basis of N14 that may be an impurity present in the coolant gas.
- The formation of C14 from O16 or O17 (n, alpha reactions) within the oxide fuel. There is then the possibility of diffusion of this C14, as CO and CO_2, to the graphite blocks.

Spent Fuel Analysis

The main radionuclides, in the form of fission products and actinides, were identified from spent fuel activities calculated over an entire fuel cycle. The masses of the main long-lived fission products and the actinides predicted for a HTR, GT-MHR type reactor are compared with those determined for LWRs operating with burn-ups of 33 GWd/tIHM and 45 GWd/tIHM. The comparison is on the basis of the same generated electrical output and after a 5 year decay period. The key results are presented in Figure 4 for fission product masses and Figure 5 for actinide masses. In most cases, the amounts of a particular radionuclide within the spent fuel are of a similar order of magnitude for both reactor designs but some differences for significant isotopes are as follows:

- Over the first few 100 years the activity of the spent fuel arises mainly from Cs137 (through β^- decay to Ba137m and then gamma emission) and from Sr90 (through β^- decay to Y90 and then gamma emission). These nuclides are also the main concern for radioactive release and potential dose to the public over the same time period. There is an advantage for HTR in that the predicted masses of Cs137 and Sr90 within the spent fuel are less than for LWR.
- In the longer term the spent fuel activity is dominated by actinide decay. Here the HTR shows a slight disadvantage with a total predicted actinide mass higher by up to ⅔ than for LWR.
- The Cm244 and Cm242 isotopes have the highest specific neutron emission rates of the identified actinides within the spent fuel. Even though the Cm244 isotope comprises less than 1% of the total actinide mass its contribution to the overall neutron production rate is significant for the first ~100 years post irradiation. Due to its short half-life (163 days) Cm242 is less important. The predicted masses of Cm (comprising predominantly the Cm244 isotope) in the fuel are similar for the HTR and for the LWR at the lower irradiation, whilst at an irradiation of 45 GWd/tIHM the LWR design has a slight advantage.

Even taking into account the specific differences identified above the predicted HTR (GT-MHR reactor design) and LWR fuel radionuclide inventories are sufficiently similar that the radiological shielding requirement for HTR spent fuel will be essential no different from that for LWR fuel. In terms of the potential for radioactive release and potential dose to the public Cs137 and Sr90 nuclides are dominant in the shorter term with Pu isotopes (particularly Pu240) becoming the most important well beyond 100 years. The HTR reactor shows a disadvantage in that the mass of Pu nuclides is up to 2 times that for LWR on the same generated output basis. It is considered that this disadvantage is more than off-set by improvements in fission product retention afforded by Si carbide (or Zr carbide) coated fuel particles with the HTR fuel design.

As a note of caution it should be emphasised that some of the predicted differences between relative nuclide masses within the spent fuel of LWR and HTR can be attributed to the different assumptions and methodologies used, for example:

- The LWR fuel nuclide inventory was derived from experiment so the data is more comprehensive (e.g. it takes into account the diffusion of contamination from fuel material).
- The HTR (GT-MHR) is loaded with PuO_2 fuel, whereas the LWR system uses UO_2 fuel. Therefore the ratios dealing with the actinides generated in the two systems may present significant differences and the normalised ratios and nuclide mass comparisons have to be considered cautiously.

3.2 Comparison of HLW-ILW Volumes arising from HTR and LWR Systems

The quantities of spent fuel and higher activity graphite are compared below for HTR and LWR systems. The fuel in the LWR is typical low enrichment U (~3% enriched) within UO_2 sintered pellets. For the HTR GT-MHR design the fuel is weapons grade Pu (more than 95% fissile isotopes) in the form of PuO_2 kernels of 200μm diameter with a multi-layer coating of porous carbon buffer, pyrolytic carbon and silicon carbide. These coated particles are embedded in a graphite matrix to make a graphite/fuel compact. The fuel compacts are inserted into channels within hexahedral graphite blocks that are designated as fuel assemblies.

Spent Fuels Comparison

The mass and corresponding volume of annual discharged spent fuels (HLW) is determined for both HTR and LWR. The LWR has been considered with burn-ups of 33 GWd/tIHM and 45 GWd/tIHM these being representative of normal and high burn-up fuel cycles employed on current LWR designs. The results and key assumptions are tabulated below. To enable a direct comparison the volumes of HLW are determined on a per GWe-year basis.

Reactor System: Type:	**LWR 33** **1300 MWe**	**LWR 45** **1300 MWe**	**HTR** **GT-MHR (278 MWe)**
Initial Heavy Metal mass (tIHM)	103.4	103.4	0.7007
Fuel residence time (months)	38	54	28
Specific volume of a fuel assembly for waste management (m^3/tIHM)	2	2	25.7 (Fuel/graphite compacts) 3.52 (Fuel only)

Annual Quantities of Discharged Spent Fuel based on the Same Generated Power:

Mass of spent fuels (tIHM/GWe-y)	**25.1**	**17.6**	**1.1**
Volume of spent fuels (m^3/GWe-y)	**50.2**	**35.2**	**28** (Fuel/graphite compacts) **3.8** (Fuel only)

With the fuel material remaining integral with the graphite of the compacts there is a difference in the annual discharged spent fuel volume per GWe-year between the LWR type and the HTR. The advantage appears to be in favour of HTR, particularly in the case of the LWR 33 GWd/tIHM, by nearly a factor of two. This advantage has to be put into its proper perspective as the volume of HTR

spent fuel is considered without encapsulation (due to the assumed good qualities of confinement of triso particles).

There is a more significant difference in the volumes of annual spent fuel discharges if the graphite of the HTR fuel compacts is no longer taken into account. In these circumstances, a ratio of 1 to 10 for the volumes of discharged spent fuels between the HTR (GT-MHR reactor) and LWR system is estimated. This ratio is increased further in the case of the LWR 33. Moreover, we have seen that the graphite of the fuel compacts could be regarded as surface waste as it is so little activated (see Figure 2). However, this hypothesis has yet to be confirmed. There are possible contamination mechanisms not accounted for in the activation analysis, for example, diffusion from the fuel particles. In any case, the HTR (GT-MHR design) shows a significant improvement in the minimisation of high-level waste.

Thus, the annual volume of spent fuels arising from an HTR reactor, GT-MHR Pu loaded, is about 3.8 m^3 of HLW per GWe-year (spent fuels without graphite) or 28 m^3 when the graphite of the fuel compacts is also considered to be spent-fuel (no separation).

It should be emphasised that the predicted advantages in spent fuel waste volume are mainly a consequence of the weapons grade Pu oxide fuel loading assumed in the HTR study. The adoption of a more typical low enrichment U oxide fuel or of a mixed oxide loading would erode much of this advantage.

Global Non-surface (HLW+ILW) Output Comparison

To provide a comparison of the non-surface disposal output account should also be taken of volumes of higher activity graphite. This waste stream is an additional one for HTR that is not applicable to LWR systems. Other potential ILW waste output resulting from operations of reactors (e.g. compacted activated materials, used resins) is not considered in this study as it amounts to only a few m^3 per reactor year in LWR systems, and it is anticipated that the volumes of equivalent wastes would be similar for HTR systems.

Over the complete HTR lifecycle, the volume of graphite requiring non-surface disposal comprises:

- The 148 m^3 of permanent reflector graphite removed during decommissioning.
- The 405 m^3 of graphite discharged as an integral part of the fuel compacts. This volume is derived assuming ⅓ of the 15.5 m^3 of compact graphite in the core is discharged every 280 days and this continues for 60 years of operation.

With 60 years of operation and a nett electrical output of 0.278 GW the volume of non-surface disposal graphite, normalised to 1 GWe-year, is 33 m^3/GWe-year.

The non-surface disposal waste volume arising from HTR is then the sum of ILW graphite and spent fuel volumes.

The annual quantities of HLW & ILW arising from HTR - LWR systems based on 1 GWe-year of generation are tabulated below:

Reactor System: Type:	**LWR 33 1300 MWe**	**LWR 45 1300 MWe**	**HTR GT-MHR (278 MWe)**
HLW Volume: discharged spent fuel per GWe (m^3/GWe-yr)	50	35	4
ILW annual average volume (m^3/GWe-yr)	-	-	33 (compact graphite and permanent reflector) 9 (permanent reflector only)
Total Annual HLW and ILW Outputs (m^3/GWe-yr)	**50**	**35**	**37** (including compact graphite) **13** (excluding compact graphite)

The global assessment of spent fuels and graphite waste output amounts to approximately 37 m^3 of non- surface waste (HLW+ILW, assuming no operations to separate the fuel from the graphite in the fuel compacts) per GWe-year for an HTR reactor. This comprises 28 m^3/GWe-year of waste arising from an HTR reactor under operations and 9 m^3 of ILW graphite resulting from the final dismantling operations.

The global volumes of non-surface waste generated by the HTR and LWR systems are quite similar falling within the range of 35 to 50 m^3 of HLW+ILW per GWe-year.

A larger gain in favour of the HTR system appears if the graphite involved in the fuel compacts could be separated from the fuel material. This has yet to be confirmed but if this graphite were really classed in the LLW category, it could lead to a considerable decrease of waste arising for the HTR system: only 13 m^3 of non-surface waste per GWe-year. The interest and feasibility of such a treatment for the graphite of the fuel compacts is certainly worth investigating.

Again, it should be emphasised that the predicted advantages of HTR over LWR systems, in terms of the volumes of higher activity wastes generated, are mainly a consequence of the weapons grade Pu oxide fuel loading assumed in the HTR study. The adoption of a more typical low enrichment U oxide fuel or of a mixed oxide loading would erode much of this advantage.

3.3 HTR Literature Archive

A literature archive has been compiled for wastes arising from the shutdown HTRs, AVR, THTR, DRAGON, FSV and Peach Bottom and is included as an appendix to the main report. The first two reactors are of the pebble bed type and the last three are of the prismatic block type. Each of the five stations are briefly described below:

AVR, located at the Juelich site in Germany, was an experimental reactor with a 15 MW electrical output. The station operated for 22 years until 1988. The load factor was 67% equating to a total output of 0.22 GWe-year. The intention now (2002) is to decommission as soon as possible, without waiting for any further safestore period.

THTR, located at Schmehausen in Germany was a power reactor with an electrical output of 300 MW. It operated for an equivalent of 427 days of full power operation giving a total output of 0.35 GWe-year before operations ceased in 1989. The burn-up was 110 GWd/t. The intention is to decommission after about 30 years of safestore.

DRAGON, located at UKAEA's Winfrith site in England was an experimental helium cooled HTR with a power rating of 22 MW thermal. Full power operation commenced in 1966 and operations ceased in 1976 with a total output of 0.053 GWe-year. The burn-up was 100 GWd/t. The intention is to decommission after about 60 years of safestore.

Fort St. Vrain (FSV), located in Colorado, USA, was a power reactor with a 330 MWe output. Approximately 1.5 full power years of operation were realised. It was fully decommissioned by 1997, dismantling operations being carried out under water. Although considerable volumes of qualitative data were available for the decommissioning of FSV, very little quantitative data could be obtained for a reasonable price.

Peach Bottom, located in Pennsylvania, USA had an electrical output of 40 MW. It operated from 1996 to 1974. Peach Bottom is in safestore condition, with minimal decommissioning data available.

Nevertheless, on the basis of information available a comparison of the quantities of the major wastes has been made, using some simplified assumptions. The results of the comparison are tabulated separately below for waste arising from HTR operations and from decommissioning:

Operational Waste

STATION	HLW Mass Kg	HLW Vol M^3	ILW Mass Kg	ILW Vol M^3	LLW Mass Kg	LLW Vol M^3	Total Output GWe-year
DRAGON[3,4,5]	21,600	12[1]	-	[2]	-	[2]	0.053
AVR[6]	32.8		-		6,293		0.22
THTR	75.7		326,030[8]		214,950		0.35
FSV	195[7]		-		-		0.50

1. Assumed to be all HLW, fuel compacts
2. Dragon data does not include ILW/LLW operational wastes – reactor said to be "very clean."
3. Some Dragon operational wastes, (e.g. clothing LLW) were dealt with as part of Winfrith generic wastes.
4. Dragon did have wastes from maintenance, but these are not quantified.
5. Dragon data is for raw, uncompacted, untreated waste
6. AVR quantities exclude gaseous wastes and very low level wastes
7. FSV spent fuel wastes are a maximum, derived by implication
8. THTR also has solid ILW operational waste (absorber and moderator elements) total activity 7.54 x 10^{14}Bq but quantities not given

Decommissioning Waste

STATION	HLW Mass Kg	HLW Vol M^3	ILW Mass Kg	ILW Vol M^3	LLW Mass Kg	LLW Vol M^3	Total Output GWe-year
DRAGON[1,2,3]	None		40,500	57.9	1,246,000	1780	0.053
AVR[4]	None		700,000		2,920,000		0.22
THTR							0.35
FSV[5]					6,820,000		0.50

1. Dragon data is for raw, uncompacted, untreated waste
2. Dragon data is after at least 20 years safestore decay
3. Dragon decommissioning wastes are future arisings, after typically 50 years safestore
4. AVR quantities exclude gaseous wastes and very low level wastes
5. FSV decommissioning wastes are as disposed

Within limits from the above comparisons it has been deduced that:

- Larger reactors produce relatively less decommissioning waste per unit of power produced.
- Reactors with longer operating lives produce relatively less decommissioning waste per unit of power produced.
- The same general deduction is not true of operating waste, particularly spent fuel.
- The 38,000 tonnes of free release material arising from AVR equates to 91% of the total station waste. This is in reasonable agreement with the 95% free release stated for Dragon.

- The volume of HLW determined on the same generated output basis is reasonably consistent for all shutdown HTRs, ranging from 164 to 390 m^3/GWe-year. Even the lowest of these values is significantly greater (by a factor of 6) than the corresponding value determined by the analysis presented in this report. This may be a consequence of the higher burn-up assumed in the analysis for the HTR GT-MHR design fuelled by weapons grade Pu (i.e. 642 GWd/t compared with 100 GWd/t typically for the shutdown HTRs).

3.4 Waste Minimisation

Waste minimisation has been considered in terms of graphite, ex-pressure vessel components and power circuit contamination.

Graphite

The CEA analysis showed that, in the case of the French disposal criteria, the dominant long lived nuclides were C14 and Cl36. These can arise from the activation of nitrogen and chlorine impurities, respectively. It therefore follows that the nitrogen and chlorine content of the graphite needs to be reduced as far as possible. Also the nitrogen content in the helium coolant needs to be kept as low as possible to avoid C14 by contamination. The purging of the pressure vessel during start-up and the implementation of an efficient coolant purification and make-up system were identified as key areas for controlling and removing air ingress.

Ex-Pressure Vessel Components

By analogy with the UK gas reactors, it was found that the activation of components and structures immediately ex vessel, i.e. the high temperature section of boilers and gas ducts, was significantly reduced by the presence of substantial neutron shields. The same would be relevant to the first stages of the HTR power circuit.

Power Circuit Component

Potential sources of contamination in HTR were identified as:

- Graphite or carbonaceous deposits carried over from the reactor.
- Tritium released from the reactor.
- Radioactive silver deposits, caused by fuel fission products.

The possible mechanisms for reducing contamination were identified as cyclones or filtration for the graphite and gas purification for the tritium.

3.5 Wigner Energy

It was concluded that Wigner energy is not a problem with regard to final disposal since the graphite will self anneal at the operating temperatures of HTR.

4 Recommended Future Actions

The following are the recommendations following from the work detailed in the main part of the report:

(i) In order to obtain a more comprehensive understanding of HTR wastes, it is recommended that activation analysis is carried out for the pebble bed type of HTR. The analysis would identify and quantify the volumes of spent fuel and graphite wastes arising; the same comparison between LWR and HTR wastes would be carried out, and would therefore also allow a comparison between the pebble bed and block type of reactors.

It is recommended that the input data for this additional analysis would be along the lines of that recorded in Section 2 of the main part of the report, thereby being in accordance with parameters being studied in WP3 of HTR-N.

This would provide an opportunity to determine the sensitivity of the results to different fuel loadings (e.g. PuO_2, UO_2 or mixed oxide fuel). This could provide confirmation that the lower HLW volume as predicted by analysis, compared to the HLW volume determined from shutdown HTR information, is a consequence of the weapons grade Pu oxide loading assumed in the analysis.

(ii) It is also recommended that an activation analysis be carried out for the steel components, e.g., the pressure vessel etc. Such analysis would identify whether, from a waste disposal point of view, reduction of impurities in the steels would be beneficial.

(iii) It has been noted that experience from UK graphite, gas cooled reactors, contamination from impurities in the primary coolant (principally nitrogen) can cause activity levels several times higher than from the impurities in the graphite. The exact contamination could be different for HTRs, but in any event will depend on the operating conditions of the helium make up and purification system, and the maintenance practices.

There appears to be a lack of written record or knowledge from HTR practice. Nevertheless, it is recommended that the subject be researched as far as possible, as a possible contribution to waste minimisation (as well as operating optimisation).

(iv) It is believed that, for block type HTRs at least, O17 within the oxide fuel in the compacts gives rise to C14. The C14 which in turn may diffuse as CO and CO_2 to the graphite bricks. This subject may require further study.

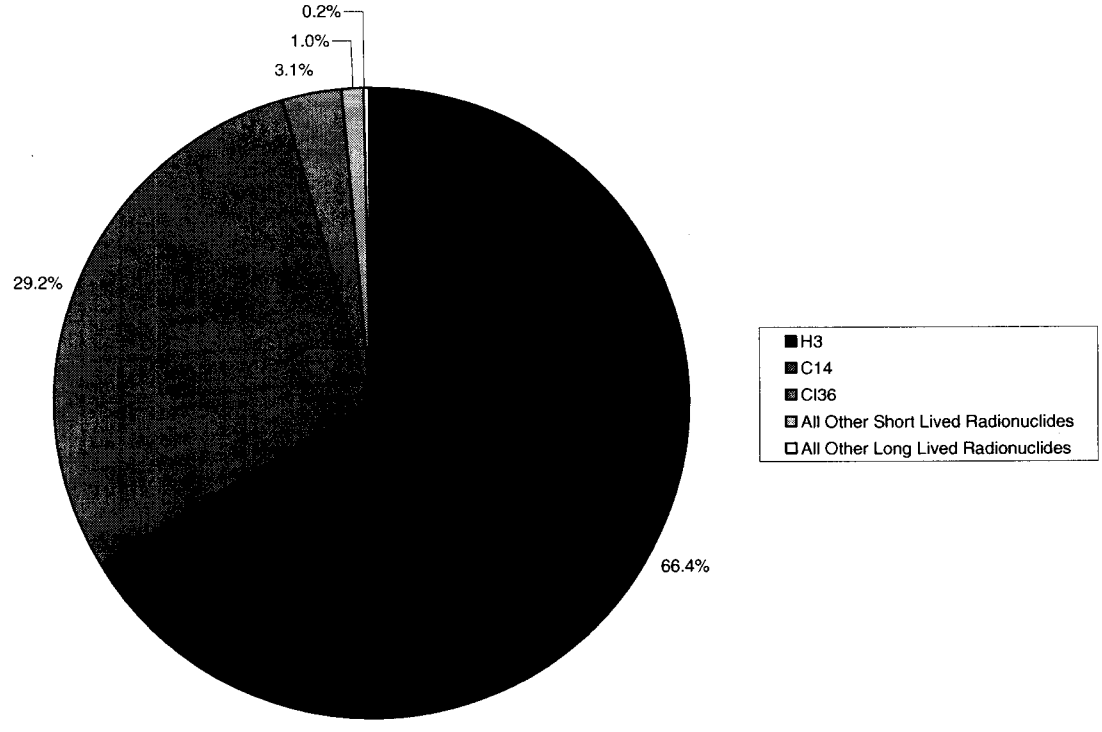
(v) It is recommended that the research, currently being conducted by others, into the potential contamination of the power circuit, be kept under review. With an understanding of this aspect developed it might be possible to modify the HTR operations (or design) to facilitate waste minimisation.

(vi) It is recommended that consultations be held with the suppliers of graphite to determine what levels of purity can be achieved, and whether the methods of purification can be changed to avoid accumulation of Cl36. Such consultation may be the responsibility of HTR-M rather than HTR-N. The co-ordinator of HTR-M has been informed.

(vii) The HTR – LWR waste comparison takes no account of the growth in volume due to undefined conditioning of the spent fuels. It will be interesting in future studies to verify that this growth in volume is not discriminatory and remains the same order of magnitude for both reactor types.

(viii) With time, the dismantling of shut down HTRs will generate further decommissioning data. The AVR at Juelich for example, will soon follow this objective. It is recommended that a formal structure be put in hand to capture quantitative wastes arising, so that at some future date the archive may be updated.

Figure 1 Radionuclide Activity Relative Contributions: Inner Replaceable Graphite Reflector after 3 Years Irradiation and no Period of Decay



Note: The magnitude of the contribution to the overall graphite activity is assumed proportional to the radionuclide activity expressed as a fraction of the corresponding ANDREA limit.

Figure 2 **Key Nuclide Activities with Respect to French Surface Disposal Limits: - Different Graphite Locations after 3 Years Irradiation and Zero Decay**

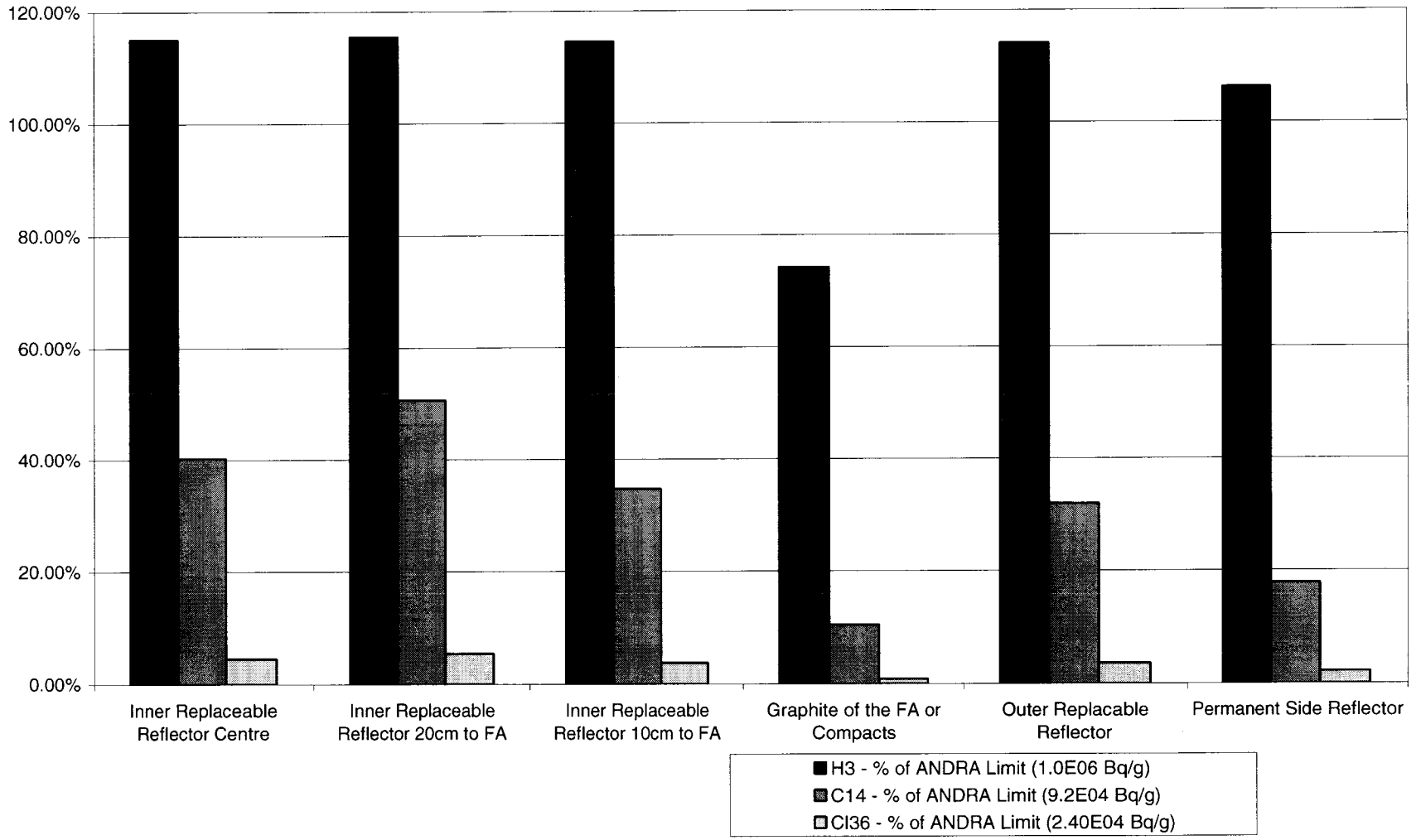


Figure 3 **Key Nuclide Activities for Inner Reflector with Respect to French Surface Disposal Limits: - Different Times of Irradiation**

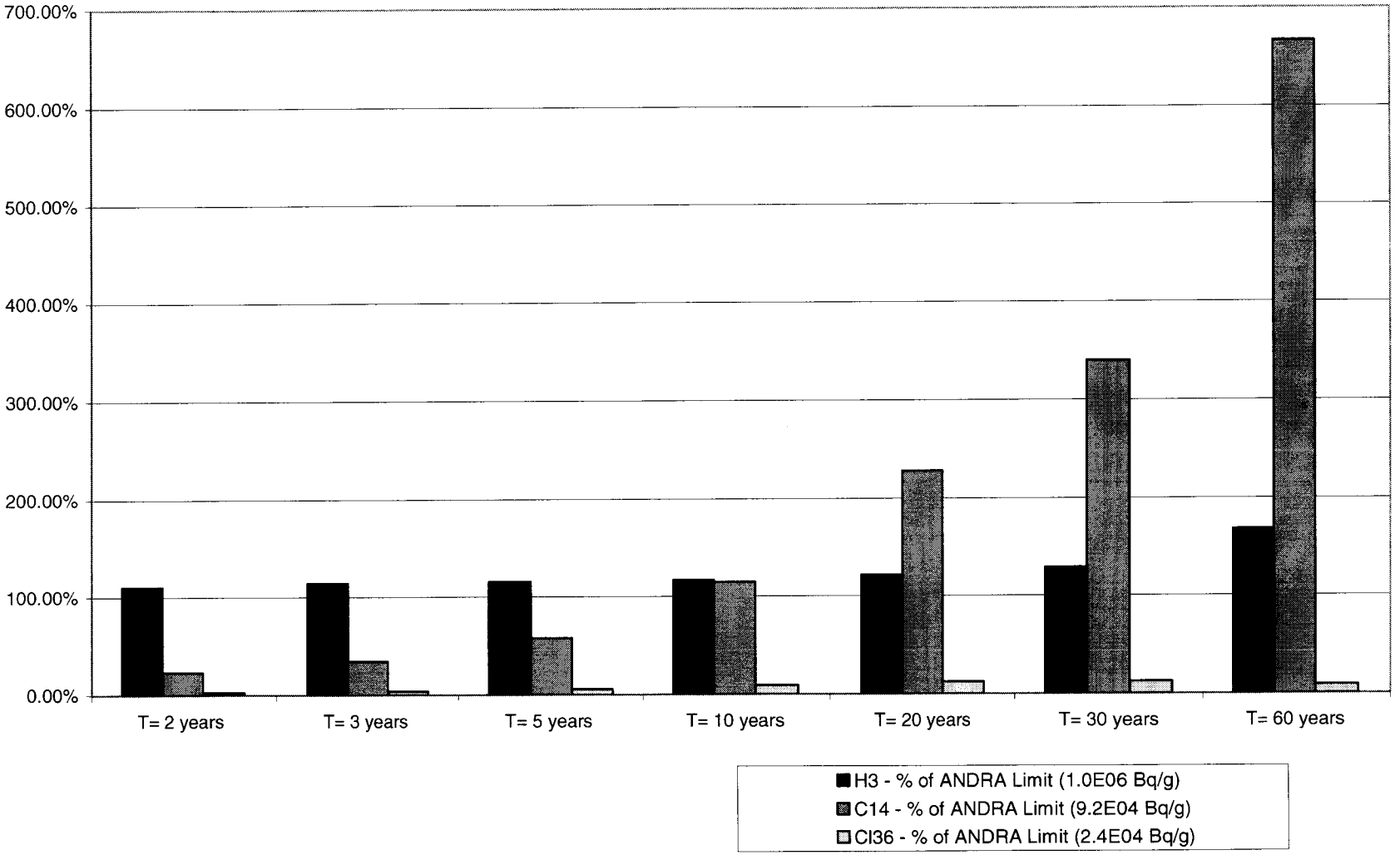


Figure 4 HTR and LWR – Spent Fuel Fission Product Masses

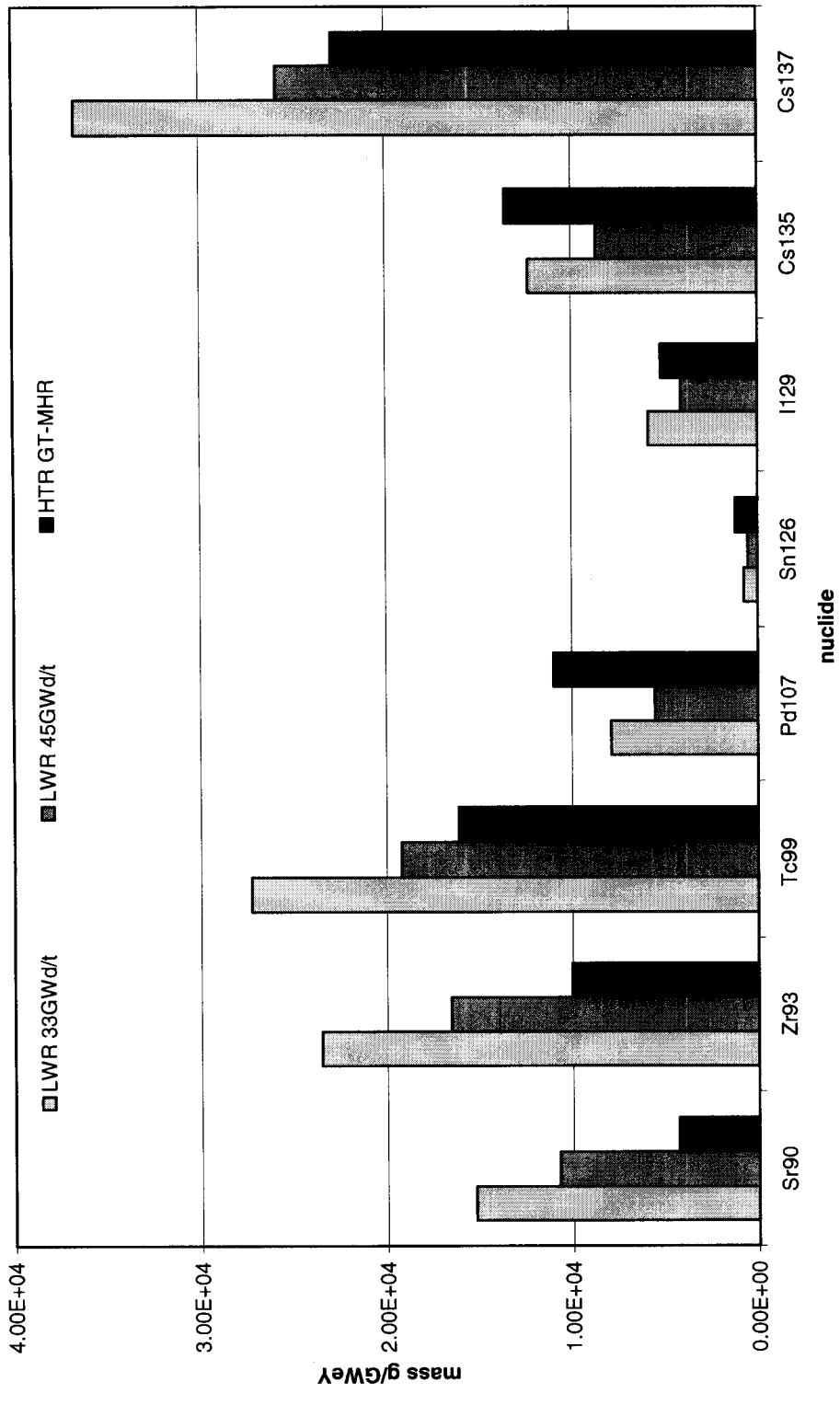


Figure 5 HTR and LWR – Spent Fuel Actinide Masses