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CPSTRESS design proposal and feasibility assessment

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Summary

This document constitutes the CP-STRESS feasibility assessment and irradiation proposal. It presents the main results obtained during the feasibility studies and initiate a design for a new irradiation on TRISO particles for HTR fuel technology. This irradiation constitutes a separate-test effect considering that mainly the thermo-mechanical stability of TRISO particles silicon carbide layer under pressure caused by the release of fission gas on the stability will be assessed. This document provides the main technical information with regards to the feasibility of such an irradiation taking the support of the Idaho National Laboratory (US) into account. A collaboration between the US and EURATOM is foreseen for the completion of the irradiation experiment. This document constitutes the deliverable D.3.1.22 of the ARCHER European project. The official NRG number for this report is 22904.20/13.123153 I&D

Approval

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1 Introduction

TRISO coated particles are one of the most important element of the HTR technology. A brief description of the composition of those TRISO particle is given below.

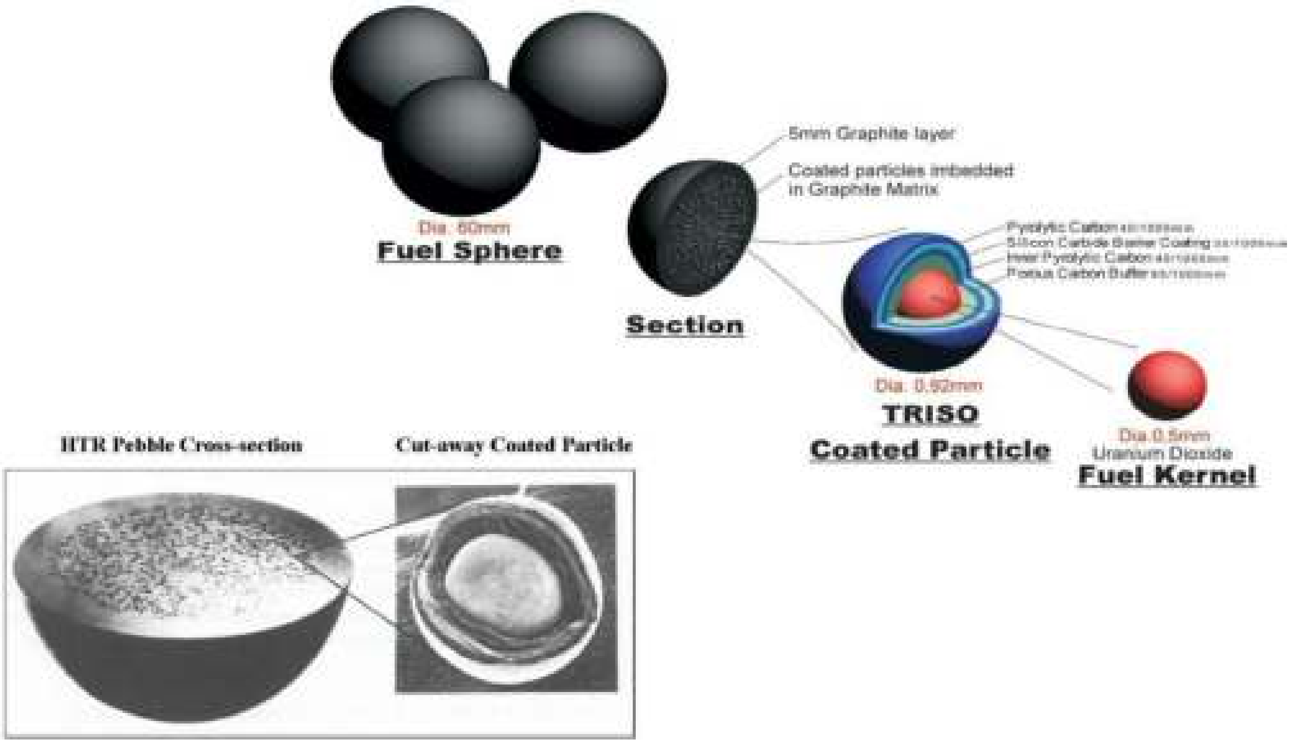

Figure 1 TRISO particles in HTR fuel technology

If the behavior of TRISO particles under irradiation has been extensively analyzed, specific irradiations driven to study the thermo-mechanical stability of silicon carbide layers under high pressure are still required to qualify software models and assess HTR fuel safety. This thermo-mechanical effect of the resistance to the build-up of inner pressure due to fission gas release, alpha particles and carbon monoxide formation on the particle (IPyC layer and SiC layer) represents an important milestone for the integrity assessment of coated particles at high burn up. Considering the specific diffusion behavior of Ag-110m during irradiation, diffusion phenomena will be scrutinized in the CPSTRESS irradiation experiment to come.

NRG, via two former irradiations experiments on TRISO particles, namely Pycasso I & II, possessed the experience required for this experiment. The following feasibility study is based on a potential irradiation within the Petten High Flux Reactor.

To simulate the build-up pressure without having any chemical interaction, surrogate kernel will be coated with Boron which under irradiation will trigger the production of Helium and then induce an inner pressure on both the PyC layer and also on the SiC layer. The same type of coating is also possible with silver to study its diffusion under irradiation.

Experimentally, the objective of the experiment is to bring particles to the failure threshold (i.e. stress of about 350 MPa) so that the thermo-mechanical effect of gas release on the particle can be observed and assessed. Most of the results presented are based on the previous experiences NRG has with the Pycasso irradiation experiments.

2 Assessment of the irradiation conditions

2.1 Temperature of irradiation

The targeted temperature will be in the range between 900 to 1200 °C. Some developments concerning the challenging point of reaching a stabilized temperature of 1200 ^{0}C during the irradiation are discussed in chapter 4.

2.2 Neutron Fluence

The neutron fluence level is chosen to be representative of values that occur at high burn-up (i.e. > 16%). At this level, the dpa values in silicon carbide and the PyC layers are also of similar levels to get representative defects and strengths. The fluence is therefore targeted at $5 \cdot 10^{25}$ m^{-2} for E>0.18 MeV. At this fluence, a dpa value of 3.7 in graphite would be achieved.

2.3 Helium production

The first coating layer (covering the kernel) is made of Boron carbide (B_4C). Boron has a natural composition of 20% ^{10}B and 80% ^{11}B, but can be enriched to a higher grade (max ≈100% ^{10}B). Capturing a thermal neutron, ^{10}B will induce the following n,α reaction:

$$^{10}B + n_{th} \rightarrow \text{He (with E}\alpha = 1{,}47\text{ MeV)} + {}^{7}\text{Li (with E}=0{,}84\text{ MeV)} + \text{gamma (0,48 MeV)}$$

This reaction will produce helium and will induce a pressurization of the coated particles. To achieve representative pressures inside the particle, the amount of produced helium needs to be carefully estimated according to the irradiation conditions within the HFR core. Using a thermal cross section of 3840 barn, the boron burn-up rate is estimated for a range of thermal fluences. The outer positions of the HFR have the most thermalized spectrum with a thermal flux up to $0.6 \cdot 10^{14} cm^{-2}s^{-1}$ (for E_n > 0.058 eV). The in-core positions have lower thermal fluxes in the range of 0.25 to $0.4 \cdot 10^{14} cm^{-2}s^{-1}$. Without taking into account the boron self-shielding, the boron burn-up to be achieved in the HFR (inner or outer positions) should at least be in the range of 90% within one year of irradiation.

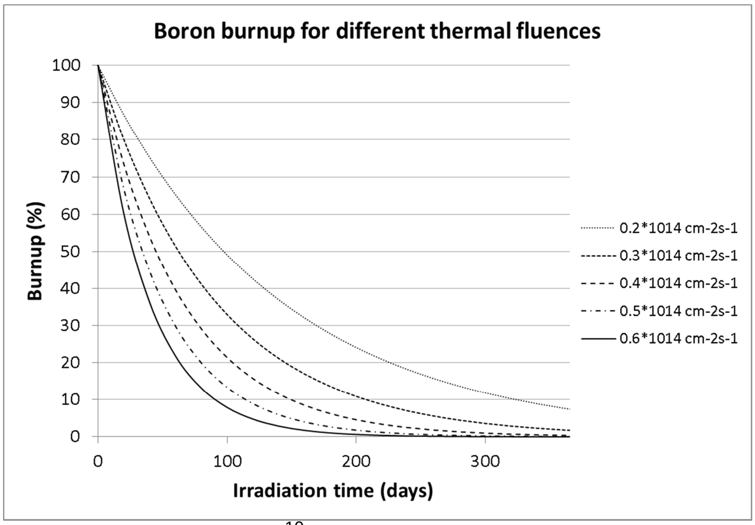

Figure 2-1: Estimated ^{10}B burnup times for different fluences

3 Coated particle fabrication studies

The fabrication of the particles is investigated within the ARCHER project in Work Package 3.1 at TU Dresden and the University of Manchester. The complete report on coating will be made available within the ARCHER project (D.3.1.31 and D.3.1.32).

3.1 Material used for the coating particles

The material used as surrogate kernel for the coating experiment is alumina (Al_2O_3). Alumina was preferred to Zirconia (ZrO_2). From experience, it is known that both zirconia and alumina may be used. However, although the zirconia weight is more representative of the fuel, the activity of the particles after irradiation is significantly higher due to hafnium traces. This complicates the handling for post-irradiation examinations significantly. In order to achieve the sphericity criteria for the kernel fabrication, some specific alumina kernels should be found and used for the final test. The preliminary tests were performed with asymmetric kernels that will generate some extra mechanical constraints that are not compliant with the requirements of the present experiment. The sphericity of the final alumina kernels should be representative of the one encountered for the original fuel kernel during the fabrication process.

3.2 Deposition method

3.2.1 Boron Carbide (B_4C) layer

The boron carbide (B_4C) layer coating is done at TU Dresden using the Pulsed Laser Deposition (PLD) method. Only the major results made available till July 2013will be presented in this report. For the time being, natural boron is used as a target for the laser. For the final particle fabrication, it might be necessary either to use enriched boron or to slightly change the PyC layer to accommodate the thicker boron layer. Nevertheless it is considered that what has been performed during this feasibility study with natural boron carbide will be easily transposed to enriched B_4C.

Different successful series of experiment were performed with the PLD technique over the past year. It appears that the present deposition of boron on alumina kernel was more complex than anticipated due to the low density of the target and the specific spraying effect induced (see difference in Figure 3-1and Figure 3-2).

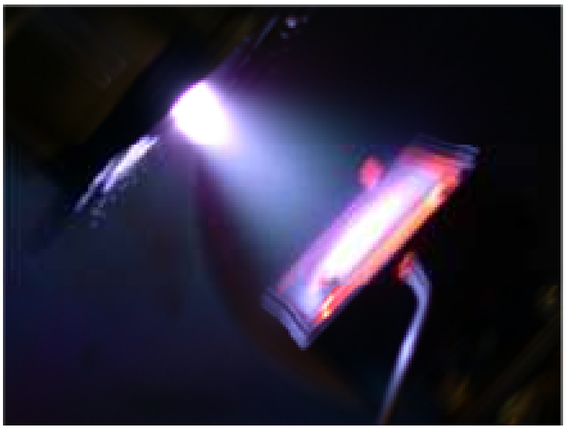

Figure 3-1 : Deposition process using Pulsed Laser Deposition

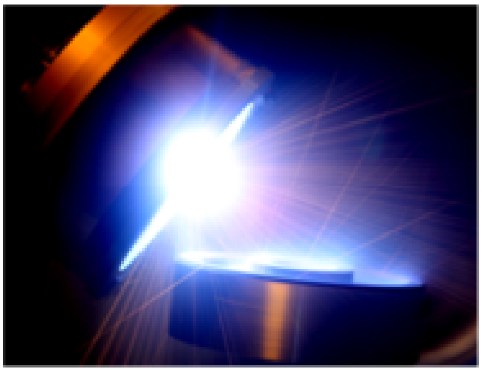

Figure 3-2 : Deposition of the Boron Carbide on the target
(spraying observed due to the low density of the target)

Nevertheless, the first results are encouraging. The deposition process is operated at 400°C and leads to the deposition of small particles on the alumina surface.
Those particles are being analyzed at the University of Manchester (UMAN). So far, the heating treatment exhibits a stability of the layer up to 1600°C. The crystallinity is still to be controlled.
Eleven hours of PLD process were required to achieve a 25 µm deposit on the alumina kernel. The surface of the boron coating is shown on Figure 3-3 and the layer is visible on the SEM image (Figure 3-4). Some new experiments will be carried out in September-October 2013 to reach a higher thickness.

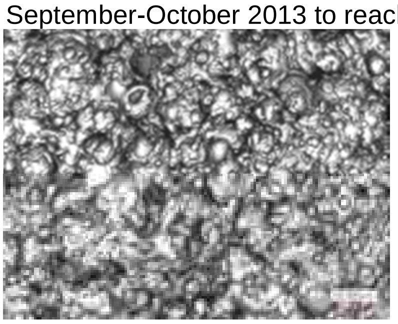

Figure 3-3 : surface of the boron carbide layer

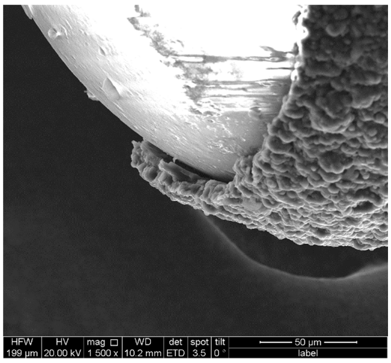

Figure 3-4 : SEM image of the deposit of supposed B_4C

3.2.2 Silver deposition

Silver deposition has also been performed on the particles already. Analysis is on-going and more details on this deposition process are upcoming.

3.3 Discussion on the results

The deposit so far exhibits a certain porosity: this porosity requires a quantification and raises some discussions. The thickness of the B_4C is to be properly assessed to reach the targeted pressure at the end of the irradiation. If the required layer is too thick even with a revised PyC layer that will absorb the extra B4C thickness, some enriched boron should be envisaged to keep the system within the internal dimensions of the SiC layer This point is to be discussed especially considering the conditions required by INL in the common irradiation and is extensively discussed in Appendix. On the other hand, the observed porosity will enhance the release of helium originating from the B_4C layer.

4 Irradiation

4.1 Design

To meet the requirements as presented in chapter 2, the experiment design is based on a similar experiment dubbed PYCASSO 2. The PYCASSO 2 experiment performed to investigate coating behavior of a variety of coated particles with dummy kernels was very much in the scope of the CP-STRESS particles examination, but without any pressurized particles. The experiment was designed for 3 different temperature regimes of 900 °C, 1000 °C and 1100 °C. PYCASSO 2 was irradiated successfully during 8 cycles with a BOI in August 2009.

The design was based on carved discs of a tungsten alloy where the particles were loaded. Due to the high density of the metal, there was enough heat production to reach temperatures in the vicinity of 1200 °C. The particles were placed in hexagonal shaped cavities in the discs, which are stacked and fixed by screws; both are shown in Figure 4-1. A specific number of discs screwed together will in a stack, were maintained at one temperature level. Between the different stacks, heat shields are placed to prevent axial heat flow, isolating the different stack temperatures one from the other. Thermocouples guided within the stack allow an accurate temperature determination. With a particle size of ~1 mm diameter, the thickness of one disc may be as low as 2 mm.

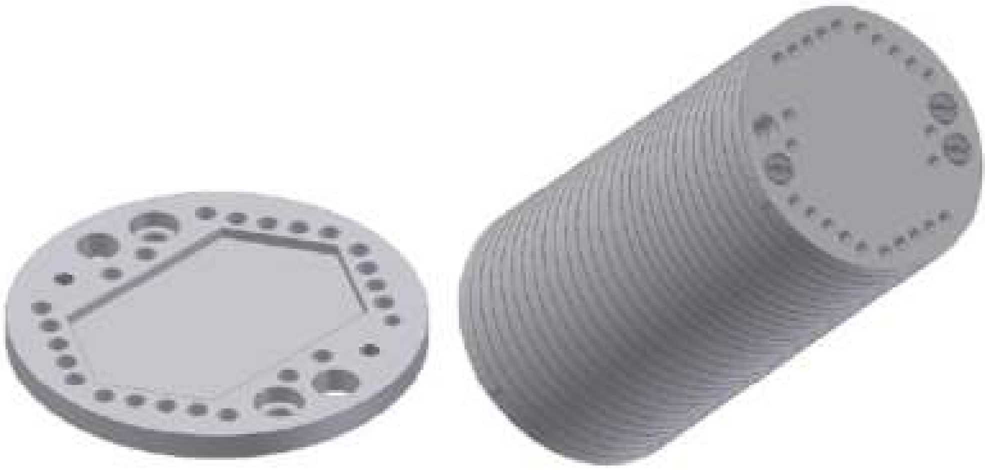

Figure 4-1 : Disc with hexagonal shape on the left, and a stack of discs on the right.

Over the complete set of stacks (Figure 4-2), a tantalum heat shield was placed to act as an extra thermal barrier. The stack with the heat shield is placed in a containment tube which constitutes the sample holder. This sample holder is finally positioned in the HFR irradiation position. Between each layer, a gas gap is present.

The temperature is achieved due to radiation heating. The gamma radiation is the main contributor of the total heating, although the amount of boron can also provide a significant contribution (to be determined), depending on the amount of particles in the experiment and the associated thermal flux.

The primary coolant water acts as a heat sink. Tailor shaped stepped gas-gaps in the axial direction between the containment tube and the irradiation rig allow the temperature control and act as a heat flow barrier. By adjusting the helium/neon gas mixture in these gas gaps, the thermal conductivity of the gas can be controlled, thereby regulating the temperature. In the thermal analyses, thermal conductivity as well as thermal radiation will be taken into account for the calculation of the heat flux through the gaps. Due to the elevated temperature levels the heat flux by thermal radiation also need to be considered.

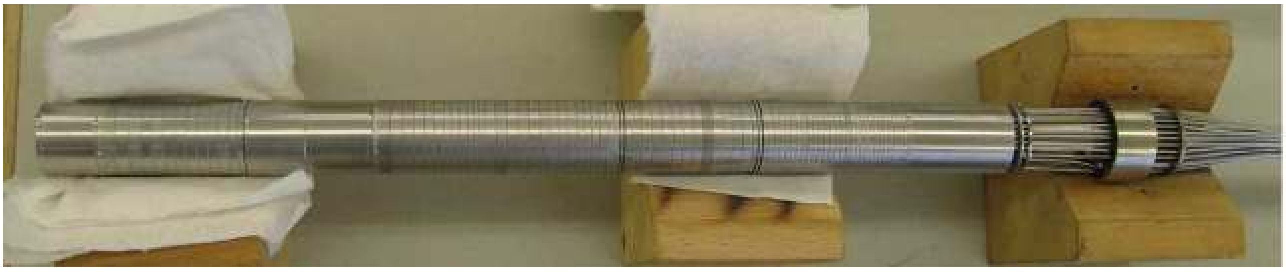

Figure 4-2 : Complete stack of discs (PYCASSO II)

The most critical part in the design of PYCASSO 2 was the containment tube [3]. High stresses may occur in the top and bottom part of the containment tube, as a result of high local temperatures.
The potential parameters that can be adjusted to duplicate the PYCASSO 2 experiment with the CP-STRESS requested operating temperatures will be discussed in this section. The generic goal is to make an optimum use of the practical experience gained during PYCASSO 2, and therefore to slightly tune some parameters in the already known design.

4.2 Number of specimen

To be statistically representative, large numbers of particles are required. Based on the design, and assuming an average particle size of 1 mm, approximately 275 particles can be fitted in each disc as depicted in Figure 5-3. The number of discs in the PYCASSO experiments was 74 per experiment. But since there were some larger particles in PYCASSO that required thicker trays, the number of discs can be increased to roughly 100. This would lead to roughly 27.500 particles in a single experiment.

Figure 4-3 : Disc with particles

4.3 Neutronics

A target fluence of $5{\cdot}10^{25}$ m^{-2} for E>0.18 MeV is foreseen. The fluence level of the position where PYCASSO was irradiated is approximately $4{,}1{\cdot}10^{24}$ m^{-2} per HFR cycle [1], which result in 13 irradiation cycles required, which is approximately 364 full power days based on 28 day cycles.
From the PYCASSO data, the relative flux profile is reconstructed to give the flux (or dpa) distribution over the experiment. The axial positions in the graph exactly cover the height of the experiment, from bottom tray to upper tray. The bottom of the capsule reached a fluence of approximately 70% of the maximum, i.e., if a maximum of $5{\cdot}10^{25}$ m^{-2} is required, the trays on the bottom will reach $3{,}5{\cdot}10^{25}$ m^{-2} . In case of larger difference required, the experiment may be positioned lower and exact additional calculations will be required.
The position that is proposed for CPSTRESS is a mid-way position between the center of the core and the periphery i.e. position G3 as depicted on the Figure 5-4. This is an 'average' position regarding the fast fluence. It is possible to go to positions more towards the center, with higher fast flux and therefore

accelerated dpa built-up. There is a trade-off however, the thermal flux *decreases* when going towards the center of the core. The lower thermal flux will decrease the rate of boron depletion, which needs to be assessed.

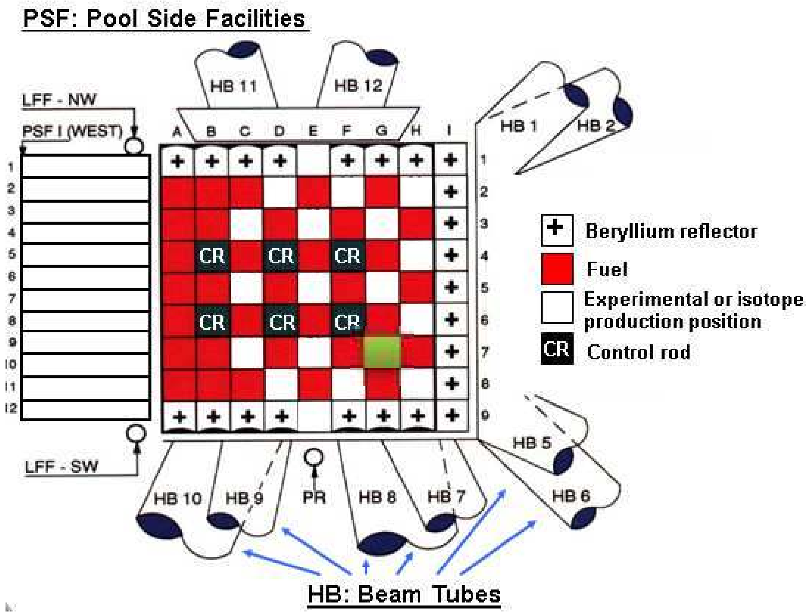

Figure 4-4 : Schematic view of the HFR core with the proposed irradiation position in green

4.4 Thermo mechanical review

A thermo-mechanical review [2] was performed to assess the feasibility of the higher temperatures compared to the case of PYCASSO 2. The following parameters can be changed to obtain a higher operating temperature for the experiment

- a higher flux position in the core;
- a different orientation (within the position, PYCASSO was facing the periphery, a core orientation will result in a higher flux);
- a different gas mixture to achieve less heat conductivity (e.g. less He, more Ne);
- an increase in the gas gaps width in the experiment;
- a different ratio between Densimet trays and sample materials, in order to achieve a higher average density (and therefore a higher gamma heating).

To make an optimum use of the practical experience gain during PYCASSO 2, only slight adjustments will be considered either in the experiment or in its environment.

The thermo-mechanical assessment is performed based on experience of PYCASSO 2, which was designed for irradiation temperatures of 900, 1000 and 1100 °C. In order to achieve the desired temperature profile, the width of each gas-gap was tailored. The inner gap (between the trays and the tantalum heat shield) varied from 150 to 500 µm. The second inner gap (between the heat shield and containment tube) varied from 100 to 300 µm. The outer gap (between the containment tube and the TRIO channel) varied from 210 to 250 µm. A gap width of 500 m is normally considered as maximum, so the gaps can either be increased or reduced in a next experiment, if needed.
Thermo-mechanical analyses were performed for PYCASSO 2 [4]. In the calculated design case, the gas for the inner gaps was a mixture of helium and neon, with some margin to compensate for lower and higher fluxes. In the design case the high temperature region becomes 1100 °C. As a safety case, also calculations were performed with 100% Ne in all the gas gaps. In that case, the high temperature region becomes approximately 1350 °C. So the targeted 1200 °C for CPSTRESS should be achieved when considering an enriched Neon gas mixture. Radial heat flow, axial heat flow and heat radiation due to the high temperatures, were all taken into account in these thermal calculations.
From the experiment logbook in DACOS, it can be deduced that the PYCASSO designed temperatures and gas mixtures being achieved [5], the targeted 1200 °C temperature area can be also achieved.

4.4.1 Stresses in the containment tube

The most critical part in the design of PYCASSO 2 was the containment tube. High stresses occur as a result of high local temperatures. The occurring stresses and temperatures were calculated for the top and bottom part of the containment tube of PYCASSO 2 [4]. In all cases the values were found to be acceptable, except for the case where 100% Ne was applied in all gaps. In that case the temperature of the containment reaches a maximum of 643 °C, σ_{max} = 295 MPa and σ_{allow} = 144 MPa, resulting in a safety factor of 0.49. This problem was solved by applying a scram of the reactor, triggered by some of the thermocouples [3]. In the CP-STRESS experiment this may also be solved with a scram induced by thermocouples, nevertheless the higher operating temperatures will make the safety margins be smaller.
Another approach is to further optimize the design of the containment bottom part, in order to achieve lower stresses and a higher safety margin. It may also be possible to reduce the temperature field in the bottom part by increasing the distance between the containment bottom and the lowest stack of trays (being the nearest heat source).
If the lowest stack of trays in the new experiment will also be representative of the 900 °C encountered in PYCASSO, then redesigning the bottom part will most probably not be necessary.

4.4.2 Conclusion of the thermo-mechanical analysis

From the thermo-mechanical analyses that were performed for PYCASSO 2, and the practical experience with PYCASSO 2, it can be concluded that an irradiation with different temperature field in the range 1200 °C - 900 °C is possible. Depending on the exact requirements, the drums can be tailored to achieve the temperatures.

There are some additional considerations, which should be discussed in a detailed final design. Some of these considerations are described below:

- If needed, larger heat shields or larger dummy blocks can be applied in order to separate the 1200°C from the 900°C regions. In that case less room is available for trays with target material.
- the design of the bottom part of the containment tube can be optimized further in order to reduce occurring stresses.
- The influence of the ^{10}B reaction on the fluence rates and the heating in the targets should be further investigated. This will likely claim some of the control margin, since the heating will decrease after boron burn-up.

4.5 Instrumentation

The instrumentation of the sample holder consists of 24 thermocouples and high temperature neutron fluence detectors. The thermocouples are spread over the height of the experiment, and also over the radius. By choosing the right distribution any radial and axial temperature gradients will be observed.

Neutron fluence will be monitored by activation monitor sets. In each stack a monitor set will be placed to determine the fluence after the irradiation. The sets are developed to cope with the high temperatures from the CPSTRESS experiment.

5 PIE

Post-irradiation examinations on the CPSTRESS experiments are crucial to scrutinize the results and to better understand the behavior of the particles during irradiation: actual temperature reached by the particles, fractured particles observations, behavior of the different layer of the particles during the experiment. This chapter sums up some destructive and non-destructive PIE processes that can be performed in the Hot Cell Laboratory at NRG. This non-exhaustive list can be considered for further discussion.

5.1 Visual inspection

One of the targets of the visual inspection is to verify the failure rate. This will be verified by visual inspection of all the particles. Based on the coating failure, failed particles will be separated and counted for each temperature regime and a failure rate will be determined.

5.2 X-Ray Tomography (XRT)

X-Ray Tomography is a powerful tool, available at NRG, to non-destructively inspect all the coating layers of the irradiated particles. Shrinkage of the buffer layer and swelling of the SiC layer will be assessed. Using XRT enables to determine the free volume of the particles (porosity of the buffer layer). The value of this volume is required to verify the internal pressure reached in the particles.

5.3 Internal pressure verification

With the known volume from the XRT measurements, the amount of helium inside the particle needs to be determined. This can be achieved by crushing the sample and measuring the amount of He released.

Since the amounts are small, this could be done best by crushing the particle in a known amount of another gas, Neon for example. By measuring the He/Ne ratio, the amount of helium can be determined and the inner pressure reached during irradiation assessed.

5.4 Heating test

Heating tests can be performed to find the failure temperature of particles that not failed during the irradiation. Particles will be heated well beyond 1600 °C to find the high temperature failure fraction. During heating, the release helium from the particles will be picked-up by another inert gas (Ne/Ar) to detect the time and temperature of failure.

5.5 Microscopy

Optical and Scanning Electron Microscope analysis can be performed to study the state of the SiC and PyC layer after the irradiation. To investigate changes in grain structure, Electron Back Scatter Diffraction measurements can be performed to show possible grain elongation or strain.

6 Conclusion

A study was performed to assess the feasibility of the CPSTRESS irradiation. This experiment focusses on irradiation surrogate coated particles with a boron carbide layer in the kernel. The helium that is produced by the neutron irradiation of boron will pressurize the particles, simulating the internal pressure of regular TRISO particles without the chemical interaction.

From prior studies, mainly the thermo-mechanical analysis performed on the PYCASSO-2 irradiation and the experience gained from the irradiation, it can be concluded that a new irradiation at higher temperature (1200°C) is technically feasible from an engineering point of view. Minor adaptations on the design of the previous irradiation should be taken into account to optimize the new irradiation and to target the requirement of the new experience.

- Fluence/dpa

The dpa is mainly dependent fromthe flux profile in the HFR. Due to the flux buckling, it is possible to achieve a flux within a range of $3{,}5 - 5 \cdot 10^{25}$ m^{-2} for the 13 expected HFR cycles.

- Temperatures – can be tailored

The temperatures can be tailored to fit the specifications. The post analysis of PYCASSO irradiations led to the conclusion that the 1200 C temperature level could be easily achieved w enough insulation material between the different temperature regimes (i.e. heat shields of dummy blocks need to be placed between the stacks).

- Internal pressures

With an irradiation position close to the HFR periphery the boron burn-up is expected to reach up to 95% within 300 full power days. Studies on B_4C coating fabrication show that layer thicknesses up to 20 micron are to be achieved, only the porosity achieved during the PLD process still needs to be assessed..

At this point, no issues are identified that cannot be solved and would be a threat for the expected experiment. However, some items still need to be further studied in order to initiate a final design calculation prior to an irradiation.

7 Objective and development of CP-STRESS

The present document is a draft: Indeed, as mentioned in the description of work of ARCHER, a collaboration with INL is foreseen as a further step for an irradiation proposal. First discussions with INL in the last quarter of 2012 were extremely successful. Consequently, INL together with NRG has submitted an irradiation proposal to the Department of Energy of the United States.

Therefore as the scope of work has slightly changed and is now also driven by INL interests, a more complete feasibility study is expected which will go beyond the deliverable discussed in ARCHER. This report presented some preliminary analysis concerning the CP-STRESS experiment and should be amended with a more in depth document to come. In the appendix, a first scoping of irradiation conditions calculated with the PARFUME code is presented.

8 References

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[3] v.Kranenburg M.A.C.: "PYCASSO-02 (370-02)", Design and Safety Report, NRG-25138/09.94941 rev.D, dated 12 August 2013

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[5] HFR Operations: Results in DACOS, experiment 370-02, logbook and temperature registrations

9 Appendix

Scoping Calculations for the CP-Stress Experiment
Idaho National Laboratory
October 14, 2013

Initial calculations have been performed to explore what particle parameters and irradiation conditions are needed to support the CP-Stress experiment. These calculations sought a combination of parameters that would ultimately result in the SiC layers experiencing a stress of about 350 MPa. Past experience with PARFUME calculations have indicated that SiC stresses in this range are required to induce particle failures. The parameters investigated in this study directly affect the particle internal pressure, and hence, the SiC stress. These parameters included the irradiation time and temperature, buffer layer thickness (void volume), initial boron-10 concentration (amount of helium that can be produced), and effective helium release fraction (amount of helium contributing to the internal pressure). Internal particle pressures were calculated assuming a nominal particle configuration, as listed in Table 1, while the parameter of interest was varied. The results of the calculations are discussed below.

Table 1. Nominal particle configuration and irradiation conditions

Parameter	**Value**
Kernel diameter (µm)	500
B_4C layer thickness (µm)	15
B_4C density (g/cm^3)	2.52
Boron-10 enrichment	Natural (19.9 wt%)
Buffer thickness (µm)	95
Buffer void fraction	0.5
Irradiation temperature (°C)	1200
$^{10}B(n,\alpha)^7Li$ cross section (barns)	3837
Neutron flux (10^{18} m^{-2} s^{-1})	0.69

The effect of irradiation temperature for a nominal particle, assuming an unrealistically high 100% effective release of the helium (all of the helium that is produced enters the buffer void volume), is shown in Figure 1. These calculations indicate that even beyond the maximum experiment design temperature of 1200 °C, significantly high internal pressures cannot be achieved with the nominal particle attributes. The results do show that saturation in helium production occurs around 200 effective full power days.

In order to achieve higher internal pressures, the boron-10 enrichment in the B_4C layer will likely have to be increased above natural levels. Figure 2 displays the internal pressures for a nominal particle as a function of boron-10 enrichment after 250 effective full power days of irradiation assuming two levels of effective helium release. Again, assuming an unrealistically high 100% helium release, a boron-10 enrichment near or above 75 % would be needed to achieve a pressure greater than 350 MPa. For the more probable 50% effective helium release (half of the helium produced enters the buffer void volume while the other half is implanted into the kernel and becomes trapped), internal pressures remain too low (below 250 MPa) even with 100% boron-10 enrichment.

To achieve internal pressures greater than 350 MPa while assuming a realistic 50% effective helium release fraction, the buffer thickness needs to be reduced from the nominal value in addition to using enriched boron-10 in the B_4C layer. Figure 3 displays internal particle pressures after 250 effective full power days of irradiation assuming 50% helium release for two extremes of boron enrichment. For 100% boron-10 enrichment, the buffer layer needs to be less than 70 µm thick to achieve pressures greater than 350 MPa. With decreasing boron-10 enrichment, the buffer will need to be even thinner.

As the experiment design evolves, these calculations will also be refined. However, it seems clear that a combination of highly enriched boron-10 in the B_4C layer and a reduced buffer thickness will be required to induce meaningful levels of particle failures.

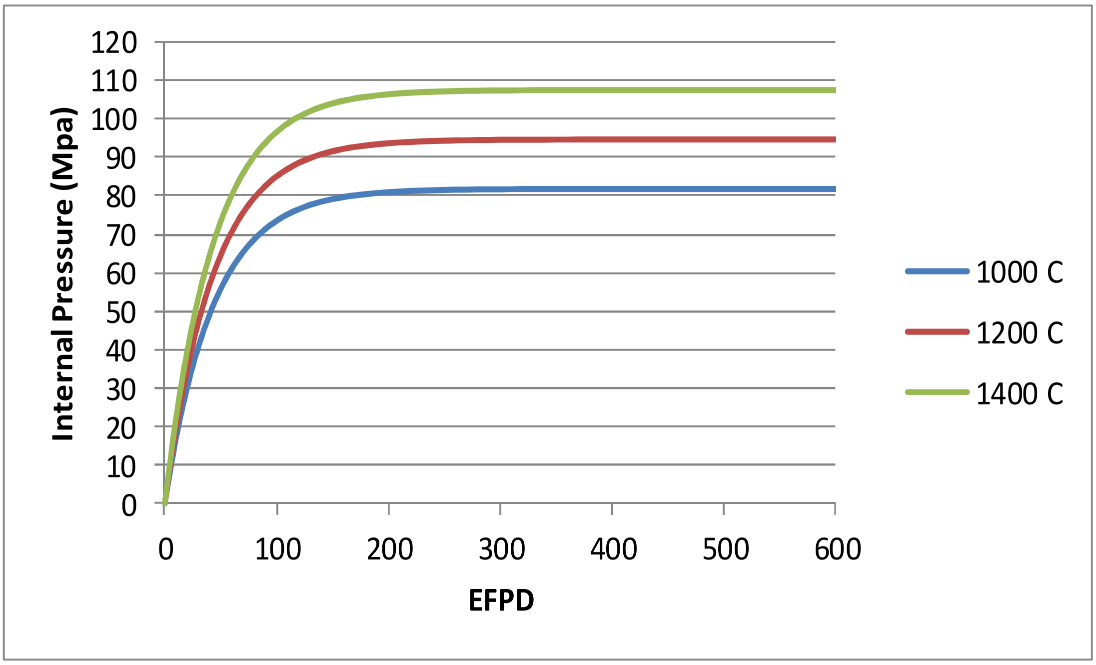

Figure 1. Nominal particle assuming 100% effective helium release.

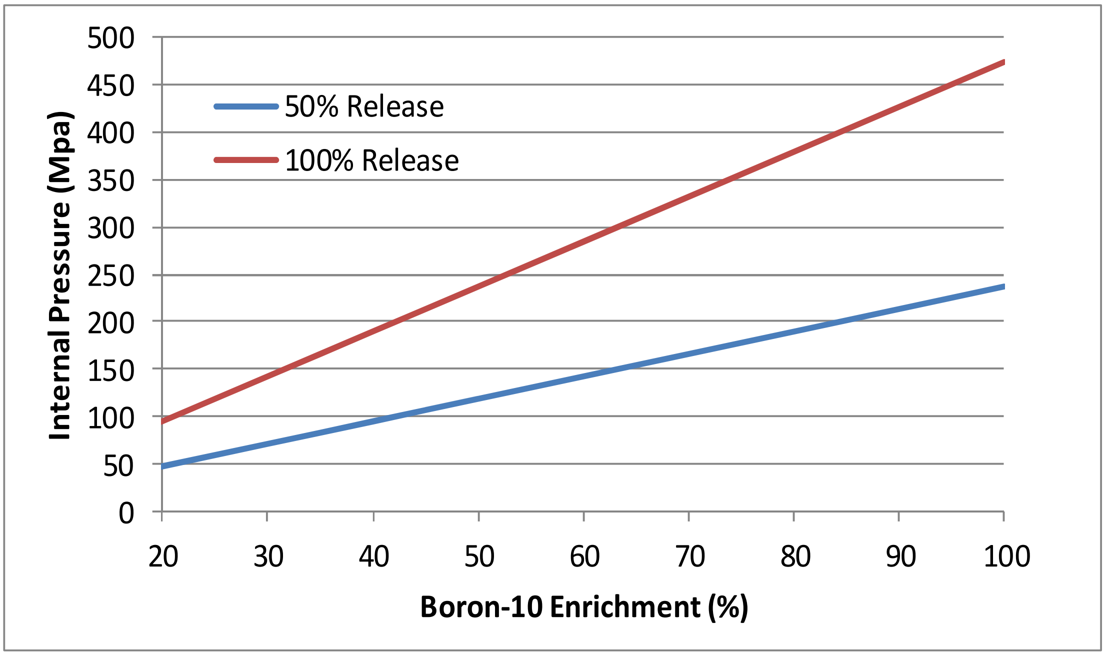

Figure 2. Nominal particle after 250 effective full power days of irradiation.

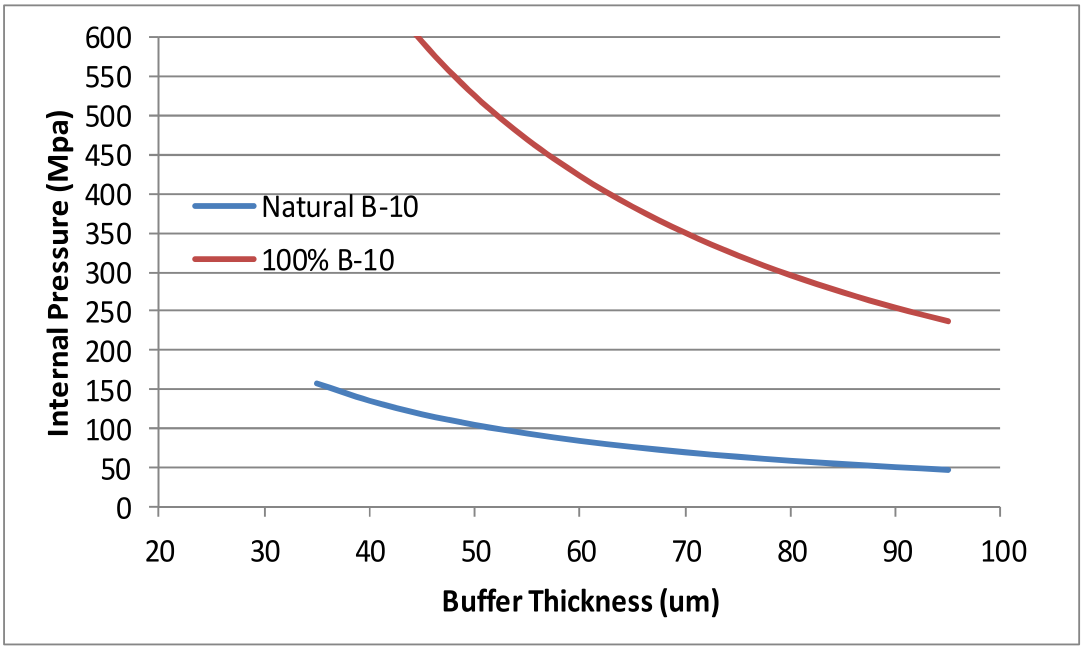

Figure 3. Nominal particle after 250 effective full power days of irradiation.