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Neutron Activation of Impurity Seeding Gases within a DEMO Environment

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In preparation for the design of a future tritium-handling plant for the DEMO fusion reactor, a study was undertaken to consider the activation of gases, in addition to those used as fuel, which are to be injected into DEMO for the purpose of reducing damage to the divertor. Likely impurity gases were identified as nitrogen, neon, argon, krypton and xenon, with no clear consensus as to which were preferred, and in what quantities these would be injected. Modelling with the FISPACT-II code, using the TENDL-2014 libraries as input, was performed to determine the products arising from their activation by the neutron flux expected at the DEMO first wall. The subsequent evolution of the progeny was investigated, along with a variation in the activation time.

Results indicate a variety of progeny are produced, most of which are short lived. However, the gamma emission from some progeny such as argon-41 from argon injection, from some metastable states and other isotopes of krypton and bromine following krypton injection, and from some metastable states of xenon following injection of xenon, could potentially cause concern.

Keywords: DEMO, tritium plant, neutron activation, impurity seeding.

1. Introduction

Plans for the operation of the proposed nuclear fusion power station DEMO require the injection of gas, in addition to the main fuelling gases. These help create good confinement of the plasma, and also act to protect plasma-facing components (PFCs), in particular the divertor. Without redress, the expected heat flux onto the strike point of the divertor is expected to be several 100 MW/m² for ITER, and similar for DEMO [1]. Current estimates of heat flux limits from materials that will be used for the divertor within ITER suggest 10 MW/m² is a maximum allowable flux, with less than 5 MW/m^2 averaged over time [2]. The presumption is that similar materials will be in use for the DEMO divertor. Injection of these impurity seeding gases, or plasma enhancement gases (PEGs), allows a significant proportion of heat to be isotropically radiated, via line-emission, which would otherwise be incident on the PFCs.

A number of research groups are considering the amounts and types of gas to be used as PEGs, both through modelling and by experiment, e.g. [3-9]. It is known that different gas species are more efficient at different locations within a tokamak, with higher mass impurities more efficient at radiating power at the higher temperatures found in the plasma core, whereas lighter atoms are better suited to the divertor region [3]. JET have experimented with nitrogen and with neon in plasmas [4] at concentrations of, respectively, 0.8% and 0.73%. AUG have also investigated nitrogen [5], as well as argon [6]. Argon has been investigated at a level of 0.5% within DEMO [7], as have argon/krypton and argon/xenon mixtures at around the 1% impurity concentration level [8], in addition to neon at 2% [9].

Due to the high neutron flux from D-T fusion reactions, activation will occur to all materials within the neutron shielding of DEMO. The work undertaken and presented here has been motivated by consideration of what gases a future DEMO tritium plant will have to process, in addition to fuel and helium ash. Priorities for the tritium plant design include determination of whether it will be necessary to take special precautions for activated progeny, whether any candidate PEGs should be avoided from the perspective of the tritium plant, and what treatment may be necessary for the activated PEGs and their progeny. This paper makes a first estimate of how much activation is expected and what products may be present within the tritium plant. Since a clear consensus has not yet been reached on which PEG will be used for DEMO, modelling has been performed for all candidate gases, namely nitrogen, neon, argon, krypton and xenon.

2. Method

Modelling was performed to determine the activation of candidate PEGs using the UKAEA code FISPACT-II [10] with the TALYS Evaluated Nuclear Data Library (TENDL) from 2014 as input. For the purposes of determining the response of how any material will respond to the neutron flux within DEMO, the UKAEA has compiled a large amount of data detailing the results of activation of 1 kg of an element placed at the outboard equatorial first wall of DEMO running at full power for two years [11]. Data within [11] are listed for all elements from hydrogen to bismuth, and the results presented in this paper are based on the methods developed therein. More complete details of the code and its history are presented in the introductory section of [11], but in summary, activation of materials is determined by irradiating each candidate impurity gas by the neutron spectrum predicted under in-vessel Monte Carlo conditions from neutron-transport simulations.

Exact amounts of PEGs to be injected are as uncertain as the species to be used, with levels under consideration for DEMO varying between 0.05% [6] and 2% [9] for different gas types. We therefore make an estimate as 1% of the total amount used to be taken as being impurities. Scaling of these amounts should be linear. Taking the 2015 DEMO design parameters [12], which gives a value $\langle n_e \rangle = 7.983 \times 10^{19}$ m⁻³ and a plasma volume of 2502 m³, an estimate of 1.997×10^{21} impurity atoms is determined within the DEMO plasma. It is this value that is used in calculations for the results presented within this paper. Whilst such a method may skew results to higher activities and dose rates for heavier atoms, if a lower amount of impurity seeding gas would be used for heavier species [3], it does allow results to be presented simply and clearly, and allow the reader to make their own linear scalings to injection rates as required.

The activation of any substance within DEMO depends on the amount of time during which it is subject to a neutron flux. The residency time could vary across many orders of magnitude depending on, for example, location in the plasma, method of injection, coefficient of adhesion to the vessel wall and pumping speed for that particular gas species. Modelling for ITER has used a gas retention time of 5 s [13], whereas the maximum within DEMO could be two years for steady state operation. Whilst scaling of impurity amounts is linear, scaling of activation time is not. The FISPACT-II code was therefore run for a variety of activation times ranging from 10 s to the maximum possible for DEMO of two years. An example case of 500 s of activation are presented in the results section, in addition to plots displaying how activity and gamma dose rate vary as a function of activation time.

In addition to increasing the amount of activation with increasing time in a neutron environment, the cooldown of activity has been calculated. In [11], the activity over 10^4 years following activation is detailed. Due to the motivation of this work relating to the DEMO

tritium plant, results are shown here for a cooldown time of ten days, since this shows the highest levels of activity and it is likely that PEGs pumped out of the DEMO torus will have been separated and either reinjected into the plasma, or disposed of.

3. Results

The total activity and gamma dose rate, both plotted as a function of activation time, are shown in, respectively, Figures 1 and 2. The values plotted are those at the time immediately after the cessation of activation. The gamma dose rate has been calculated at a distance of one metre assuming a point source.

A variety of progeny result from each candidate PEG, with increasing numbers from higher-Z species.



Fig. 1. Activity from progeny of candidate PEGs for different activation times.



Fig. 2. Gamma dose rate from progeny of candidate PEGs for different activation times.

Figures 3 to 7 show the highest levels of radioactive progeny from each of the candidate PEGs, respectively nitrogen, neon, argon, krypton and xenon. These are plotted to show how the evolution of each progeny develops over time after activation. This then presents the activity present in the tritium plant, assuming these isotopes are sent immediately to plant. These are shown here for an activation time of 500 s, although other times were output from the model, as indicated in Figure 1. An

estimate of activity from other activation times can be gained from comparison of Figures 3 to 7 with Figure 1.



Fig. 3. Activity vs time from progeny of nitrogen.



Fig. 4. Activity vs time from progeny of neon.



Fig. 5. Activity vs time from progeny of argon.



Fig. 6. Activity vs time from progeny of krypton.



Fig. 7. Activity vs time from progeny of xenon.

Finally within the results presented here, Figure 8 shows how the gamma dose rate falls over time following a 500-s activation period.



Fig. 8. Decay of gamma dose rate from progeny of candidate PEGs from a neutron activation of 500 s.

4. Discussion

The following discussion is based upon the normalised number of 2×10^{21} atoms of injected PEGs, with an activation time of 500 s, as presented above.

Nitrogen, as shown in Figure 3, produces progeny which are all beta emitters. ${}^{13}_{\Box}N$ is the most active of these

with a half-life of around 10 minutes. The progeny following the decay of the four nuclides shown are all stable. The gamma dose rate from activated nitrogen is the lowest of all candidate PEGs, see Figure 8, at less than 0.1 μ Sv/h, and falls rapidly below this after about one hour. Figures 1 and 2 show that there is very little increase in activity and in dose rate from neutron exposures longer than about 1000 s.

Nitrogen use within fusion devices is known to create ammonia. This is difficult to pump, readily adhering to surfaces even above its boiling point, and its existence in a tritium environment would create tritium-containing isotopologues, potentially increasing the tritium inventory in DEMO beyond acceptable levels. Ammonia can also form nitric acid.

Neon has initially the highest activity for activation times less about an hour, and the highest gamma dose rate for activation times of up to about two days, out of all candidate PEGs. However, activity falls rapidly; 20 minutes after activation the total activity and gamma dose rate from neon progeny fall by ten orders of magnitude. For longer activation times, progeny from krypton and xenon give a higher activity and gamma dose rates. As with nitrogen, the progeny from the activation products are all stable.

One of the products of activated neon is fluorine. In a hydrogen isotope environment, the production of hydrofluoric acid is therefore possible.

Argon initially has a relatively high gamma dose rate compared to other candidate PEGs. Most of the progeny are beta emitters, with some also emitting MeV gammas. Once the shorter-lived isotopes of ${}^{40}_{\Box}Cl$ and ${}^{37}_{\Box}S$ have decayed, activity is dominated by ${}^{41}_{\Box}Ar$, a 1.3-MeV gamma emitter with half-life 1.8 hours which also emits a beta.

Krypton progeny, following activation, consists of a number of gamma and beta emitters with half-lives ranging from sub-seconds to hours. Gamma emission is initially dominated by ${}^{81m}_{\Box}Kr$ ($t_{\frac{1}{2}} = 13 \text{ s}, E_{\gamma}$ = 190 keV), until after about a minute when ${}^{83m}_{\Box}Kr$ ($t_{\frac{1}{2}} = 1.8 \text{ h}, E_{\gamma} = 42 \text{ keV}$) dominates. After the initial decays have occurred, other beta emitters become dominant, most additionally emitting gammas.

Xenon also has a large number of metastable states with 530-keV gamma-emitting ${}^{135m}_{\Box}Xe$ dominant for the first hour, after which the beta-emitting ${}^{135}_{\Box}Xe$ dominates, with a half-life of 9.1 h. A large number of other beta and gamma emitters are also produced. Other progeny from the metastable states are also radioactive, with half-lives varying from days to years.

Xenon has the highest gamma dose rate, out of all candidate PEGs, for activation times greater than about two days.

The point source approximation for the gamma dose rates presented above may underestimate the true value of a dose rate in the vicinity of the DEMO tritium plant if cryogenic pumping methods similar to those used at JET [14] or planned for ITER [15], with large cryopanels, are used. Whilst other methods are being investigated as the baseline design for pumping DEMO [16], cryogenic pumping exists as a back-up technology. Conversely, however, some gamma flux attenuation will exist due to the pipework containing the gas.

Nitrogen therefore appears to be the most favourable PEG in terms of the activity that a tritium plant will have to process. However, the production of ammonia is particularly troublesome and how to alleviate its presence deserves special consideration.

Of the other candidate PEGs, the initially high levels of radioactivity could be ameliorated by temporary storage; tens of minutes would be necessary if neon were to be employed, whereas hours may be needed for argon. Such storage would lead to an increase in the tritium inventory of the tritium plant, which should be minimised from a safety perspective. However, the tritium inventory could be kept low if separation of the PEG and its progeny from the tritium prior to tritium processing would be possible, for example by differential pumping at the exhaust stage of DEMO. Any temporary storage of radioactive gas would obviously still need to be in a shielded area.

4. Conclusion

Injection of PEGs is necessary for the long-term operation of DEMO. The gases will become activated by neutrons from D-T fusion within the tokamak, and these activation products will need to be processed by the tritium plant. The candidate PEGs currently are nitrogen, neon, argon, krypton and xenon. Of these, nitrogen appears safest from a radiological perspective, although the associated problem of ammonia production is important. The effects of activity from the activation of neon and argon could be avoided if temporary storage of these PEGs occurs pre-processing. Activation of krypton and xenon produce large numbers of gamma emitters. Discussion is needed between the tritium plant designers and the DEMO plasma physicists to determine which of the candidate PEGs are least problematic.

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