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Continuity of High Quality Research and Medical Isotope Production in Petten

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1. Introduction

The High Flux Reactor, HFR, in Petten is designed and built in the fifties of the 20th century for science and engineering purposes [1]. Full operation started in 1962 with experiments and isotope production in the core and neutron diffraction experiments around the pool.
The HFR experiments contribute to the development of nuclear fuel elements, in particular high temperature reactor pebble fuel development and testing. In addition structural materials for light water reactors, high temperature reactors and fast sodium cooled reactors are investigated. The poolside facility is functional for fuel ramping and loss of coolant tests. More recently the research on fusion materials and components such as helium and lead cooled blankets has developed.
The HFR produces with the NRU reactor in Canada the majority of the medical isotopes used for millions of treatments in the world today. Besides large production volume of for example Mo isotopes, the HFR has new developments underway such as the production of Lutetium.
The horizontal beam tubes serve a wide range of scientific micro-structural investigations of a wide range of materials. As an example: the evolution of residual stress patterns in welds are studied with neutron diffraction. The neutron beams have also applications in the medical domain for example boron neutron capture therapy development BNCT.
At present the HFR is owned for a lease of 99 years by the Joint Research Centre of the European Commission [2]. The HFR has been operated the past decennia under Supplementary Programs of the European Commission. NRG holds since several years the operation license under the Netherlands Nuclear Energy Law. NRG operates and maintains the plant under contract, and manages the commercial activities around the reactor since 2002.
The successes of the HFR research and development programs depend for a large part on the Hot Cell and Isotope Laboratories, located in sight of the reactor. Their experienced personnel and

regularly updated infrastructure, and equipment contribute greatly to the results produced by the HFR [3].

2.Major HFR refurbishments and upgrades

Reactor vessel renewal

The reactor is of the tank-in-pool type using an aluminum alloy reactor vessel with neutron beam guide tubes. The vessel surveillance program measures mechanical properties of vessel samples irradiated in the core. The level of embrittlement of the material samples is the indicator for the integrity of the reactor vessel, which is mainly affected by thermal neutron radiation increasing the silicon content [4]. On the basis of the surveillance program results the vessel was replaced in 1984. The alloy composition was adjusted, but of course also the second vessel develops an increasing silicon content leading to embrittlement. The present surveillance program uses fracture mechanics results to predict the allowable defect in the irradiated material. The yearly in-service inspection measures the defect dimensions in the vessel to monitor the integrity. The most recent inspection and surveillance results provide indications that the vessel life will last beyond 2015.

Conversion to Low enriched Uranium Fuel

In the latter part of the 20 century the international research reactor community was convinced that the non-proliferation was served by using Low Enriched Uranium, LEU, with less than 20% fissile material. The deliberate preparations for the HFR conversion included qualification of the new LEU fuel elements for the HFR. The actual conversion from Highly Enriched Uranium to Low Enriched Uranium (<20%) fuel elements took about one year. The conversion was successfully completed in 2006 without any reduction in full power days during the process [5]. Though some change in neutron spectrum locally could be observed in the LEU core. Some position re-arrangement of the isotope and experimental devices within the core has effectively allowed the irradiation program to continue as before.

Safety and Ageing

The reactor vessel is not the only ageing component of the HFR. Heat exchangers and piping have been renewed as well to avoid malfunction. The non-destructive inspections in regular maintenance outages are invaluable for the surveillance of the integrity of the major HFR components.

During the summer maintenance stop 2008 small gas bubbles traces were observed in the bottom plug liner pointing at corrosive processes affecting the liner from the concrete side. After in depth investigation and analyses of nearly half a year it was decided to prepare a major repair operation in spring 2010.

The unforeseen outage caused by the bottom plug liner issue is another indicator for the importance of the HFR isotope production capacity in the world. The expectations are that the HFR has a technical life beyond 2015, but the risk of another major repair cannot be excluded. The same holds for the other ageing research reactors.

4.Status of HFR in Research, Development, and isotope production

The HFR program objectives for the next decade cover quite different fields. The programs on fission and fusion energy have in common the safe production of energy with the minimization of the environmental effects. The isotope production aims for the improvement of the health conditions, whereas the utilization of neutron beams contributes both to improved health care and the knowledge of the microstructure of materials

Fission energy

The AMES project continues the JRC coordinated projects AMES and NESC. In LYRA-rigs specimens are subjected to irradiation conditions relevant for water cooled power plants. Recent experiments use compositions of WWER-1000 and PWR reactor pressure vessel materials with systematic variation of the chemical composition. The focus is on the effects of neutron radiation on the fracture properties of pressure vessels of water cooled reactors. Another highly relevant subject for plant life extension is the development of a loop to be used in the HFR for irradiation assisted stress corrosion crack testing.

The work on innovative reactors in the HFR concentrates on high temperature reactor, HTR, applications for both the electricity production and high temperature process heat. Fuel particles, used for HTR fuel elements, and high temperature structural materials development are part of the experiments conducted in the HFR core. Much of the work is carried out in the high temperature reactor technology network – HTR-TN operated by JRC-IE currently with 21 partners. New grades of graphite are under investigation. In the project RAPHAEL, NRG plays a leading role in the research on graphite behavior for high-temperature reactors. Rigs have been developed for tests up to 24 dpa, figure 1, and are used intensively for temperatures up to 1000 ^{o} C [6]. The non linear behavior needs to be quantified accurately for design of HTR's cores.

The reduction of the mass and radio toxicity of nuclear waste is an important issue for the safe generation of nuclear energy. Typical examples of projects addressing that issue are: CONFIRM, and HELIOS. The CONFIRM irradiation will tests two plutonium fuel pins with 30% plutonium nitride in a zirconium-nitride inert matrix; $(Pu_{0.3},Zr_{0.7})N$, fabricated at the Paul Scherrer Institute in Switzerland. To simulate fast reactor neutron conditions the test material is shielded from thermal neutrons applying a hafnium. The inert matrix stability will be tested in this way using PIE. The HELIOS project is as part of the FP6 EUROTRANS Integrated Project. Partitioning and Transmutation. The main objective is the study the behavior of the actinides in uranium free fuel in ceramics and cermet microstructures such as CerCer $(Pu, Am, Zr)O_2$ and $Am_2Zr_2O_7$+MgO or CerMet $(Pu, Am)O_2$ +Mo. The effect of microstructure and temperature on gas release and swelling on the fuel properties will be measured, to understand to potential of these fuel types for the reduction radioactivity in waste.

Fusion energy development

The operation of fusion power plants will take decennia of materials and component development. The first step towards fusion power technology is the building and operation of ITER. The HFR

Petten has contributed generously to the materials development for the ITER vacuum vessel in research and development of low heat input welding technologies for austenitic steels to be used for ITER. Steel irradiated in the HFR has been welded to study the effects of helium generated in the steel during irradiation, leading to limits of allowable helium contents in steels to be welded in irradiated condition. The results of the determination of the radiation effects on the properties of plate and welds have been used for the Materials Handbook for the design and safety analyses of ITER.

The functional materials development for tritium generation in blankets has continued for a long time. The work has presently two program branches:

- one for the application in the testing blanket modules, TBM, for ITER, figure 2, and
- the development of advanced lithium and ceramics and lithium lead properties relevant for the design of a demonstration fusion power plant, DEMO, after ITER.

The EXOTIC (EXtraction Of Tritium In Ceramics) test series running in the HFR focuses presently on the in-pile tritium release characteristics of ceramics [7]. Parallel to the functional material tests for the ITER TBM the effect of in-situ oxidation on the permeability of Eurofer-97 has been experimentally simulated with moisture purge gas.

The HIDOBE (High Dose Beryllium irradiation) project series in the HFR has the goal to measure the long-term behavior of beryllium pebbles used in the blankets for neutron multiplication. The determination of neutron radiation effects on swelling, creep and tritium retention leads to verification of models used by designers of blankets. This work has along term character aiming for end of test helium contents in the beryllium of 5000 appm. The IEA implementing agreement on Radiation Damage Effects in Fusion Materials has a strong co-ordinating function. Partners in the experiments are several institutes in the EU, Japan and the Russian Federation, providing different grades of beryllium specimens for the HFR in-pile experiments.

The HFR testing for ITER is not limited to testing materials properties. Highly integrated experiments demonstrated the feasibility of design concepts for blankets through in-pile testing of test blanket sub-assemblies with all the ingredients for potential interaction of steel, lithium ceramic pebbles, beryllium pebbles, helium, and tritium. Such in-pile simulations provide designers with verification data of their models for the blanket behavior. Such results can also become a signal for unforeseen interactions in the blanket operation leading to design and layout improvements.

Isotope Production

The HFR produces on the average over 30 % of Mo isotopes demand in the world. About five reactors contribute with communication on their planned outages to the world production. That this is a small number of producers is exemplified by the fact that the HFR had to operate the only major Isotope Production Reactor in Europe for a record level of more than 100 days during 2007. The majority of the production at present is Mo, table 1. New potential isotopes, such as lutetium are produced in the HFR to prepare for the next generations of treatments and diagnostics, and more are in the pipeline. It must be admitted that some older therapy products

have been discontinued from the production list. These reductions are offset by the development prospects for the new generation of isotopes.
Besides the production of isotopes for medical use technical applications of isotope production grow inn importance. The use of semiconductors in power applications has boosted the demand for silicon doping. The natural ^{30}Si isotope exposed to thermal neutrons catches a neutron and consequently transmutes into stable ^{31}P. Increasing phosphorus content can reduces the resistance of the silicon to the electrical properties needed for the application. The usage of phosphorus doped silicon in automotive components for hybrid cars will therefore be on the rise for the forseeable future.

Beam applications
Originally the neutron beams were intensively used for diffraction to resolve complex microstructures. The micro-structural application is still continued in the HFR, though specialized research reactors, such as FRM-2 in Munich and ILL in Grenoble with very high quality beams, including higher strength, now have taken over parts of the area. Important specialties where the HFR is most instrumental today are the measurement of diffraction patterns in and around welds. The method allows the precise determination of residual stresses in the welded construction, following the thermal and mechanical distortion of the weld heat input. JRC-IE Petten hosted recently the specialists in this area for an international workshop on the subject in the frame of the Network on neutron techniques standardization for structural integrity, NET [2].
Small Angle Neutron Scattering, SANS is another method applied in the HFR which has more ties with the earlier micro-structural beam usage.
JRC-IE uses thermal neutron beams clinical trials for boron neutron capture therapy. The thermal neutrons are used to transmute ^{10}B into helium and lithium particles which energy destroys the cell they are formed in. The first series of brain treatments has been concluded. After renewal of the license for the trials the next research program will concentrate on the treatment of glioblastoma and head and neck cancers. Ex corporal treatment of liver metastases might be taken up later as well. It is not the intention to use the HFR for regular treatment purposes. The programs are set up for clinical trials. It is expected that when successful treatments have been established. Much smaller reactor can create neutron beams with the right specification allowing treatment near large (academic) hospitals.

5.Programs for the first half of the 21st Century

Research organization
Pre-competitive research co-operation will remain mainly centered around scientific and high tech developers organizations that share work on test matrixes. The co-operation can be bi-lateral and multi-lateral. Usually the results are shared along guidelines and delivery definitions before the projects start. Such type of research is with closed purses with in-kind contributions of all partners. The results will usually be published in open literature, with combined authorship. The effectiveness of the knowledge dissemination can be measured with the quantities of open publications and their citation indexes.

In the European Union Framework programs presently many parties work together to reach goals that require co-ordination of different scientific and technological knowledge domains and laboratories. It is expected that this type of research organization will continue along similar lines for decades. The indicators for success of the Framework programs can be measured with the results with the deliverables defined in the project contracts as yardsticks. In many occasions co-ordinated research efforts are published in open literature where number and quality in the form of quotation can serve as primary instrument for effective determination

Contract research will contractually affirm which part of the results will be disclosed and what will remain confidential property of the commissioner. PALLAS will make possible the complete chain of research from project design to final reports. Parts of the chain can also be ordered from design of capsules to post-irradiation testing in isolation. In several contracts the opportunity it will be left open to report with the commissioner in open literature. Then the result will be measurable.

The use of Pallas for R&D purposes will be contained in international programs, as is presently the case. At present the energy and environment R&D is performed under the umbrella of Euratom, which launches framework program within the nuclear R&D. NRG participates - with the support of the Dutch ministry of Economic Affairs - actively in these framework programs, and performs together with European partners strong irradiation programs. Specific programs are performed for fusion R&D within the Fusion for Energy (F4E) and European Fusion Development Agreement (EFDA) framework.

Within these programs long-term relationships have been developed with many partners. Some of these relationship are already active for 40 years. Examples for the domains of fusion and Generation-4 fission energy are given as an example in table 2.

Research Programs

The Nuclear R & D strategy in the EU, and their major instruments for the first half of the 21st century have recently been addressed in [8]. As major research reactors are identified the Réacteur Jules Horowitz, PALLAS, MYRRHA, a fast demonstration reactor, and, others, figure 3. These tools are needed for the advancement of fission and fusion energy and the production of isotopes for medical diagnoses and therapy foreseen for the first half of the 21st century.

Safety is the most important issue in extending the lifetime for existing light water reactors, which lives will be well extended up to at least 60 years end-of-life, thus deep into this century. Based on the latest capacity building forecasts it is expected that in the first half of the 21st century, the nuclear fraction of power generation will double or triple. The safety features for new reactor designs require experimental verification in research reactors, offering a safe testing environment. The Generation 4 class of reactors will be in the centre of the development field. Presently the focus is on the HTR development requiring tests of materials and components of new materials. Those new materials such as carbon fiber reinforced graphite and silicon carbide composites might be used for components such as control devices and advanced fuel elements in the core of the HTR producing high temperature process heat. These materials may also become important for fusion components such as divertors and high temperature blankets with silicon carbide as structural materials.

The development of a safe "closed fuel cycle" is another area, which contributes to the projects needed to ensure the delivery of shorter-lived waste and safe storage solutions.

For the development of fusion energy the development of new low activation materials is mandatory to keep the environmental impact of the fusion power plants to a minimum. This statement is true for functional and structural materials. At present the ferritic, martensitic steels have become the reference structural steel for blankets [9]. In those steels the impurity control has the potency to allow re-processing after less than 100 years. This steel class has limitations in operating temperature and radiation resistance. Those properties might be improved in the oxide dispersion strengthened ferritic steels that could extend the life of blankets in fusion power plants, figure 4. SiCSiC composite structures show good potential for blanket devices beyond DEMO, but these materials need lots of development time, and component testing before wide application is ascertained. Also in tungsten developments are needed to increaser the toughness and radiation resistance.

In the EU the integrated Project "EXTREMAT" stimulates the synergy in the development of advanced fission and fusion materials. Major sub-objectives of this project are the generation of science and engineering knowledge that can be applied by the industry:

- develop materials with improved resistance of materials to environments with high chemical reactivity, and cyclic mechanical loads at high temperatures [10]
- increased lives of materials under high heat flux and thermal shock conditions at high temperatures
- provide materials with neutron radiation resistance factors higher than the present limits up to damage levels over 200 displacement per annum, dpa.
- Invent process schedules that allow that result in materials with graded properties and heterogeneous composition to promote the application in extreme environments.

The EXTREMAT program has set in motion a development path that will take decennia to complete. The use of research reactors in the first half of this century will be indispensable as test beds for the materials development and the verification of design of advanced components in the first half of this century. High neutron fluxes and test rigs allowing to simulate extreme environments are part of the development them selves.

In the coming decade's nuclear medicine will become even more important for staging, monitoring and treatment of cancer. It will be used to determine the unique molecular, metabolic and functional make-up of the diseased organism and help in the formulation of individual specific care. The scientific and quality-of-life improvements will be of large societal importance. For BNCT application it is expected in the first half of this century that specialized small scale dedicated medical neutron beam devices near a university hospitals could be a good solution for the application of neutrons for patient treatments.

6.Requirements for the HFR successor.

Upgrading the HFR for the expected programs and with standards needed for the middle of this century, would take many years of isotope production and research results loss. The discontinuity arising from such a major overhaul would be very capital intensive, also in comparison with building a new reactor. Still there would remain some limitations in the upgraded HFR, because of the existing structure and layout. The replacement of the HFR, by a new research reactor named PALLAS, is the most feasible solution for the continuation of the safe and reliable nuclear infrastructure in Petten.

The requirements for PALLAS, the HFR successor, are derived from the trends expected in the first half of the century in research, development and isotope production in the EU and the world. Of course the Réacteur Jules Horowitz, RJH, will be the major platform for testing in the EU. The PALLAS reactor will continue to be the major isotope supplier in the EU and for the world. There will be agreements on the back-up function of RJH for the isotope supply by PALLAS.

The nuclear infrastructure in Petten, and its expertise concentrated in JRC-IE, COVIDIEN and NRG attracting many other parties will make possible the intense use of PALLAS for experimental investigations for energy and medical applications.

The tender process for the conceptual design of PALLAS started in September 2007, preceded with vendor qualification, in order to have the reactor operational in 2016. Major requirements for PALLAS are:

- Peak fast neutron flux about twice the value of the HFR with ample space for test rigs and isotope production and a peak thermal neutron flux two to three times the HFR value
- Compact core with Beryllium reflectors to economize on the fissile and reflector material and with sufficient space for loops and components for accelerated testing and isotope production.
- Tank-in-pool type for simple reliable handling of experiments and isotope production.
- The reactor power interval should be flexible within the boundaries of 30 MW to 80 MW maximum to optimize utilization in line with the cyclic demands for irradiation services.
- At least 300 full power days per year to increase the flow of scientific experiments, provide a regular supply of isotopes and allow shorter experimental sequences.
- The residual reactor heat would not be wasted, but would be used by several enterprises as low temperature process heat in the local community.

In the design of PALLAS, NRG has not opted for neutron beams, because these would add significantly to the build- and operating costs. There is sufficient high beam capacity, so the neutron beams would not have been competitive with the features of dedicated reactors with high intensity beams such as the FRM-II in Munich and the ILL in Grenoble. The reactor of the Technical University in Delft provides facilities in The Netherlands for research that requires less intensive neutron beams.

The scientific, economic and societal relevance, goals and scope have been used for the definition of the requirements. Besides the technical specification for the design and building, the need of a sound financial operating regime has been included in the operational requirements. The balancing mechanism of capital and operation cost is built into the requirements and award criteria of the tender.

7. The design and construction of PALLAS.

The tenders have produced conceptual designs for PALLAS on the basis of the NRG requirements in line with the IAEA regulations and Netherlands licensing authority rules, including expected additions such as the ability of the containment to withstand a fighter plane impact. The tender has concluded in September 2009. NRG has set the first step in September 2009 for the Environmental Assessment issuing the "Startnotitie" as required by the Netherlands authorities. The site selection has two options:

- one near the hot cell laboratories on the Petten location,
- the other is near the Borssele power plant and the COVRA interim waste storage organization.

These options will be analyzed with respect to their environmental and socio – economic impact. The pre-design developed in parallel to Environmental Assessment must lead to the completion of the pre-design in early 2011. The building license should become available in 2012, immediately followed by the start of the building phase. Hot commissioning is expected in 2016 followed by the full power operation of PALLAS, and the preparation of the HFR decommissioning in 2017. This procedure assures a smooth continuation of the nuclear infrastructure in Petten with strong contributions to the 21^{st} century societal demands for the security of medical isotope and energy supply.

8.Conclusions

1. The HFR is ageing just as its contemporary research reactors. Nevertheless it will play a crucial role in the supply chain of isotopes, and research until its successor becomes fully operational
2. The responsibility the Petten site feels for the continuity in supply of products for health care are supported by the future projections for isotope demands.
3. The development of the Generation-4 fission reactors and fusion power plants are indicated on the roadmap for research, and infrastructures in the European Union, and elsewhere.
4.Until the middle of this century fission reactors will remain important for fusion development. Nearing the middle of the century, dedicated fusion devices will come available for fusion component testing reducing the work for fission research reactor for fusion.
5. Replacement of the HFR is necessary to comply with the 21^{st} century requirements for the isotope production, and materials and component research in support of the nuclear power renaissance.
6. The main PALLAS requirements are:
    - neutron fluxes double those of the present HFR
    - flexible core reducing fuel cost and waste production
    - more mechanization in operation, with double hot-cells
7. The EC prescribed tender process has lead to the selection of the best conceptual design in September 2009, lead to first criticality of PALLAS in 2016.
8. The PALLAS research reactor will continue to increase the high quality research and medical isotope production in Petten with benefits for the nuclear infrastructure in the European Union, and the world.

Acknowledgements
The authors are grateful for the strong and intense support of the personnel of NRG and JRC-IE contributing to the specification and tendering process for the design and building of PALLAS. Unfortunately only one tender can be awarded the contract for the design and building of PALLAS, but we are very grateful for the efforts put in all the tenders with their comprehensive and inventive conceptual designs.

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Tables

Table 1 Treatment spectrum of Molybdenum and Lutetium

Molybdenum	
1. Cardiology	(Heart diseases)
2. Oncology	(Cancer diagnosis/treatment)
3. Urology	(Kidney function)
4. Neurology	(Brain function)
5. Infections	(Unknown origin)
6. Many others	

Lutetium
GEP NET (Gastro Entero Pancreas Neuro Endocriene Tumoren):

Table 2 Co-operation in the domains of fusion and Generation-4 fission energy

- CEA, GEN-4 & Fusion
- CRPP, Swiss, Generation-4
- ENEA, Italy, Fusion
- ESKOM, South-Africa, Generation-4
- FZJ, Germany, Fusion
- FZK, Germany, Generation-4
- JAEA, Japan, Generation-4, Fusion
- JRC-IE, EU, Generation-4
- KAERI, South-Korea, Generation-4
- KURCHATOV, Russian Federation, Fusion
- ORNL, Tennessee, US, Generation-4
- PNL, Washington, US, Fusion
- RID, Delft
- SCK, BelgiumGeneration-4, Fusion

Figures

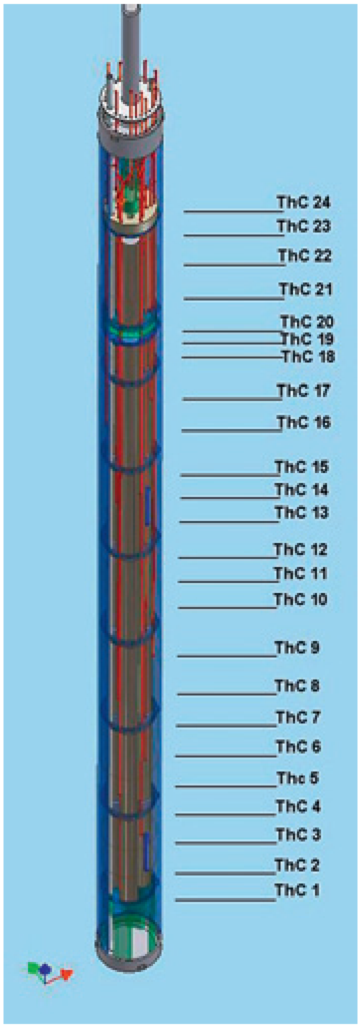

Figure 1 HFR rig for graphite experiments with thermocouple locations indicated. Rig length over 500 mm [reference 2].

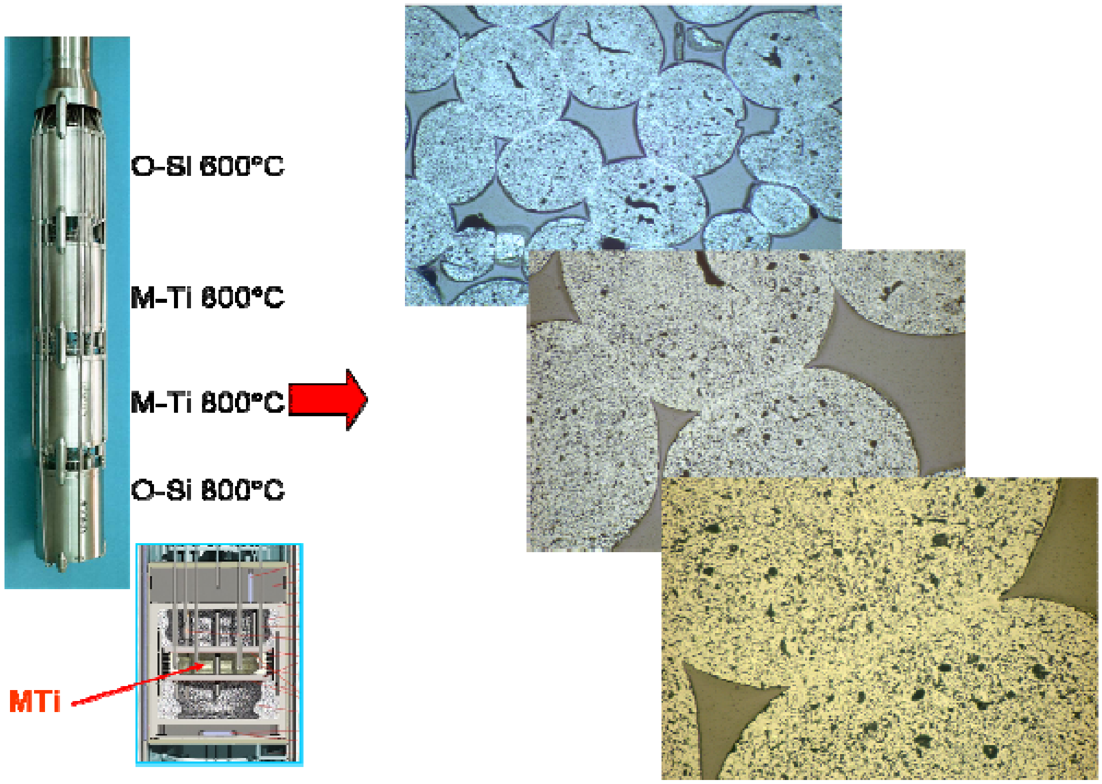

Figure 2 ITER test blanket module relevant test in HFR. Titanate particles show sintering, particle size diameter about 1 mm, [reference 7].

Figure 3 Research Reactors foreseen in the European Union, [reference 8].

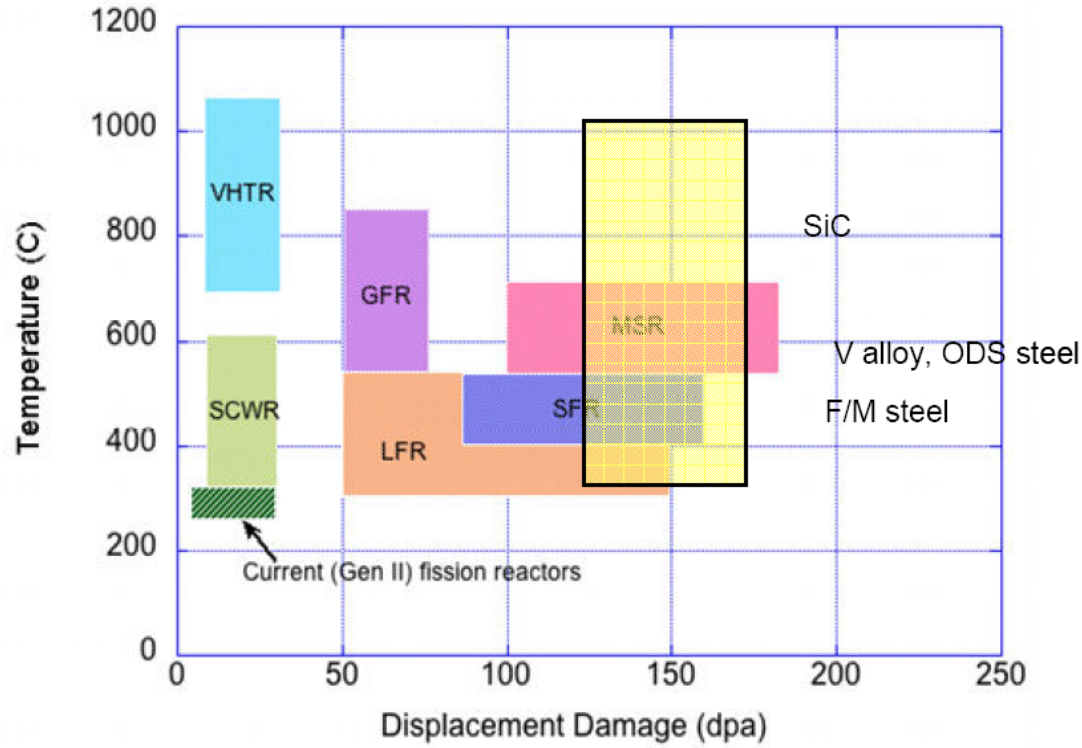

Figure 4 Main application temperature and displacement damage windows for Materials Research in the fisson (fields) fusion (box) domain, after Zinkle ORNL, Oak Ridge, [reference 8].