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New evaluation of displacement damage and gas production for breeder ceramics under IFMIF, fusion and fission neutron irradiation

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Abstract

Design and development of accelerator driven neutron sources, in particular the International Fusion Materials Irradiation Facility (IFMIF), demands neutron cross-sections with extended energy range up to 50 MeV, as standard data libraries are truncated at 20 MeV. Based on recently released new cross-section data for lithium isotopes, damage and gas production rates were re-evaluated for the lithium-based breeder ceramics Li_4SiO_4 and $LiAlO_2$ under several neutron irradiation conditions. Calculated irradiation responses for mixed spectrum neutrons (HFR-Petten), fusion neutrons (DEMO reactor) and the accelerator-based d-Li neutron source IFMIF are presented. This study indicates, that IFMIF provides not only favorable irradiation conditions for structural materials but also can be a suitable test bed for fusion breeder materials. In the latter case an appropriate neutron moderator/reflector should be used to adjust the low energy tail of the IFMIF spectrum to that one of a typical DEMO reactor blanket.

1. Introduction

As long as a suitable intense fusion neutron source is not available, the irradiation behavior under fusion specific conditions remains a critical issue [1,2]. Since an appropriate fusion neutron source does neither exist at present nor will be available within this decade, irradiation performance studies have to rely in a foreseeable timeframe still on fission reactors and ion accelerators. As in major international fusion strategy scenarios the timely availability of reliable material databases is on a critical path, an early accessibility of an intense fusion neutron source is indispensable [3]. Various neutron source studies and high ranking advisory boards came after thorough and comparative assessments of relevant concepts including spallation sources to the conclusion that an accelerator-based d-Li stripping source (IFMIF) is the most advanced and most suitable option [4,5].

The majority of specimens made of structural materials will be irradiated in the high flux test module (HFTM) of IFMIF followed by post-irradiation examination in hot cells [6,7]. In addition, more sophisticated in situ push–pull creep fatigue tests and in situ tritium release tests on Be and various breeder materials are foreseen in the medium flux region. The present study is mainly aimed to provide the irradiation conditions in the medium flux test modules (MFTMs) of IFMIF for candidate solid breeder materials, like Li_4SiO_4 and $LiAlO_2$, by fitting as close as possible fusion power reactor conditions. The neutronic responses for MFTMs are compared with those for a typical rig position in the mixed spectrum high flux reactor (HFR-Petten) and the front wall position of the helium cooled pebble bed (HCPB) blanket of a fusion DEMO reactor.

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2. Adaptation of IFMIF neutron spectra

In the IFMIF reference design concept, the vertical test assembly of the medium flux region is equipped with two MFTMs: (i) a module for in situ creep fatigue experiments housing a miniaturized universal testing machine for simultaneous and independent testing of three push–pull fatigue specimens, and immediately downstream the beam axis, (ii) a module for in situ tritium release experiments on Be and various ceramic breeder materials. While for structural materials like Fe-based alloys, IFMIF has shown to meet DEMO reactor relevant H, He and dpa production rates as well as H/dpa and He/dpa ratios very well [6,8], the population of low energy neutrons in the IFMIF MFTMs was not sufficient to match in addition HCPB blanket relevant tritium production rates for breeder materials properly. As the cross-section of the ^{6}Li(n,t)^{4}He reaction increases at low energies as $\sim 1/\sqrt{E}$, the low energy tail of the neutron spectrum should have sufficient flux density. In order to meet this additional requirement, the former MFTM reference design of the conceptual design analyses (CDA) phase (1995–1996) – labeled as 'MFTM, no moderator' in this work – was improved by systematic neutron transport studies with the aim to soften the IFMIF neutron spectrum in the MFTMs by enhancing the fraction of low energy neutrons. The related neutron spectra were calculated by means of MCNP4c Monte Carlo code [9] using several realistic geometry models of the facility.

2.1. Materials screening for MFTM neuron moderator

The initial neutron transport calculations [10] have been extended and include now a variety of materials including Fe- and Ni-based alloys, W, SiC/SiC, graphite and H_2O. A major result of the screening study is that the integration of two 30 mm thick tungsten plates at the front and back side of the IFMIF creep–fatigue test module levels very effectively the high energy neutrons peaked around 12–16 MeV and, at the same time, notedly increase the neutron population in the range 5×10^{-3}–5×10^{-1} MeV mainly due to backscattering. Between 10^{-2} and 10^{-1} MeV the neutron flux increase is more than fourfold. This spectral shifter effect is most pronounced immediately behind the W-plates, that is, in the test module dedicated to in situ tritium release experiments on Be and ceramic breeder materials.

These neutron transport calculations also revealed that a replacement of tungsten by e.g. iron or nickel-based alloys would have only a very moderate spectral shifter effect. A substitution of tungsten by SiC_f/SiC and graphite would increase the high-energy part (>3 MeV) of the neutron spectrum by about 50% and 80%, respectively, mainly due to elastic scattering but would have practically no 'spectral shifter' effect in the medium and low energy parts.

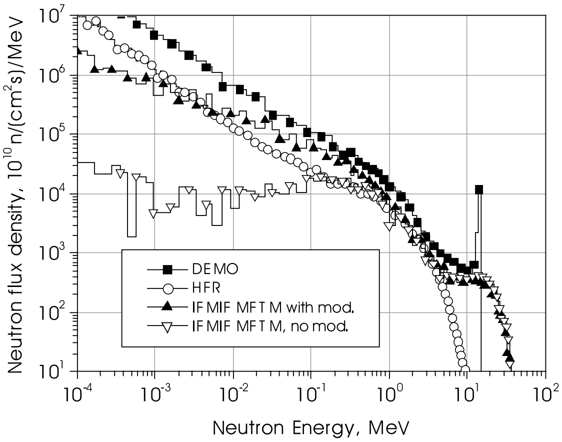

Fig. 1. Neutron spectra of IFMIF HFTM and MFTM with tungsten moderator plates and graphite reflector in comparison with the mixed HFR spectrum and HCPB blanket of fusion DEMO reactor.

2.2. Materials screening for a coating of the test modules

After a significant fraction of high-energy neutrons could be shifted to lower energies by using an appropriate tungsten moderator, it was tried in a second step to increase substantially the flux of low energetic neutrons. To achieve this goal, the test modules were encased by neutron reflector materials in the geometry models for the MCNP4c calculations.

Several coating materials and designs were analyzed to assess their influence on the neutron spectrum. Particularly with regard to technical realization, the best results were obtained by an additional carbon jacket of 30–40 cm attached to the MFTM and the low flux test module. Such a carbon coating significantly increases the population of low energy neutrons and thus greatly improves the testing conditions for breeding ceramics, e.g. below 10^{-2} MeV, the flux increase is more than two orders of magnitude. Fig. 1 shows typical neutron spectra of the HCPB DEMO blanket [11] and the HFR [12] in comparison with different spectra inside IFMIF MFTM. Obviously, a major result of the neutronics calculations is, that with two tungsten plates acting as 'spectral shifter' and additional carbon coating acting as 'reflector', the shape of the neutron spectrum in the MFTM follows over several orders of magnitude closely that one of a DEMO-type HCPB breeding blanket.

3. Comparison of irradiation responses

3.1. Damage and gas production rates in ceramic breeders

Although substantial efforts are being made during the past few years to extend existing nuclear data libraries from 20 to 50 MeV or even beyond, evaluated nuclear data are still available only for a limited number of isotopes. Therefore, calculations were made only for Li_4SiO_4 and $LiAlO_2$, but not yet for other ceramic breeder materials of interest like Li_2TiO_3 or Li_2ZrO_3. The high-energy nuclear data files LANL [13] and INPE [14] have been used for this work.

For breeder ceramics composed of elements with significantly different masses a physically correct treatment of the displacement damage is necessary. While for quasi-monoatomic matter the neutron response can be calculated with common computational tools, a sound calculation and evaluation of the displacement damage in multi-component materials like breeder ceramics requires improved codes beyond the classic NRT-model. Therefore, the primary knocked-on atom (PKA) spectra have been calculated using the NJOY module GROUPR [15], and for a proper treatment of sub-lattice specific threshold energies and damage rates for the breeder ceramics, the Boltzmann transport code BOLT [16,17] has been selected. For this model of displacement damage calculation the displacement threshold energies of each element in compound are the important input parameters. In the present work we have utilized E_d values provided with the SPECTER code [18]: 10 eV for Li, 25 eV for Si, 30 eV for O and 27 eV for Al. The values suggested by Greenwood are widely used for dpa damage calculations in compound materials. Nevertheless a careful assessment of the displacement threshold energies is necessary (see e.g. [19]), but it is outside of the scope of the present paper which is focused on the different neutron spectra comparison.

As the neutron responses for the ceramics Li_4SiO_4 and $LiAlO_2$ are qualitatively very similar in a given neutron source, figures are shown only for Li_4SiO_4 enriched with 30 at.% ^{6}Li. Fig. 2 reveals that the implementation of moderators and reflectors increase the damage production rates of all isotopes in the ceramic by more than a factor of 2 in the MFTM of IFMIF. Although not foreseen in the present reference test matrixes, accelerated irradiation of breeder ceramics would only be possible in the HFTM.

With respect to gaseous transmutation products, the implementation of moderators and reflectors improves significantly the helium and tritium production in the MFTM by a factor of 4 and 10, respectively, but has only a moderate effect on the less relevant proton and deuteron production (see Table 1). On the other hand, the mixed spectrum reactor HFR with its overwhelming density of low energy neutrons has comparatively high T and He production rates coming from the ^{6}Li(n,t)^{6}He reaction, but, on the other hand, due to the lack of high energy neutrons a negligible H and D production.

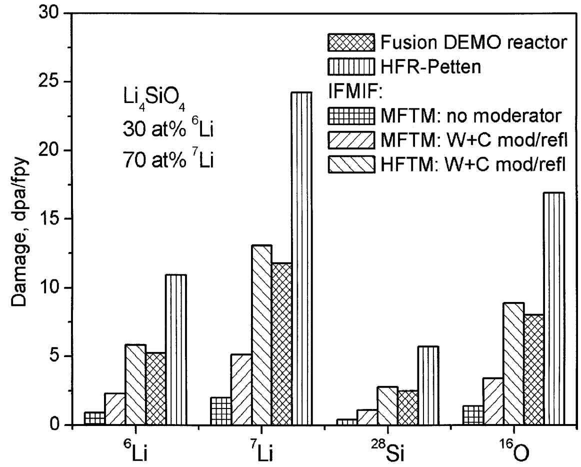

Fig. 2. Comparison of damage production in the breeder ceramic Li_4SiO_4 for IFMIF MFTM (CFTM position), IFMIF HFTM, fusion DEMO reactor (the first wall of HCPB blanket) and HFR (F8 rig position).

In Table 1 the different neutron sources are compared with respect to damage and gas production rates in Li_4SiO_4 and $LiAlO_2$. It is interesting to note that damage rates in meta-aluminate are systematically higher than in orthosilicate for all neutron spectra considered. This is mainly due to high displacement rate of aluminum as well as due to more efficient oxygen displacement in meta-aluminate. Two oxygen atoms in meta-aluminate posses more displacements than four atoms in orthosilicate. This effect cannot be explained by direct summation of displacements of different components as is usually done by extending NRT approach to multi-component materials. We could suppose that oxygen in meta-aluminate is additionally displaced by collisions with aluminum, while silicon in orthosilicate is displaced much less effective. Smaller damage rates in orthosilicate make it more preferable candidate solid breeder material. On the other hand, compared to orthosilicate, meta-aluminate has generally higher gas/dpa ratios in all neutron sources investigated, mainly due to the Li^6 content. Although the T and He production rates and consequently the T/dpa and He/dpa ratios could be significantly increased for breeder materials in the MFTM of IFMIF, the later are still about a factor of 1.7–1.9 below DEMO reactor typical values, while for the HFR these ratios are too high. In mixed spectrum reactors, DEMO typical T/dpa and He/dpa ratios can be adjusted in breeder materials quite well, e.g. by an implementation of suitable Cd-coatings that act as thermal neutron shielding, or by using different ^{6}Li enrichments [11]. It is interesting to note that in the case of the lithium-based breeder materials damage rate is pronouncedly dependent on ^{6}Li contents and low energy neutron part of the spectrum.

Table 1
Dpa and gas production in Li_4SiO_4 and $LiAlO_2$ in medium (MFTM) and high (HFTM) flux test modules of IFMIF, in the F8 rig position of HFR and in the HCPB DEMO fusion reactor blanket

Irradiation parameter	Demo reactor	HFR position F8	MFTM no moderator	MFTM W + C moderator	HFTM W + C moderator
Total flux, 10^{14} n/cm^2 s	11.94	3.83	1.54	4.84	10.09
Li_4SiO_4 (30 at.% ^6Li)					
Damage, dpa/fpy	27.5	57.8	4.8	11.9	30.6
H, appm/fpy	329.9	2.6	216.1	142.8	607.3
D, appm/fpy	508.9	40.3	242.6	200.0	927.4
T, appm/fpy	1.8×10^4	1.9×10^5	0.4×10^3	4.2×10^3	3.2×10^3
He, appm/fpy	2.0×10^4	1.9×10^5	1.0×10^3	4.7×10^3	5.4×10^3
H/dpa	12.0	0.0	45.3	12.0	19.9
D/dpa	18.5	0.7	50.8	16.9	30.3
T/dpa	664.5	3216.4	75.9	355.7	104.9
He/dpa	712.0	3217.2	213.9	396.9	176.0
$LiAlO_2$ (90 at.% ^6Li)					
Damage, dpa/fpy	30.2	82.4	5.0	12.6	31.9
H, appm/fpy	594.3	4.0	315.5	207.2	871.8
D, appm/fpy	823.3	68.0	402.1	332.7	1544.9
T, appm/fpy	3.0×10^4	3.1×10^5	0.2×10^3	7.0×10^3	4.2×10^3
He, appm/fpy	3.2×10^4	3.1×10^5	1.1×10^3	7.5×10^3	7.1×10^3
H/dpa	19.7	0.1	63.5	16.4	27.4
D/dpa	27.3	0.8	81.0	26.4	48.5
T/dpa	990.0	3806.3	48.9	542.2	130.7
He/dpa	1048.2	3807.3	225.0	594.5	223.4

3.2. Weighted-average recoil spectrum

It is well known that different PKA spectra can produce completely different damage morphologies. A low energy PKA produces Frenkel defects, that is, isolated vacancies and interstitials. The significant fraction of these defects survives recombination. On the other hand, high-energy recoils generate atomic collision cascades in which a high fraction of produced defects is subject to in-cascade recombination. Also for a high energy PKA, the formation of sub-cascades becomes more probable, which can lead to more intense radiation hardening. Therefore, recoils of different energy produce different damage morphologies that result in different mechanical properties of irradiated materials.

For the evaluation of the entire PKA spectrum, it is useful to construct a function that weights each recoil by its associated damage energy. The usual way to characterize a PKA spectrum is thus to use the cumulative damage production function $W(T)$, which represents the fractional damage energy created in all PKA recoils with energies between the threshold energy E_d and the energy T.

For ceramic breeder materials irradiated in a HCPB DEMO reactor blanket, this damage production function $W(T)$ continuously increases over about two orders of magnitude with increasing PKA energy, revealing a broad energy spectrum of recoils. A specific feature of breeder ceramics is the sudden increase around 2 MeV caused by an additional contribution coming from nearly mono-energetic tritium (2.6 MeV) and α-particles (2.0 MeV) emitted by the $^6Li(n,\alpha)t$ reaction. As shown in Fig. 3, all values within the hatched area can be achieved by a proper selection of W-moderators and C-reflectors. That is, except of the range 0.8–3 MeV, over the whole

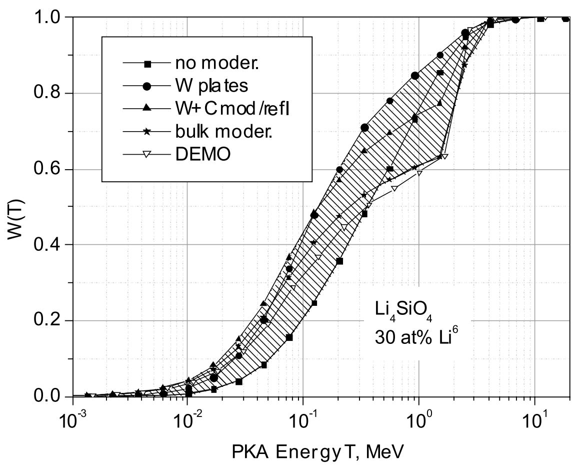

Fig. 3. Damage production function $W(T)$ in Li_4SiO_4 for the medium flux volume of IFMIF (closed symbols) in comparison with HCPB DEMO blanket.

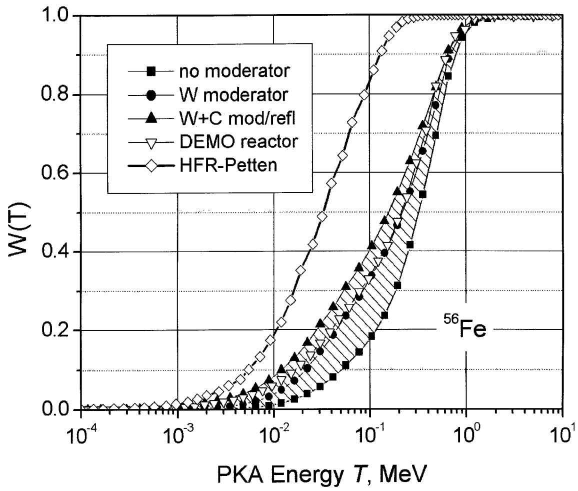

Fig. 4. Damage production function $W(T)$ in iron for HCPB blanket of DEMO reactor in comparison with IFMIF (hatched area) and HFR-Petten.

PKA spectrum DEMO specific recoil energy distributions can be adjusted in breeder ceramics irradiated in the present MFTM layout of IFMIF.

In contrast to lithium containing ceramics, the $W(T)$ function continuously increases without steps in structural materials of a fusion DEMO reactor. The hatched area in Fig. 4 shows that the HFTM of IFMIF meets perfectly over the entire PKA energy range DEMO reactor conditions in iron-based alloys, as the shape of the $W(T)$ function can be adjusted by using an appropriate combination of W-moderator and C-reflector. On the other hand, the PKA spectrum of the HFR is too soft by about an order of magnitude. Even if future experimental studies further confirm recent results of multi-scale modeling on a complete decomposition of high energetic cascades into sub-cascades [20] with the implication that for various materials the exact shape of the high energy part of fusion reactor $W(T)$ functions becomes less important, mixed spectrum reactors are still too soft for a fusion typical balance between isolated Frenkel pair type and cascade type irradiation damage.

4. Conclusions

The main goal of the present study was to achieve for ceramic breeder materials the same level of fusion irradiation simulation in the MFTM of IFMIF as already achieved for structural materials in the HFTM.

- Based on a screening of different materials and module designs, neutron transport calculations have shown that a suitable combination of W-moderator acting as 'spectral shifter' and C-coating acting as 'reflector', the population of low energetic neutrons can be substantially increased in the MFTM of IFMIF. Herewith the shape of the MFTM neutron spectrum follows over a wide range of neutron energies nearly that one of a HCPB DEMO reactor blanket.
- Displacement damage production in breeder materials: Compared to a HCPB blanket ('first wall' side), accelerated irradiation would be possible in the IFMIF HFTM.
- Gas production rates in breeder materials: Although in the MFTM of IFMIF the T and He production rates and consequently the T/dpa and He/dpa ratios could be significantly increased, the later are still about a factor of 1.7–1.9 below DEMO reactor typical values, while for the HFR these ratios are too high. Neutronics calculations for IFMIF MFTM are ongoing to further improve tritium and helium production rates.
- An appropriate combination of W 'spectral shifters' and C 'coating' allows for breeder ceramics in the MFTM a suitable adaptation, and for structural materials in the HFTM a perfect adaptation of the damage production function $W(T)$ for DEMO reactor conditions over the entire recoil energy range.

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