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Structural materials for fusion and spallation sources

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Abstract

Experimental investigation of neutron-induced irradiation damage in structural materials is fundamental to the development of magnetic confinement fusion. Proposals for the testing of candidate materials are described, indicating that a period of at least 10 years will elapse before a suitable high neutron fluence fusion test facility becomes available. In this circumstance, the possibility that neutron spallation sources could be exploited to shorten the time-scale of fusion materials development is attractive. Although fusion displacement and transmutation reaction rates can be replicated in spallation sources, there are significant differences arising from the harder neutron spectra and the presence of energetic protons. These differences, including higher energy PKA, electron heating effects, transmutation rates and pulsing are described and their consequences discussed, together with the concomitant development of theoretical models, needed to understand the effects. It is concluded that spallation source experiments could make a significant contribution to the database required for the validation of theoretical models, and hence reduce the time scale of fusion materials development.

1. Introduction: fusion development and materials requirements

As the time approaches when a final decision is made on the construction of the international 'next step' ITER tokamak device [1], the fusion community has to form a strategy leading to the development of a practical and economic electricity-generating device. Following an initiative from the UK government, the European Union has discussed the adoption of a 'fast-track' approach to fusion, which would reach this programmatic aim in the shortest possible time. The 'fast track' approach has immediate implications for the development and testing of materials.

The ITER device is planned to operate at a fusion power of 500 MW for pulses of about 0.5 h, and would demonstrate much of the technology required for the commercialisation of fusion power. However, a combination of its relatively low power density and low availability will limit the lifetime radiation damage dose to a maximum value in structural materials of 3–4 dpa. The 'fast-track' programme leads directly on to the construction of a single upgradeable DEMO device, which would demonstrate both the technological feasibility of fusion electricity generation and its economic performance, with target damage fluences of up to 150 dpa for the structural component replacement lifetimes. An important distinction between these devices arises in the choice of materials. ITER employs austenitic steel as its principal structural material, this choice being made because it is sufficiently well qualified for the purposes of immediately constructing and licensing the device. However austenitic steels are known to be too life-limited by high temperature helium embrittlement for use at higher fluences, and there are no useful fusion-relevant low activation versions of this class of material. The principal candidate material for DEMO is low-activation ferritic–martensitic (LAFM) steel, with vanadium alloys and SiC/SiC as longer-term possibilities.

The transition from ITER to DEMO cannot be made reliably unless a structural material has already been qualified for the high fluences required, and this can only be achieved by testing in a separate special purpose

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device. The fusion community has produced a conceptual design of a deuterium–lithium neutron-stripping source IFMIF (International Fusion Materials Irradiation Facility [2]), which would produce a reasonable approximation to the appropriate neutron spectrum and provide the necessary fluence for testing at DEMO conditions. Its main limitation is a small irradiation volume ($\sim$0.5 l at the highest flux), so that results are dependent on small-scale testing methods and are thus restricted to material and joint samples rather than components.

Two possibilities exist for component testing. The most optimistic assumes that the results from IFMIF, taken together with the results of low-fluence testing of tritium-breeding modules on ITER, would be sufficient to provide the initial design of DEMO, and that further component development would be a task for the DEMO operating programme. The alternative is that a volume neutron source – for instance a compact current-driven spherical tokamak – would succeed IFMIF and be used before and/or in parallel with DEMO. However, even completion of the engineering design of IFMIF followed by its construction would take roughly 10 years. In these circumstances the possibility of proceeding with the experimental investigation of fusion materials using existing neutron sources is being investigated. Of the range of available neutron sources, only spallation devices produce a fusion-relevant ratio of transmutation helium (in atomic parts per million or appm) to atomic displacement reaction rate. In DEMO this parameter is in the range 10–15 appm/dpa in the highest neutron flux positions.

In this paper, the similarities and differences between fusion and spallation source irradiation conditions are explored, and the possible use of the pulsed ISIS spallation source (Rutherford-Appleton-Laboratory, UK) for low-fluence testing discussed. It is noted that the helium production/displacement damage ratios within the target volume of the proposed European Spallation Source (ESS) [3] are similar to predicted fusion values. However there are some obvious differences between spallation and fusion, for instance the electronic interactions of high-energy protons, spectral effects on the formation of displacement cascades, the different mix of transmutation species, the operating and shutdown temperatures of materials, and the effect of pulsing. Tantalum target plates, irradiated with 800 keV protons up to 11 dpa in the ISIS machine have already been analysed. Such data, in collaboration with irradiation on a continuous source such as SINQ (Paul-Scherrer Institute, Switzerland), could provide a direct assessment of pulsing effects. Other possibilities would result if an actively cooled irradiation facility were developed that permitted the insertion and removal of samples independently of target replacement operations. In principle, such a facility could be used to control the temperature of samples, and would also permit control of the duration of irradiations so that comparisons of the effect of different spectra at the same fluence could be made.

2. Comparison of materials testing methods and properties

In a fusion power plant the irradiation damage inflicted immediately behind the plasma facing surface is caused by 14 MeV neutrons from D–T fusion reactions in the plasma. Through collisions in the first wall, support structures, coolant and blanket, the neutron energy is degraded and the spectrum flattened. Most of the damage is in fact due to backscattered neutrons with energies $E < 14$ MeV. The monochromatic 14 MeV elastic primary knock-on atom (PKA) energy spectrum is flat with a cut-off of around 1 MeV for iron, whereas the mean PKA energy at the first wall, resulting from the degraded neutron spectrum and complicated by inelastic scattering, is $\sim$50 keV [4]. Primary damage is normally measured in terms of the displaced atom production, and it is convenient to use it here for comparison purposes. The PKA energy corresponding to half of the damage energy is $\sim$160 keV. As regards materials properties, the situation is even more complicated and is determined by processes occurring after the damage process (e.g. recombination, clustering, migration to sinks, etc. of defects).

The two main damage mechanisms generally considered in the case of fusion are the creation of displaced atoms and the generation of gas, particularly helium. Table 1 gives typical values of these quantities for a fusion power plant. However, there is no fusion irradiation facility capable of such irradiation; the most intense 14 MeV neutron source ever constructed (RTNS-II) has produced a maximum dose that is four orders of magnitude less than is required with about $\sim$0.01 dpa after about six months of irradiation [5]. Until a dedicated d-Li stripping facility is constructed, the main present-day options for fusion damage simulations are: mixed spectra materials testing reactors, fast reactors, and spallation neutron sources. The range of displacement rates and helium production rates for various neutron sources are compared in Fig. 1.

Mixed spectrum reactors used for fusion materials testing have three main limitations. These are: the damage is produced mainly by low energy recoils under 100 keV, the damage rate is a factor of 2–10 times too small and the gas production rate in materials like F82H is a factor of 50 too low. In fast reactors (such as Phénix or EBR-2), most of the damage is from recoils with under 200 keV (peaking around 50 keV), displacement rates similar to the fusion case but gas production rates similar to those in mixed spectrum materials testing reactors. Stripping sources (such as the proposed IFMIF facility) provide a good match to fusion conditions in

Table 1
Comparison of neutron irradiation conditions between a proposed magnetic confinement fusion power plant (DEMO) and a spallation neutron materials testing source

	D–T fusion power plant	Spallation source
Neutron energy (MeV)	14	<1000 (target) <300 (reflector) [typical values]
Maximum (and mean) energies (MeV) of elastically scattered PKA Fe nucleus	0.95 ($\sim$0.05)	69 (target) 21 (reflector)
Time behaviour	Quasi-steady-state	Pulsed: ISIS (160 kW, 1 μs, 50 Hz); ESS (5 MW, 1 μs, 50 Hz); steady-state: SINQ (500 kW)
Neutron flux ($\mathrm{n\,m^{-2}\,s^{-1}}$)	7×10^{18}	3×10^{17}–3×10^{19}
Damage rate (dpa/s)	$\sim10^{-6}$	3×10^{-8}–3×10^{-6}
He rate (appm/s)	$\sim10^{-5}$	10^{-6}–10^{-4}
H rate (appm/s)	$\sim2\times10^{-5}$	10^{-5}–10^{-3}

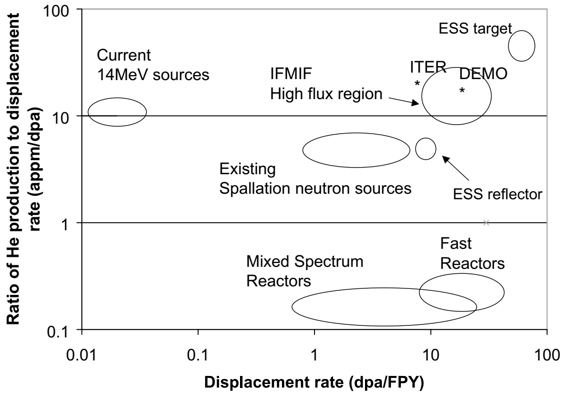

Fig. 1. Representation of the typical ranges of ratio of displacement rate to helium production rate (He appm/dpa) as a function of the displacement rate per full power year (dpa/FPY) for various neutron irradiation situations. Typical example data are for: current 14 MeV source (RTNS-II); mixed spectrum reactors (HFIR (USA), HFR (NL), SM2 (RF)); existing spallation sources (PIREX, SINQ, LAMPF and ISIS in the neutron-only irradiation regimes).

terms of recoil spectrum, displacement and gas production rates, but in a volume of $\sim$0.5 l. The maximum neutron and PKA energies are about twice those for the fusion case; such sources are generally recognised as the best alternative to a fusion neutron source. The d-Li stripping source concept is the one that best matches the requirements of fusion simulation, but the conditions in ESS straddle those of fusion, making it an interesting possibility.

According to data provided as part of the IFMIF assessment [2], an irradiation facility adjacent to the reflector of an unspecified spallation neutron source can match fusion DEMO displacement rates. New data, however, from the ESS project [6] indicate that the contribution from neutrons in the energy range 10 MeV to 1 GeV is significant in the proton beam region of the target. The transmutation rate from such energetic particles is large and reactions may affect the phase stability of alloys that are particularly sensitive to small changes in chemical composition. Thus studies are needed of such possible changes and the impact that transmutations might have on mechanical properties. Away from the axis/reflector position, the bulk of the atom displacements arise from neutrons with energies similar to fission energies, and the gas rates are lower than the fusion case by an order of magnitude. The gas generation rates of transmutation H and He have been measured at high energies (800–2500 MeV) [7,8].

Subject to engineering considerations, it may be possible to place samples in an intermediate region, just outside the path of the proton beam, where the transmutation rate is reduced to a tolerable level and the damage conditions (appm He/dpa) resemble fusion-like values. Thus there is the possibility that a large spallation source like ESS could provide a useful small-volume irradiation facility. At such a location, the 5 MW target of ESS can provide up to about 70% of the rate calculated for the DEMO power-plant with a wall loading of 2 MW/m^2. The size of the usable irradiation volume is limited by the steep gradient in the high-energy component of the neutron spectrum away from the beam axis. The resemblance to the fusion-like conditions would obviously be sensitive to the exact position in the intermediate region. Hence it would be important to include, with the irradiation sample, a monitoring device to ensure that the sample is in the optimum fusion-relevant region. Even here, however, the highest energy neutrons (albeit with low fluxes) are much more energetic than in the fusion environment and we may need to consider related effects.

Information on the neutron spectrum for the ESS was provided in [4]. Fig. 2 combines this information with data from the IFMIF project so that the conditions

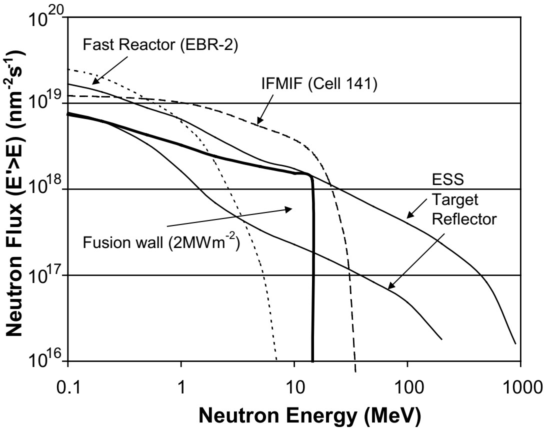

Fig. 2. Neutron flux spectra for a range of sources [2,6].

can be properly compared. The higher energy neutron tails that arise from direct collisions between spallation protons and neutrons are significant.

A more realistic appraisal of the relative importance of high-energy neutrons in the ESS cases can be seen in Fig. 3. Here the differential spectral contributions of energy carried by the neutrons are plotted as a function of neutron energy. In the case of the ESS rigs 505 and 506, it can be seen that although there is a significant high-energy neutron tail, extending above 14 MeV (where the fusion peak is visible) and up to ~1 GeV, the relative number of neutrons in this component is relatively small. The only significant difference between the IFMIF case and the fusion case is the contribution of about 50% of the energy from neutrons between 14 and 35 MeV.

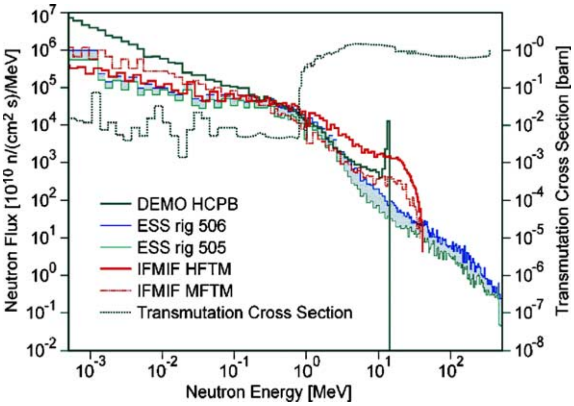

Fig. 3. Comparison of differential neutron spectra for the ESS irradiation rigs, IFMIF high and medium flux test modules (IFMIF HFTM and IFMIF MFTM) and DEMO HCPB. Typical energy dependence of the total transmutation cross section for iron (right hand scale) is shown.

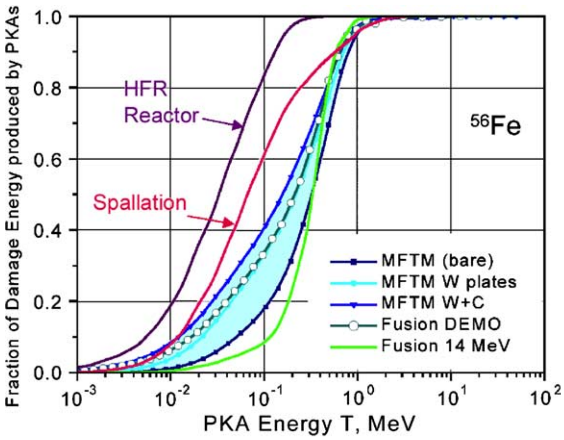

Fig. 4. Fraction of damage energy versus the PKA energy for various positions in IFMIF (hatched area), an advanced blanket (HCPB type), a fusion DEMO reactor (open symbols); the 14 MeV fusion peak, the mixed spectrum of fission reactor HFR, and for a typical position inside the ESS test module.

As noted above, collisions of the neutrons in materials spread the resulting PKA energies. This can be seen in Fig. 4, where we have plotted the fraction of the damage energy produced by iron PKAs versus the PKA energy for regions of IFMIF, a mixed-spectrum reactor, a fusion blanket and the ESS test module [9]. At high energies an increasing fraction of the energy goes into inelastic processes.

In the target region there is a greater contribution of neutrons that result from direct collisions with protons and have energies that extend up to the proton beam energy, i.e. up to ~1 GeV. This would mean that recoils in iron could be as energetic as 69 MeV. Such very energetic recoils have energies almost as high as fission product recoils and are quite different in character to lower energy recoils. These high-energy neutrons can create a larger number of transmutations. The irradiation field is also more intense in the target region.

3. Discussion

It is clear that evaluation of the possible use of spallation sources for fusion materials testing must depend on the special (i.e. non-fusion like) effects of the high energy PKAs produced, and their local heating effects, as well as the presence of the energetic protons in samples placed near the beam axis. Here we focus on the effect of the high-energy tail of the spallation particles. We have noted that 14 MeV fusion neutrons from D–T reactions produce elastically scattered Fe PKAs with kinetic energies up to ~1 MeV in fusion plant structures. This energy is significant since the PKA loses more than half its energy by electronic excitation. Indeed PKAs above 1 MeV will lose most of their energy by electronic scattering. One can see from Table 1 that a significant difference exists between PKAs produced in the fusion and spallation cases: a significantly higher fraction of the energy is given to the electrons in the target material in the spallation case.

For PKAs in the energy range: 1–35 MeV, maximum electronic energy loss rates in Fe are of the order of 10 MeV/μm. A 30 MeV PKA will travel a few μm, giving up energy to the electrons, before it can start to transfer its remaining energy to atomic displacements. At high energy the interaction between the PKA and the nuclei of atoms is by Rutherford scattering which decreases with increasing energy, while at low energies 'hard sphere' scattering occurs. The result is a long thermal 'rod' with a few isolated cascades, ending in a displacement spike. The difference in time and spatial scales of the two types of energy loss are large. Displacement damage is confined to a volume of less than about 10^{-23} m^3 (scale of around 100 nm) and the lifetime is of the order of a few picoseconds. The above considerations indicate that one should consider carefully the effects of the dissipation of electronic energy in the material [10]. Thermal deposition and local heating are known to lead to microstructural changes in materials, for electronic stopping powers above a critical value which is typically ~30 MeV/μm in metals and ~5–30 MeV/μm in ceramic insulators [11,12]. These threshold values are sufficiently close to the expected 1–35 MeV PKAs in the fusion case that further investigation may be appropriate.

Calculating the deposition of the energy and the subsequent diffusion of the energy is not simple and will be both structurally and temperature dependent. Even in a metal like copper the energy will be deposited within a few nm of the thermal rod. With an energy density of 20 MeV/μm it is possible to heat all materials above their melting point over a timescale of the order of 100 ps. We would expect a substantial shock to arise from the rapid expansion. Further, there will be an electrostatic component as well as a mechanical component to the shock, even in metals, arising from the separation of the electrons from the positively charged cores. We believe it is a process that deserves some investigation in the future.

The rapid energy deposition in the thermal rod may produce a number of effects including the punching of prismatic dislocation loops and the production of shocks. The final conclusion is that dislocation loop punching is a realistic mechanism and may already be responsible for loops observed in recent MD simulations [13]. However, interpretation of ion-beam mixing data with MD simulations in Ni, Pd and Pt [14] suggests that the electron-phonon coupling, thought to greatly enhance the cooling rate of thermal spikes in many metals, may only play a minor role. The behaviour for very energetic PKAs therefore deserves some careful study both experimentally and theoretically.

The leading candidate structural materials currently proposed for a fusion power plant are swelling-resistant LAFM steels, such as EUROFER [15] which will operate at temperatures of ~450 °C. For higher temperatures (<800 °C), and therefore with higher thermal efficiencies, advanced oxide-dispersion strengthened (ODS) ferritic alloys, containing yttria or TiC nanometre-sized strengthening particles, are being considered. The behaviour of such complex alloys under fusion neutron irradiation and high-energy PKA spectra, particularly the dynamics of precipitates, is difficult to predict. Experimental work in this area would therefore be essential. From light water reactor studies, it is known that mechanical properties are critically affected by the presence of minor components in steel, e.g. Cu. It is known that fine metal precipitates of Cu are nucleated from solution during irradiation and stabilised by PKAs of ~500 keV which give rise to hardening and embrittlement. Annealing coarsens such precipitates. Increasing the energy of the neutrons will most likely alter the precipitate behaviour in an unknown and complex manner. Possible outcomes include the complete removal of the smallest precipitates or the growth of the very largest. Thus it is not possible to say how much irradiation hardening will occur. The presence of the strong heating effect (thermal rod behaviour) and the disorganisation (liquid-like state) of the lattice during a cascade will profoundly increase the rate of resolution of precipitates in the material. There will be strong effects on swelling and mechanical properties.

The issue of whether the pulsed nature of spallation sources affect radiation damage behaviour has been considered recently [16]. In this work it was shown that the relation between the lifetimes of mobile self-interstitial atoms (SIAs) and vacancies and the time scales for pulsing in a source like ESS is such that pulsing would not alter the damage compared with equivalent steady-state conditions. However, the conclusion in [16] depends strongly on the irradiation temperature [17,18]. The higher temperature ranges of fusion applications may therefore render the materials susceptible to pulsed irradiation effects. It was also concluded in [16] that pulsing would neither affect the rate of growth of cavities nor significantly alter the irradiation creep rate via the mechanism of climb-controlled glide. Pulsing of a 10 MeV deuteron beam, however, has been shown experimentally to affect the irradiation creep rate of austenitic stainless steels at low doses [19]. In these experiments a 'resonance' was found between the pulse period of the beam (at a pulse width ~100 s) and an anomalously enhanced creep rate. We note that this critical beam pulse period is much longer than the pulse periods given for the sources in Table 1; no enhanced creep effects were seen for periods as low as 2 ms.

4. Possible near-term experiments

The ISIS pulsed spallation source has been used with Ta target plates. Spent plates, irradiated at temperatures less than 200 °C, and with a total damage of up to ~11 dpa by 800 keV protons have been analysed by microhardness, bending and tensile tests [20]. The results showed the expected irradiation hardening increasing with dose but, also, surprisingly, that the ductility remained high. No irradiation creep measurements were reported on the Ta specimens. At present it is not clear whether the lack of irradiation embrittlement (observed in other irradiation experiments on Ta at even lower doses) is due to the pulsing of the source or the different impurities of Ta specimens.

With such experiments in mind, consideration is being given to the possibility of irradiating candidate materials in the ISIS spallation source to obtain low-fluence results in advance of the availability of facilities capable of providing power plant doses. Recalling that ISIS is a pulsed source and incapable of providing the flux levels available on SINQ, the reasons for proposing such a programme rest on the following points.

- Although many of the possible effects of pulsing have been dismissed on the basis of theoretical arguments [16] there are still significant doubts on the matter, which would best be resolved experimentally. Specifically, the investigation of the evolution of the microstructure under equivalent pulsed and steady state conditions would provide a sound basis of understanding. Obviously this comparison would require baseline irradiations on a continuous source at similar temperatures and doses.
- The irradiation of samples at temperatures appropriate to fusion (450–500 °C for steels) would provide complementary information to the lower temperature experiments conducted on SINQ.
- The ability to insert and withdraw samples independently of target loading operations would allow controlled dose experiments to be conducted.

These requirements can only be met by the construction of an actively cooled irradiation facility incorporated into the design of a solid target or immersed in a liquid one. The plans to construct a new target station for ISIS offer the possibility of providing a facility. The planned second target station for ISIS will receive 80 kW of beam power. If high transmutation rates are to be avoided, an irradiation facility must be placed outside the proton beam. Then a maximum damage rate of some 10^{-9} dpa/s can be achieved, i.e. a dose of several dpa would take 20–30 years of full power operation. However before a feasibility study and preliminary design are commenced, a consensus from the potential user community is needed on the scientific case for the experimental programme.

Experiments to determine the effect of the PKA energy (at constant dpa, temperature and at low doses) could possibly be done by placing a group of identical fusion-representative test samples at different radial positions from the spallation beam axis. Those samples close to the axis will suffer damage with higher energy PKAs than samples placed further away. Each sample will need to be kept in the irradiation field for different times to keep the damage level (total dpa per sample) constant. Samples placed at different radial locations, however, will experience different damage rates (dpa/s), a parameter potentially important for microstructural evolution. Thus a prerequisite for this experiment is to obtain a set of data assessing the likely effect of damage rate, independent of the incident particle energy. PIE microstructural examination might then reveal any differences that can be attributed to the PKA spectrum.

5. Summary and conclusions

There can be no doubt that a d-Li stripping source, such as the proposed IFMIF project, provides the best match of 14 MeV neutron irradiation conditions for fusion materials testing in small volumes (~0.5 l). We have considered the main damage parameters: dpa/FPY, He appm/dpa and PKA spectrum. None of the other types of neutron sources considered here can be considered as a genuine alternative to IFMIF. However, the minimum timescale for the design, construction and commissioning of the IFMIF device is of the order of 10 years. Therefore, in order to consider how best to follow the proposed 'fast track initiative' route to fusion energy, this paper has looked at how the gap between the very different materials requirements of the ITER and DEMO devices may be bridged. In particular, we have investigated how, in the near-term, spallation neutron sources could contribute to fusion materials testing and we have considered the key issues connected with their use in this context.

Although spallation sources can provide fusion-like gas generation and displacement damage rates in test samples, they are characterised by higher energy neutrons and protons. The main points arising from this are:

- The high neutron energies in the target region of spallation sources generate PKA atoms with energies much larger (typically $E < 69$ MeV) than would be the case in fusion power plant ($E < 1$ MeV).
- The high energies of the PKAs in a spallation source may give rise to strong local electron heating effects: a 'thermal rod' is produced which may produce shocks, electrostatic effects, and the punching of prismatic dislocation loops.
- In low-activation ferritic alloys proposed as structural components, the disorder (liquid-like state) in the irradiation-produced cascade is likely to affect

the coarsening behaviour of precipitates in the steel which give rise to hardening and embrittlement. These effects are hard to predict and therefore experiments are needed.
- An experiment to determine the effect of the hardness of neutron spectrum at low fluence and at constant dpa on irradiation damage is proposed for the ISIS source. This may help resolve some of the issues above. Concomitant measurements of the effect of damage rate (at constant incident particle energy) on microstructural evolution are also needed.
- The effect of transmutation by high-energy neutron or proton reactions must be limited.

With regard to the pulsed nature of the sources, the main point arising from this is:

- Theoretical analysis suggests that source pulsing, with typical pulse types, will not affect the nature of irradiation damage provided that the irradiation temperatures are low. However, there is a need to validate this conclusion using simple experiments, comparing pulsed and steady state irradiations in similar samples and at temperatures relevant to fusion power-plant operation. A possible experiment is proposed.

In conclusion, despite the obvious differences that we have reviewed here between 14 MeV fusion and spallation source neutron damage, the potential fundamental physics data obtained from the use of spallation sources are likely to help develop and validate models. At present it is possible only to extrapolate from existing thermal and fast reactor irradiation data towards fusion-like conditions. However, by using spallation data we would extend the range of parameters to encompass the conditions in fusion structural materials.

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