PAPER

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Waste implications from minor impurities in European DEMO materials

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Abstract

Waste-production predictions for the future demonstration fusion power plant (DEMO) are necessary to produce an accurate picture of the likely environmental and economic costs of radioactive waste disposal at end-of-life. An integrated simulation process combining Monte-Carlo neutron transport simulations, inventory calculations, and extensive and reproducible post-processing algorithms has been used for the evolving European DEMO designs to quantify the time-varying mass inventories in different waste classes for individual regions and components of the reactor vessel, as well as for the reactor as a whole. Waste categories based on UK and French regulations reveal that minor impurities contained in structural steels, particularly Eurofer, as well as in functional materials such as tungsten, and beryllium, can have a significant impact on their waste classification prospects. Predictions for current European DEMO concepts suggest that there may be an issue in disposing of fusion structural-steel waste as low-level waste (LLW) in near-surface repositories. Detailed analysis of the subtleties of these predictions, particularly with regard to the production of long-lived radionuclides such as ^{14}C and ^{94}Nb, reveal that the threshold for acceptance as LLW is only just exceeded in some situations. Several mitigation approaches are discussed in this context.

The computational framework developed for these assessments can be rapidly and continuously applied to the maturing DEMO design, helping to guide design choices to mitigate long-lived waste production and ensure that most waste becomes LLW or better within a few decades.

Keywords: radioactive waste classification, fusion DEMO design, neutron transport and inventory calculations, activated steel in fusion, material impurities, tungsten, tritium breeding materials

(Some figures may appear in colour only in the online journal)

Abbreviations used in this paper:

DEMO	— DEMOnstration fusion power plant	EOL	— (DEMO) End-Of-Life
VV	— (reactor) vacuum vessel	MCNP	— Monte-Carlo neutron-particle transport code
WCLL	— water-cooled lithium-lead breeder blanket	CAD	— computer-aided design
LLW	— low-level (radioactive) waste	CSA	— Centre Stockage de l'Aube waste facility
HCPB	— helium-cooled pebble-bed breeder blanket	SS316	— stainless steel type 316
ILW	— intermediate-level waste	wppm	— weight parts per million
HCLL	— helium-cooled lithium-lead breeder blanket	IG	— ITER-grade material composition
		appm	—atomic parts per million

1. Introduction

One of the desirable features of nuclear fusion energy production is the relatively benign nature of the waste products from the fusion reaction itself. This contrasts strongly with the situation in the nuclear fission industry, where the waste associated with the fission fuel cycle is extremely radioactive for thousands or millions of years. The primary solution for dealing with such high-level waste is disposal in deep geological repositories [1], so that it can be isolated on the million-year timescale [2]. In fusion, such dangerous long-lived radioactive products are not produced during the deuterium–tritium plasma burn.

On the other hand, the resulting high-energy neutrons emitted from the plasma must be slowed (moderated) and captured (absorbed) in the walls of the containment vessel surrounding the plasma—both to extract their energy as heat and hence to generate electricity, but also to breed more tritium to sustain the fusion reaction. Unfortunately, the interaction of these neutrons with the materials of the reactor causes those materials to become radioactive (most commonly by the direct absorption of a neutron in the atomic nucleus of a stable isotope to produce a heavier unstable daughter). This process also occurs in the structural materials of a fission reactor but the resulting intermediate-level waste is of secondary importance compared to the hazardous high-level fuel waste, which typically accounts for 95% of the radioactivity despite being only 3% of the waste volume [3].

The current ideal scenario in fusion energy research is for the next-step demonstration fusion power plant (DEMO) to avoid generating any long-term, higher-level radioactive waste and thence to make the licensing requirements and regulatory constraints much less than compared to fission [4]. DEMO must be designed, via appropriate choice and design of materials, shielding configurations, etc, so that the majority of waste produced becomes suitable, after an initial 50–100 years of storage [5], for disposal as low-level waste in shallow repositories. These DEMO design choices must rely on simulated predictions of waste production, including the time evolution of its severity and mass/volume. Even though the design uncertainties are likely to be quite significant while DEMO is still conceptual, the approximate predictions can nonetheless highlight potential problems, such as the unforeseen production of long-lived activity from a candidate fusion material in a particular reactor component, and thus indicate where further design effort is required.

This paper presents the latest results from an integrated simulation process that combines Monte-Carlo neutron transport simulations, high-fidelity inventory calculations, and extensive and reproducible post-processing algorithms to quantify the time-evolution in radioactive waste. This work builds on earlier work by the authors [6, 7], which were amongst the first detailed whole-reactor waste assessments for DEMO fusion reactor designs. Limited fusion radioactive waste assessments have been performed previously for DEMO designs (e.g. [8, 9]), other fusion experiments, such as the Joint European Torus (JET) [10] and ITER [11], and other concepts (e.g. for IFMIF [12]), but the present works are part of a coordinated effort within the European programme to perform assessments efficiently and continuously as the design of DEMO evolves. For this reason, the work described in this paper (and previously [6, 7]) focussed on creating automated computational methodologies and effort is continuing to improve this further. The detailed interrogations described below exemplify the flexibility and the (relative) ease with which such analysis can now be performed.

For future iterations of the DEMO concept, the waste assessment results that feedback to evolve the design will not be restricted as in earlier works to, for example, considering only one material's waste prospects (e.g. for W [13]), at only specific reactor components (e.g. the blanket modules [9]), or at the prospects of a single DEMO concept (as in earlier European projects [8, 9]). This flexible and rapid assessment objective has been further extended in the present work, whereby it is now straightforward to consider the relative waste disposal prospects of DEMO concepts under different regulatory systems and for different waste repositories. Waste severity of radioactive material is assessed (categorised) according to nuclear regulations currently applied in the UK and France, and results are presented for the latest conceptual designs of the European DEMO design programme.

The computational approach used in the assessment is briefly described in the next section, and the subsequent results section focusses on scenarios that demonstrate the issues that can arise due to minor constituents of fusion materials. These impurities may be present unavoidably due to natural contamination of raw materials or as a consequence of manufacturing, or intentionally to create an improved material performance. The implications that impurities have on the waste disposal prospects of a material after it has been exposed to fusion neutron irradiation environment for a prolonged period need to be properly investigated. Such analysis could lead to re-evaluation of certain materials currently used in DEMO designs, perhaps via a refinement of manufacturing processes to reduce an unintentional impurity, or by planned compositional adjustment to remove a problem impurity whilst maintaining performance. Alternative mitigation, including alteration of DEMO operation schedules or planning for a 'DEMO-specific' waste disposal facility, are also discussed.

2. Methodology

The simulation and processing schema used to predict waste evolution has been described in detail previously [6, 7]. Briefly, the three main steps are:

(i) Neutron transport simulations are performed for a DEMO design. This involves using a computational model of the design in a time-independent simulation, where particles (neutrons) are transported one-at-a-time using Monte-Carlo-based random selection to create variation between different neutrons. Data from the separate neutron 'histories' are combined (tallied) to build-up statistical predictions of the neutron environment (fluxes and energy spectra) in the reactor. The widely used MCNP [14] transport code was used for the present work with

a DEMO model based on the CAD geometry of the 'EU DEMO1 2015' design [15]—see [7] for further details, including a description of the different tritium-breeding concepts that can be considered with this design. Figure 1 shows a typical MCNP geometry used in the simulations, showing the reduced 10° toroidal extent of the model. It would be computationally too expensive to simulate the full 360° torus and the symmetry of the problem makes this unnecessary. Various in-vessel and ex-vessel components are labelled in the figure, including the tritium-breeding blanket modules, which in this case are those of the helium-cooled pebble-bed (HCPB) concept. The DEMO design program in Europe is also considering concepts for a water-cooled lithium-lead (WCLL—also considered in the present paper) and helium cooled lithium-lead (HCLL), which only differ from the HPCB model shown in the material composition and geometry design of the blanket modules.

(ii) Nuclide inventory simulations are used to evolve material compositions. The neutron flux spectra (one for each region of the model) from the transport simulations provide the input irradiation conditions to simulations with the FISPACT-II [16] inventory code, which solves coupled differential equations governing the time-dependent concentration of each nuclide. Initial material compositions (based on either the homogenised material compositions used in the Monte-Carlo simulations above, or based on appropriate nuclide concentrations of specific fusion materials) were evolved for a series of irradiation and cooling steps that reflect the current operational plan for a European DEMO, which is a 22 year long, two-phase, pulsed operation with several maintenance ('shutdown') periods (see [6, 17] for more detail, as well as discussions elsewhere [5, 18]). End-of-life (EOL) cooling periods of up to 1000 years were included in the simulations to chart the inventory evolution for waste assessment purposes. At each recorded time-step of irradiation or cooling, FISPACT-II outputs the composition of the material at that point in time as well as the derived total activity, decay-heat, gamma dose, etc, based on the decay parameters of the unstable nuclides in the composition.

(iii) The inventory results are analysed and combined to define the classification of materials as a function of time. For the full reactor analysis, automated post-processing routines have been developed to read FISPACT-II outputs and attribute the appropriate mass of each simulated region of the DEMO geometry to the appropriate waste class according to the time-evolving activity after final shutdown (EOL). The processing can consider limits based on the total activity (separated into α, β, γ contributions as required) or for limits of specific radionuclides—an approach made possible by the detailed nuclide-specific output produced by FISPACT-II (see [19] for further illustration of this). The same processing can also be used to predict the waste classification of arbitrary materials, or of specific fusion materials that only appear as part of homogenised mixtures in the simplified MCNP transport geometry, such as the steels used in blanket modules; subject to the running of appropriate extra inventory simulations for the specific nuclide composition of interest in step (ii) above.

Note that in these conceptual assessments it is also assumed, optimistically, that 100% of any tritium present in the blanket breeding zones will be removed before components are released for waste disposal. Near complete removal will be necessary to maintain the (re)fueling cycle of DEMO and subsequent fusion power stations, but is unlikely to be achieved in reality. Even if less than 1% of the tritium (^{3}H) remains, this could still have significant waste categorization implications since many storage repositories have strict limits for this nuclide despite its relatively short, 12.3 year half-life.

3. Results

Figure 2 displays the total neutron fluxes in units of n cm^{-2} s^{-1} as a function of DEMO location in a colour-mapped toroidal slice through the HCPB DEMO model. The totals shown are sums over the energy-separated flux tallies recorded by MCNP, which are the necessary inputs for FISPACT-II to multiply with energy-dependent nuclear reaction cross sections (probabilities in cm^2 units) and thence to define reaction rates (reactions per second). A similar plot (not shown) of the statistical uncertainties shows that these are less than 5% for all in-vessel components, while some of the deeper outboard (opposite side of the plasma chamber to the centre column—see figure 1) vacuum vessel (VV) regions have somewhat higher uncertainties, but these are still acceptable for the purposes of estimated waste assessment.

The highest levels of neutron flux (greater than 1×10^{14} n cm^{-2} s^{-1}) are within the plasma chamber and in the most-exposed in-vessel components including the divertor and blanket-module armour. The vacuum vessel and the equatorial port plugs act as shields reducing the ex-vessel neutron fluxes to the order of 1×10^9 n cm^{-2} s^{-1} in the region below the equatorial port plug and to around 1×10^8 n cm^{-2} s^{-1} above it. Notice, however, that in this model there is significant neutron leakage though the port, illustrating the conceptual nature of the current designs but also potentially suggesting that the walls of the lower port should be thickened.

FISPACT-II simulations performed using the homogenised, average material starting compositions in each region of the MCNP model have been used to predict the waste classifications of those regions of DEMO. Various different waste category regulations exist around the world, and the study, applicability and comparison of these for potential fusion-waste disposal is the subject of ongoing European studies. In the present work, two different country regulations are considered—chosen to highlight different approaches to waste classification. One applies global activity limits to categorize waste, the other applies limits on individual radionuclides. The first is the general waste classification system applied under UK regulations [20, 21], where low-level waste (LLW) must satisfy *total* limits of 4 MBq kg^{-1} for α-activity and 12 MBq kg^{-1} for combined $\beta + \gamma$ emissions. Any material with activity above these limits would be considered as

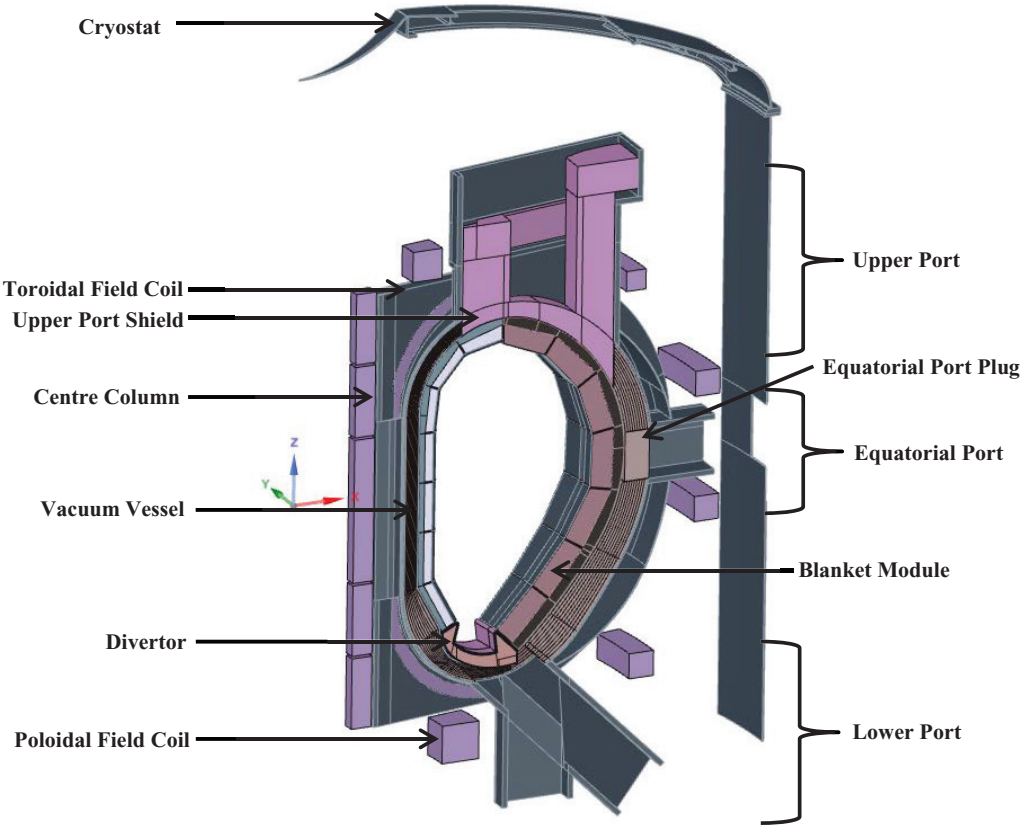

Figure 1. MCNP model geometry of the helium-cooled pebble-bed (HCPB) DEMO design concept. See figure 3 for an indication of the scale of the reactor.

intermediate-level waste (ILW) (or worse), and would not be eligible for LLW waste facilities. This regulatory system is particularly relevant for fusion as it has been used to plan the decommissioning and waste handling of JET [10], which is sited in the UK and is the largest fusion experiment so far constructed. The JET experience, and hence UK regulations, are also useful in the planning of waste management for ITER [21]—the next big experiment in magnetic confinement fusion research.

A second categorization was defined according to the *nuclide-specific* limits for France's main LLW disposal facility at Centre Stockage de l'Aube (CSA) at Soulaine-Dhuys, Aube département, in eastern France—a repository that will handle the low and intermediate level waste from ITER [22]. Limits for individual radionuclides are defined according to their half-lives, decay type, and general radiotoxicity, and more than 40 nuclides have defined limits [23, 24]. For the present studies, only a few of these are relevant for the materials envisaged for DEMO, including the following with their associated *Limite Maximale d'Admissibilite* (LMA):

- ^{14}C—9.2×10^4 Bq g^{-1}
- ^{94}Nb—1.2×10^2 Bq g^{-1}
- ^{63}Ni—3.2×10^6 Bq g^{-1}
- ^{59}Ni—1.1×10^5 Bq g^{-1}
- ^{60}Co—1.3×10^8 Bq g^{-1}
- ^{93}Mo—3.8×10^4 Bq g^{-1}.

Note that the regulations for this French facility, and for other facilities around the world, also consider secondary characteristics when classifying the acceptance of radioactive waste, including the waste origin, chemical composition, and reprocessing potential [25]. This will be discussed again later.

Waste can also be classified as non-active (NAW) if it meets the IAEA clearance criteria [26], but little of the DEMO device, and certainly none of the in-vessel or VV region, satisfies this in the current simulated predictions.

Figure 3 shows a set of toroidal slices through the DEMO model geometry, illustrating how long different regions of the reactor take to satisfy the LLW criteria of the two sets of acceptance limits. Figures 3(*a*) and (*b*) are cross-sections of the HCPB-blanket DEMO design, with regions coloured according to the time-window (measured from DEMO EOL) during which the material in that region is predicted to be acceptable as LLW according to the UK and France limits, respectively. Figures 3(*c*) and (*d*) show equivalent results for the WCLL-blanket concept, but notice that the cross-sectional slice in these cases is at a different toroidal angle to that used for the HCPB slices—these WCLL slices include the port regions (as labelled in figure 3(*c*))

For the breeder blanket modules of the HCPB concept the results in figure 3 are not very favourable—all such zones are predicted to remain as intermediate-level waste for more than 1000 years beyond DEMO EOL under both classification systems, but for different reasons (see later, section 3.2). In

the WCLL concept the predictions appear to be more favourable—under UK regulations most of the blanket regions are predicted to be LLW within 100–300 years. However, in this concept, the lithium-lead (Li–Pb) breeder material included in the homogenised blanket material makes up a relatively larger proportion of the total material mass than compared to the much lighter beryllium and lithium-orthosilicate mixture of the HCPB concepts (see [6]). In both cases the majority of the long-term activity originates from the 10–15 volume % Eurofer steel in the material mixture, and it is clear that using activity per-unit-mass limits on these mixtures produces a skewed result in the WCLL, Pb-mass-dominated concept. Since the majority of these 'functional' materials are likely to be recycled rather than being sent for disposal, and in either case will be largely separated from the structural (steel) materials, it is more instructive to consider the waste categorization of the steel in isolation.

Figure 4 shows the same toroidal cross-sections as figure 3, but this time regions are coloured according to the LLW acceptance times of the isolated structural steels in that zone. For the in-vessel components Eurofer is the assumed steel in the design, while stainless steel 316 (SS316) is used in the vacuum vessel and other ex-vessel components (SS304 is also present in some ex-vessel regions)—typical compositions for these two primary structural steels (used in the FISPACT-II calculations) are given in [6]. This new, steel-only assessment required additional FISPACT-II inventory simulations for each relevant cell of the geometry (one for each breeder concept) and assumes that the neutron flux spectra would not be strongly altered in the presence of fully-separated materials (i.e. no new MCNP calculations were performed), which is a reasonable assumption in most cases. However, it is still recommended to use as much detail as possible in MCNP designs as there could be localised variations that cannot be accounted for in the current homogenised models. For example, in regions near to water-filled cooling pipes there could be an influence on the neutron spectra in surrounding material due to the highly moderating nature of water. Detailed investigations elsewhere [27] have shown the importance of heterogeneity in the case of the tungsten armour in DEMO, but there is not yet enough detail in blanket designs to make an accurate assessment for the DEMO vessel in general and this is beyond the scope of the present work.

Regions that do not contain steels, which are mainly tungsten armour regions, are omitted from this material-specific analysis but, in any case, such regions are largely invisible on the reactor scale of figure 4.

Figure 4 shows steel-assessment results for the HCPB and WCLL concepts according to both the UK and France regulations. In contrast to previous works [6] and to the results in figure 3, now the prospects for the WCLL concept are not that different to those of the HCPB design, with all of the blanket regions not acceptable as LLW under either system within 1000 years. Notice that the HCPB results in figure 4 are almost indistinguishable from those in figure 3 because the change in activity-per-unit-mass values are small in the steel-mass-dominated blanket modules of that design. The above result clearly illustrates the need for care when using conceptual

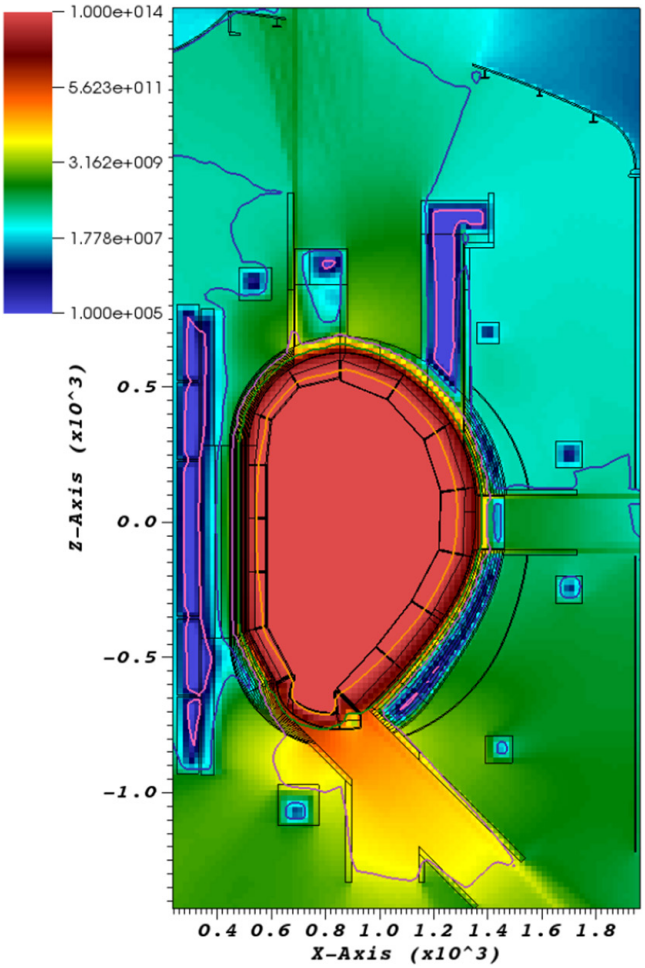

Figure 2. Toroidal slice of the HCPB DEMO model. Regions (cells) are coloured according to the predicted total neutron flux during plasma operation. The fluxes, as defined by the colour legend, are measured in neutrons cm^{-2} s^{-1}. The vertical ('z-axis') and radial ('x-axis') positions are given in 10^3cm units.

DEMO designs (converted from CAD to MCNP input, or otherwise) to make large scale predictions such as waste classification—here we have shown that the characteristics of the material assessed, particularly its density and volume (for waste regulations that apply activity-per-unit-volume limits), should realistically reflect the material released for waste disposal. For this reason, all remaining results in this paper consider isolated materials at their nominal density, and not the homogenised material mixtures assumed for the production of figure 3.

Note that in the HCPB concept (figures 4(a) and (b)) the MCNP geometry was modified to investigate the impact of radial sub-division of both the breeder blanket and divertor body zones on waste categorization, which has previously been shown to produce beneficial waste disposal prospects for the VV (see [7]). The VV in the DEMO designs considered here includes this sub-division and figures 3 and 4 demonstrate the benefit. However, for the blanket, only in figure 4(a) is there any obvious change from the fully homogenised blanket—in a few of the outer blanket layers in the lower, outboard region there are improved prospects (reduced acceptance times) for the Eurofer steel used in these regions.

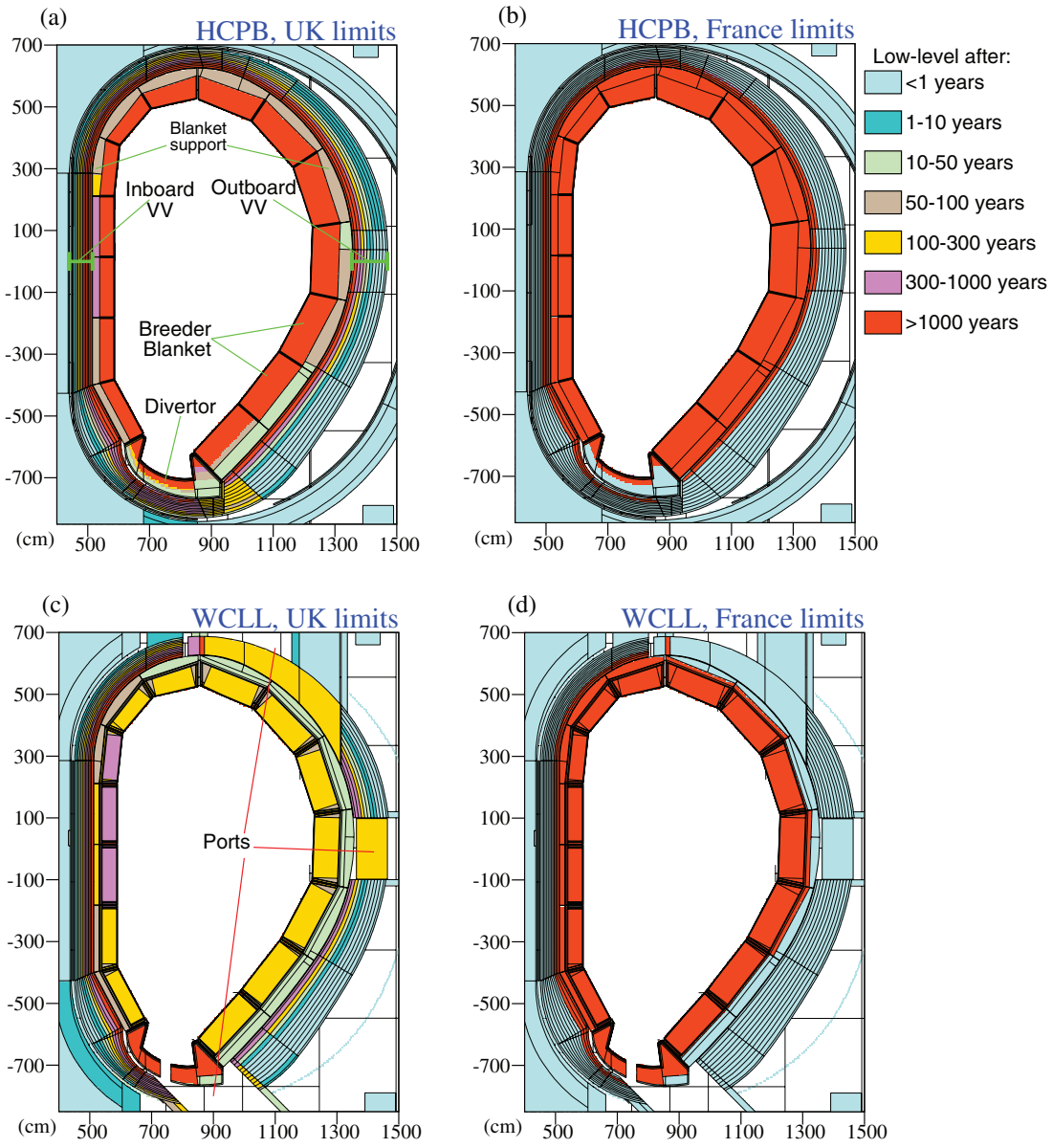

Figure 3. Toroidal slices of the DEMO model with ((*a*) and (*b*)) HCPB and ((*c*) and (*d*)) WCLL breeder blanket concepts. In each cross-section, homogenised material regions (cells) are coloured according to the time-interval (shown in the key and measured from DEMO EOL) during which the material is predicted to satisfy the criteria to be classified as low-level waste (LLW) according to ((*a*) and (*c*)) UK or ((*b*) and (*d*)) French regulations.

In contrast to the authors' previous assessment for the VV [7], which recommended designing a VV that could be radially separated before disposal, there does not appear to be a significant similar benefit for the blanket modules at the current level of modelling detail.

On the other hand, the predictions reveal a potential benefit for the divertor body. Note that divertor activation results for a model with either a HCPB or WCLL blanket should be indistinguishable because the divertor design is the same. However, comparing figures 4(*b*) and (*d*), where the latter does not have the radial sub-division in either the blanket or divertor, shows there is a potential improvement in the waste prospects at the rear (bottom) of the divertor; some of the outer parts of the body may be immediately LLW-disposable. The exact origin of this result will be explored later (see section 4).

In contrast to the breeder blanket, deeper radial components of the WCLL design often have shorter LLW acceptance times compared to similar regions in the HCPB concept. For example, in figure 4(*b*), for HCPB under French regulations, all of the blanket support (shield) regions remain above LLW limits for more than 100 years, while for the WCLL concept (figure 4(*d*)) the majority of outboard blanket support regions could be accepted at the CSA facility immediately after the DEMO experiment has finished. This stark contrast is related to the slow growth (build-up) of long-lived radionuclides in Eurofer steel, which will be discussed in the following sections. Similarly, some of the inner VV regions are predicted to be 'LLW-accepted' sooner behind the WCLL blankets than behind the HCPB ones—again due to specific levels of long-lived radionuclides in SS316. The radionuclide origins of

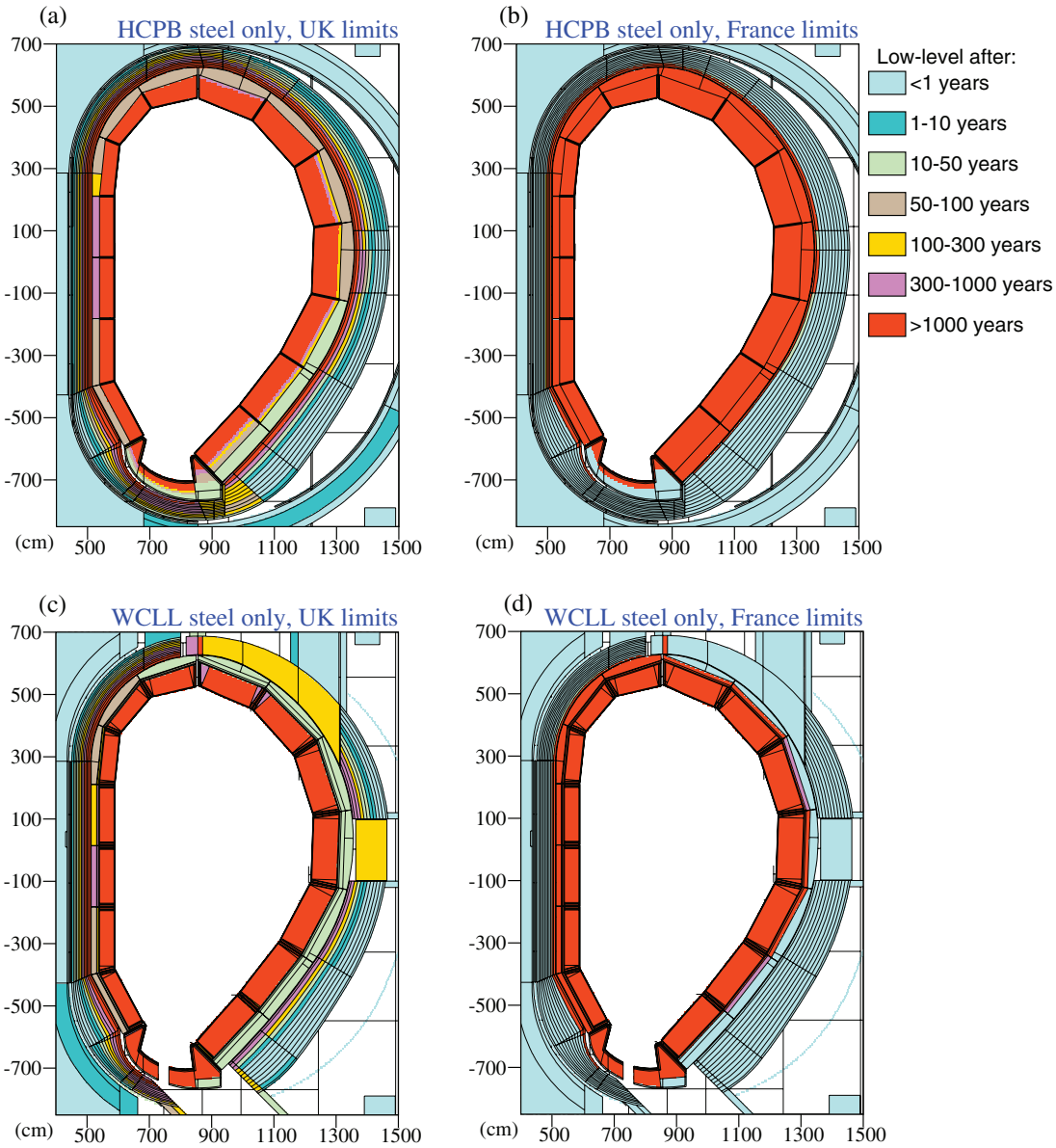

Figure 4. Toroidal slices of the DEMO model with ((*a*) and (*b*)) HCPB and ((*c*) and (*d*)) WCLL breeder blanket concepts. In each cross-section, regions (cells) are coloured according to the time-interval (shown in the key) during which the steel in that region is predicted to satisfy the criteria to be classified as low-level waste (LLW) according to ((*a*) and (*c*)) UK or ((*b*) and (*d*)) French regulations.

these results in SS316, Eurofer, and detrimental waste prospects in selected function fusion materials are explored in the next sections.

3.1. 316 stainless steel

In the above global reactor analysis it was observed that many of the inner (closest to the plasma) layers of the radially subdivided VV (into 10 equal 'interspace' layers contained within inner and outer shell layers [7]) were predicted to remain above LLW limits under UK or France regulations for more than 1000 years. For the HCPB concept this was true regardless of whether the SS316 stainless steel of the VV was considered in isolation at its full density ($\sim$8 g cm^{-3})—see figures 4(*a*) and (*b*)—or as part of reduced-density, homogeneous volume mixes of water, 316 and vacuum in the interspace—figures 3(*a*) and (*b*).

Radionuclide breakdowns of the $\beta + \gamma$ activity for SS316 in different layers of the inboard (inner radius of the DEMO torus, adjacent to the centre column in figure 1) equatorial VV in the HCPB model in figure 5 reveal the origin of this prediction. Activity is plotted as a function of decay time beyond EOL on both a logarithmic (figure 5(*a*)) and linear (figure 5(*b*)) year-scale to make it easier to observe, respectively, both the shorter-term (less than a year) activity levels that are relevant for maintenance operations, and the long-term behaviour that determines the waste disposal prospects. As well as the total activity in the material the contribution curves from individual important radionuclides are shown. These 'nuclide contribution' plots are now part of the standard

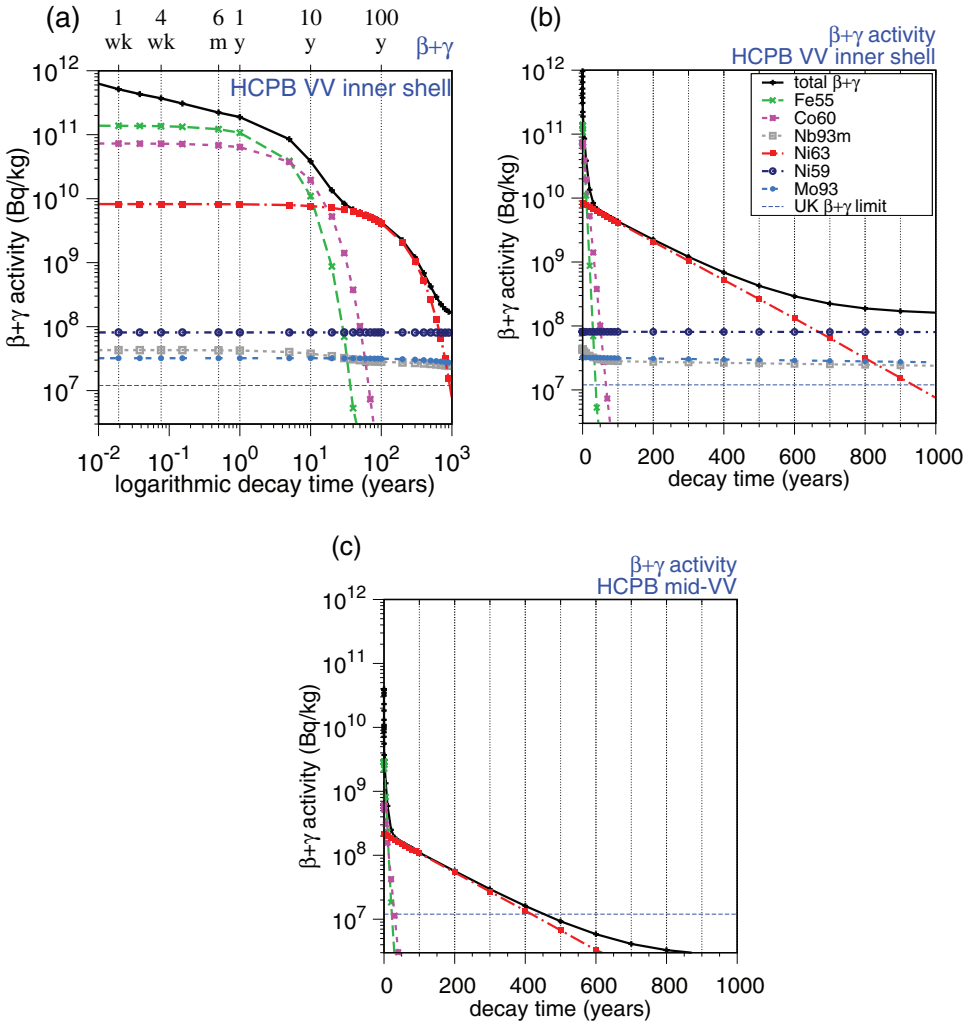

Figure 5. Nuclide contributions to SS316 activity under the predicted conditions in the inboard equatorial VV behind the HCPB breeder blanket concept. In (*a*) the variation in activity for the VV inner shell (nearest the plasma) is shown against a logarithmic decay timescale (measured from DEMO EOL), while in (*b*) a linear timescale is used for the same VV zone. (*c*) The activity decay profile (on a linear time *x*-axis) for a middle layer of the VV interspace (between the inner and outer shells). In each plot the total activity is shown, together with curves representing the contributions from important radionuclides. The UK-specific LLW limit for $\beta + \gamma$ emissions (none of the dominant radionuclides in this case are alpha emitters) is also shown as a horizontal dashed line.

output provided by FISPACT-II and have proven useful in, for example, interpreting the performance of simulations relative to experimental decay-heat measurements [19].

For the most exposed inner shell of this part of the VV (figures 5(*a*) and (*b*)) the activity of several β/γ emitting nuclides are above the UK LLW limit. The long-lived ^{59}Ni and ^{93}Mo radionuclides with half-lives of 76 000 and 4000 years, respectively, and the shorter-lived ^{93m}Nb ($T_{1/2} = 16$ years) produced by the decay of ^{93}Mo, all produce a near-constant decay-activity output exceeding the UK 12 MBq kg^{-1} limit. However, deeper into the vacuum vessel, at the middle of the interspace, the activity from these three radionuclides is far below this LLW limit (and not visible in figure 5(*c*)). In this case, the only nuclide determining the predicted acceptance time of SS316 is ^{63}Ni and this decays with a half-life of 101 years, leading to a LLW-acceptance time of around 450 years (figure 5(*c*)).

Using the France activity limits for the CSA facility, on the other hand, it is a different radionuclide that impacts on the acceptance of VV SS316. The nuclide-specific limits of ^{59}Ni, ^{93}Mo, and ^{63}Ni are relatively high—the lowest, for ^{93}Mo, is still more than three times the total $\beta + \gamma$ limit for the UK criteria. Instead, ^{94}Nb, with a 20 000 year half-life, has a low, 120 kBq kg^{-1} limit, which is exceeded in most of the VV inner shells, and in several layers of the interspace, behind the HCPB blanket modules.

As discussed in the previous section, behind the WCLL breeder blanket the predictions for the VV are more favourable due to the lower levels of production of the problematic radionuclides highlighted above, and all of the outboard VV, in particular, is predicted to be immediately (at DEMO EOL) releasable as LLW.

3.2. Eurofer

^{14}C production in Eurofer steel is the primary cause of the result for the blanket regions under the UK-based waste classification system in figures 4(*a*) and (*c*). As was discussed in

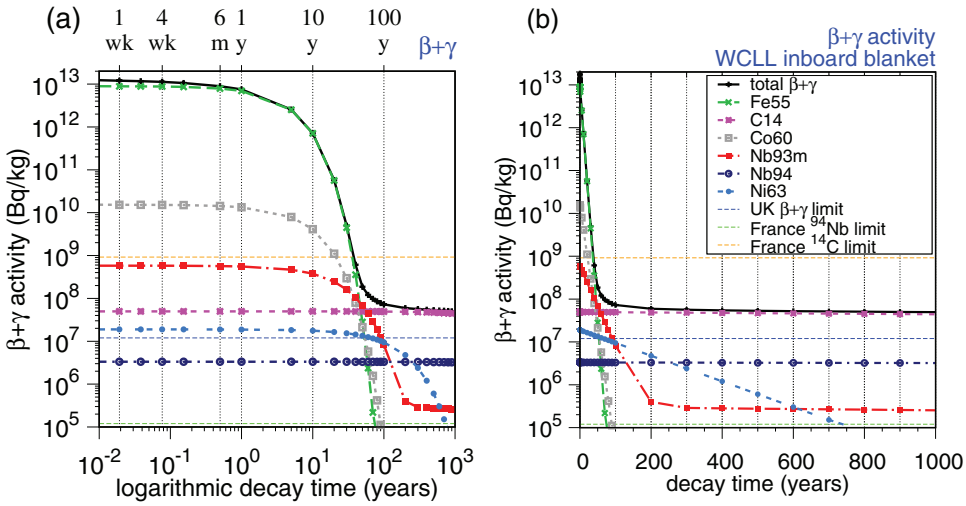

Figure 6. Nuclide contributions to Eurofer activity under the predicted conditions in the inboard equatorial breeder blanket zone of the WCLL concept. In (*a*) the variation in activity is shown against a logarithmic 'decay-time since DEMO EOL' scale, while in (*b*) a linear timescale is used. The total activity is shown, together with curves representing the contributions from important radionuclides. The UK-specific low-level waste (LLW) limit for $\beta + \gamma$ emissions and the France CSA repository limits for ^{12}C and ^{94}Nb are also shown as horizontal dashed lines.

[7], the ^{14}C originates from (n,p) (neutron capture followed by proton emission) reactions on the ^{14}N isotopes (99.6% of natural nitrogen) from the few hundredths of weight % nitrogen in Eurofer steel, which improves its high-temperature strength through nitride precipitation [28]. In earlier DEMO activation studies (see, for example [4]) ^{14}C production from Eurofer was not highlighted as an issue because in those cases the focus was on γ-dose for recycling and remote-handling considerations. However, for radioactive waste disposal and storage in underground, near-surface repositories β and α emissions must also be considered and limited appropriately. Since ^{14}C is a pure β emitter it does not contribute to γ dose, but it contributes significantly to the total $\beta + \gamma$ output and is solely responsible for Eurofer exceeding the UK-limit LLW considered here at DEMO EOL in many locations within the blanket of either the HCPB or WCLL concepts.

Figure 6 presents the post-EOL activity of Eurofer after being subjected to the predicted conditions in the inboard equatorial breeder blanket region of the WCLL model (the highest flux zone for the blanket modules in that model) during the ~16 year schedule of the second phase of planned DEMO operation [17] (blanket replacement is foreseen after an initial ~5 year phase-1 campaign). As was the case for SS316 (figure 5), there is the usual dominance of ^{55}Fe during the first few decades after shutdown (produced via (n,2n) reactions on the primary ^{56}Fe isotope), but then, at longer times, the near-constant contribution from ^{14}C with its 5715 year half-life, which, in this case, is clearly above the UK-LLW $\beta + \gamma$ activity limit (also shown in the plots for reference).

^{14}C appears as a particular problem under the UK classification system because those regulations do not have separate limits for different nuclides—the total radionuclide inventory must satisfy the same global activity limits. For waste categorization systems with nuclide-specific limits, such as the French limits considered here for the CSA facility, ^{14}C has a generous limit reflecting its relatively minor radiotoxicity. The specific ^{14}C activity limit for the near-surface disposal CSA facility of 9.2×10^7 Bq kg^{-1} is also shown in figure 6. For Eurofer in this blanket zone the ^{14}C activity is well below the French limit. However, the level of ^{94}Nb activity is again, as it was for SS316, above the low limit for this radionuclide (also shown in figure 6) and so the steel would still not be allowable for near-surface disposal on the 1000 year timescale. In either case—UK or France classification system—extremely small concentrations (less than 0.5 weight parts per million or wppm) of radionuclides produced from minor impurities—0.045 wt.% N and 0.005% Nb for ^{14}C and ^{94}Nb, respectively—are predicted to be responsible for not being able to dispose of Eurofer steel from a DEMO blanket as LLW.

3.3. Beryllium

Figure 7 shows the contributions from different radionuclides to the total α activity of beryllium after an operational life in the inboard equatorial HCPB breeder zone. The 'pure' Be composition assumed in the DEMO model contained 0.01 weight %, or 100 wppm, of uranium (as well as 0.9% O, 0.08% Mg, 0.09% Al, 0.06% Si, 0.01% Mn, 0.1% Fe, and 0.001% Co), which in a ~1000-tonne blanket containing 300–400 tonnes of Be would equate to 30–40 kg of uranium. The contribution from radioactive α-emitting actinide impurities is significant enough to exceed the UK-LLW limit of 4 MBq kg^{-1}. Several nuclides, but in particular by the long-lived ($T_{1/2} \approx 24\,000$ years) ^{239}Pu, exceed the limit.

Actinide impurities are only potentially a problem for the HCPB concept since other breeder blanket concepts use, instead, Pb for neutron multiplication and moderation, where the long-lived radioactive nuclides after neutron irradiation are not significant enough to exceed LLW limits. It is likely that much of the Be used would be extracted for possible reuse rather than being disposed of as waste with the other blanket components, which would largely negate the problem.

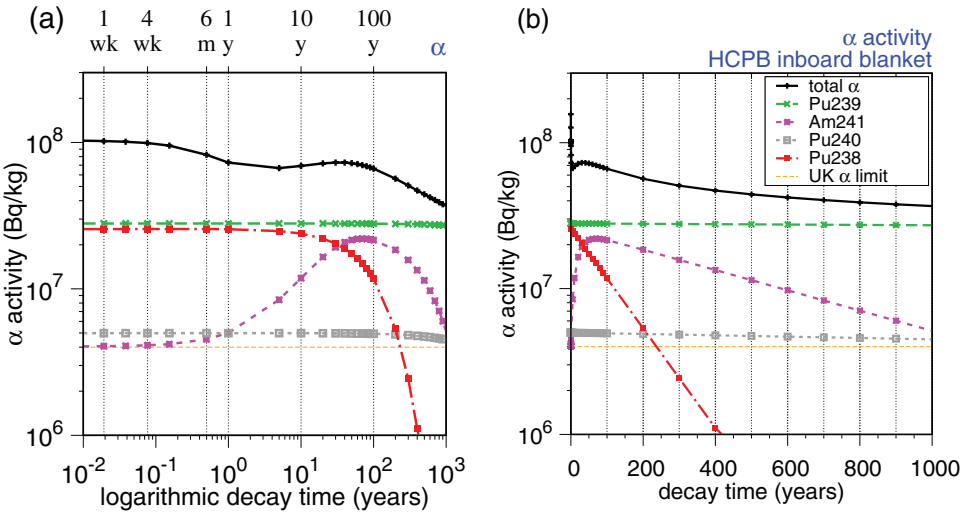

Figure 7. Nuclide contributions to α activity in 'pure' beryllium after exposure to the predicted conditions in the inboard equatorial HCPB breeder blanket zone. In (*a*) the variation in activity is shown against a logarithmic decay-time scale, while in (*b*) a linear timescale is used. The total activity is shown together with curves representing the contributions from important radionuclides. The UK-specific LLW limit for α activity is also shown as a horizontal dashed line.

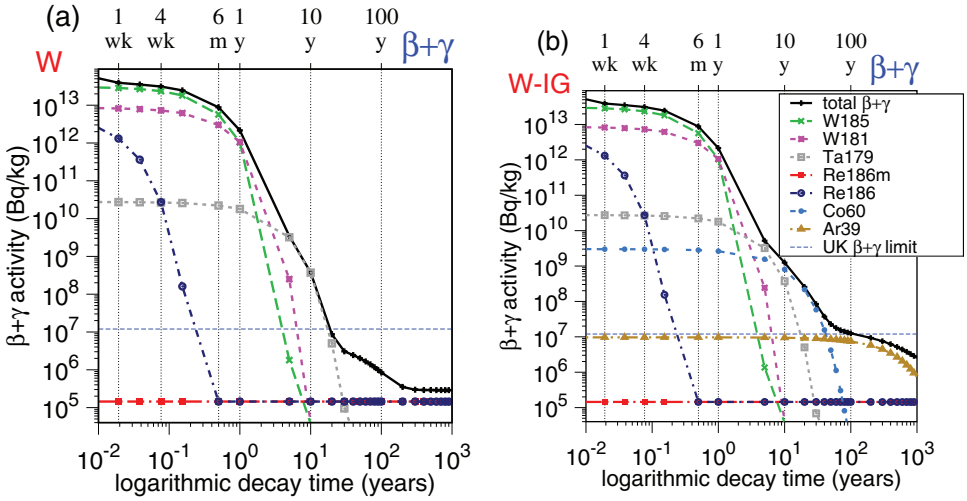

Figure 8. Nuclide contributions to (*a*) pure tungsten and (*b*) ITER grade tungsten after exposure to the conditions predicted for the armour layer at the inboard strike-point of the divertor in the DEMO model. Total activity and curves representing the activity contributions from important radionuclides are plotted against a logarithmic decay-timescale (measured from DEMO EOL). The UK-specific low-level waste (LLW) limit for $\beta + \gamma$ emissions is also shown as a horizontal dashed line.

However, it is still a potentially serious issue that might make this tritium-breeding option less desirable, or at least demonstrate the need for more careful processing and selection of natural Be sources. Uranium concentrations in mined Be can vary significantly and the 100 wppm assumed here is near the upper limit from US and Chinese sources, while Russian deposits often have much lower U content. For example, in [29] the average U content in commercial Be from Russian–Kazakhstan sources was calculated to be only 5.2 wppm. At these levels of uranium impurity the amount of ^{239}Pu activity, which dominates the α-emissions in figure 7, would drop from the ≈28 MBq kg^{-1} shown in the figure to around 1.5 MBq kg^{-1}, and would thus be below the UK-LLW α-limit of 4MBq kg^{-1}.

3.4. Grades of tungsten

Manufacturing impurities in the ITER-grade (IG) tungsten assumed in European DEMO designs can also produce long-lived dominant activation products [6]. Figure 8, shows the decaying $\beta + \gamma$ activity beyond DEMO of both pure W (figure 8(*a*)) and the ITER-grade (figure 8(*b*)) after an operational lifetime (∼5 years) in the predicted neutron spectrum for the first-wall armour of the inboard divertor strike-point. Such results are relatively independent of the breeder blanket concept choice, except at very short decay times [6].

In pure tungsten the total $\beta + \gamma$ activity from contributing radionuclides—primarily ^{186}Re and the long-lived ^{186m}Re ($T_{1/2} = 2 \times 10^5$ years) that decays to it—at long decay times

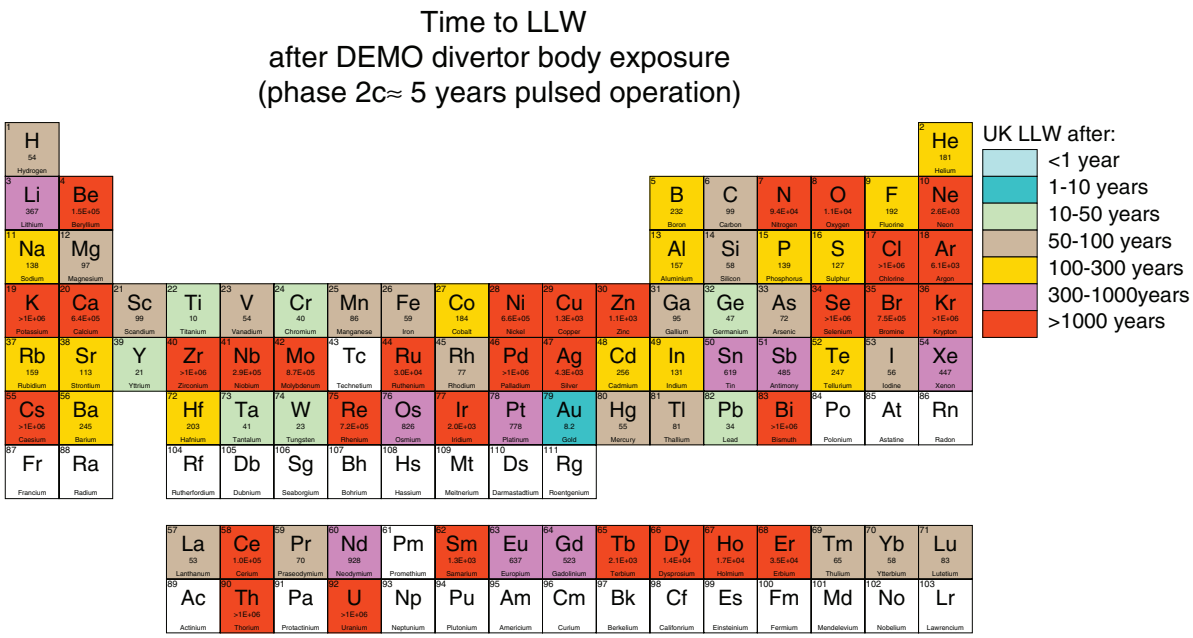

Figure 9. Periodic table with each naturally occurring element coloured according to the time-interval during which it would satisfy the UK-LLW criteria after irradiation during one full operation cycle in the divertor cassette body. The actual 'time-to-LLW' values are given below each element symbol. Note that no tritium removal is considered for this divertor environment, and this radionuclide is responsible for the long-lived waste in some of the lighter elements, such as He, and Li.

is orders of magnitude below the UK-LLW limit. However, in IG-W there is a significant delay in reaching the LLW limit; in pure W under identical conditions the UK-LLW limit is satisfied after around 20 years, while it takes IG-W around 100 years to become acceptable under this limit. Figure 8 shows that this remarkable change in response for a material that is 99.96 wt.% W is caused by the production of primarily ^{60}Co via neutron-capture reactions on the ^{59}Co isotope that makes up 100% of the tiny amount of residual cobalt in the as-manufactured IG-W; the assumed cobalt concentration is 0.001 wt.% or 10 wppm (≈30 appm) leading to around 0.2 appm (0.07 wppm) ^{60}Co at the end of operation.

3.5. Lithium ceramic

In the HCPB design the main tritium-breeding material is lithium orthosilicate (Li_4SiO_4) made up of 23.6 wt.% lithium, 52.6% oxygen, and 23.7% silicon. Calculations show that none of these three main constituents are likely to cause a delay in acceptance of this ceramic as LLW in either the UK or French regulatory systems. However, the engineering composition assumed in the DEMO concept also includes various minor impurities, including 10 wppm potassium, around 90 wppm platinum, and a few wppm of cobalt. The potassium (K), in particular, leads to the formation of ^{39}Ar ($T_{1/2} = 269$ years) via (n,p) reactions, and ^{36}Cl ($T_{1/2} = 3 \times 10^5$ years) via (n,α) (neutron capture followed by α-particle emission); both from ^{39}K (∼93.2 atomic % of potassium).

^{36}Cl has a relatively low acceptance limit (5 Bq g^{-1}) in the CSA facility and its generation in Li_4SiO_4 irradiated in the first few centimetres (radially) of each blanket module region (see figure 1) is predicted to exceed this limit for more than 1000 years. Similarly, and perhaps more severely, production of ^{39}Ar (and to a lesser extent ^{60}Co and ^{193}Pt from cobalt and platinum, respectively) in the present assessment delays acceptance of the *entirety* of the blanket modules as LLW in UK repositories for potentially hundreds of years due to high-levels of β emissions. These findings give a strong incentive to reduce potassium content in manufactured lithium ceramics.

3.6. Elemental scoping

The above fusion material discussions and assessment have highlighted how minor impurities can play a significant role in the waste disposal prospects of fusion materials. As the DEMO design evolves it will be vital to consider concentration limits for minor constituents so that appropriate design choices, changes in materials, and/or refinement of manufacturing processes can be included in the engineering concepts. The methodology developed in this work can be rapidly applied to perform a waste assessment on the full range of possible impurities.

As an example, figure 9 shows a 'time-to-LLW' analysis (UK-LLW) for all possible pure, naturally-occurring elements after an operational exposure to the neutron flux conditions predicted for the divertor cassette body. Since this component of DEMO is primarily composed of the complex Eurofer alloy in the current designs, it is useful to explore the relative impact of impurities. The periodic table tableau in the figure confirms the aforementioned problems with ^{14}C production leading to nitrogen being predicted to take more than 1000 years to become UK-LLW acceptable if irradiated in a pure state. This relative waste assessment indicates which elements are likely to delay the acceptance of a material as UK-LLW if they form part of that material's composition, and the length of decay-time required before a particular element is acceptable

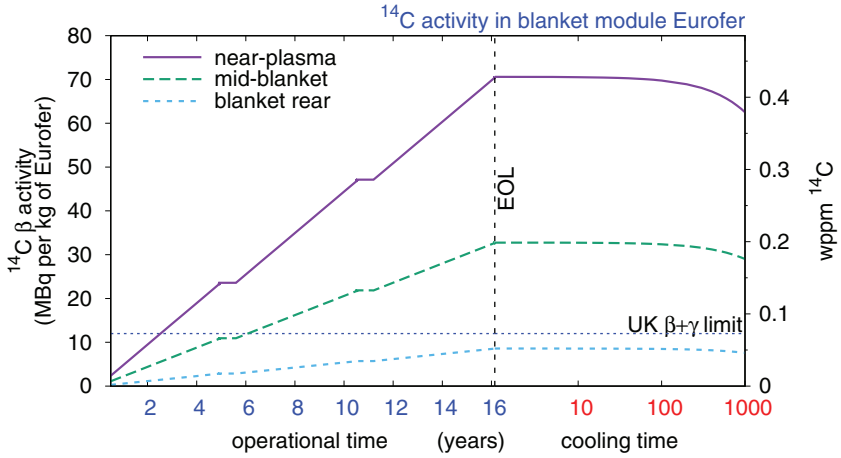

Figure 10. ^{14}C evolution in Eurofer during (DEMO phase-2 operation) and after exposure in three layers of the outboard equatorial HCPB breeder blanket. The threshold between operational exposure on a linear timescale and decay cooling on a logarithmic scale is the vertical dashed line marking DEMO EOL. ^{14}C is plotted as absolute β activity (left y-axis) and wppm concentration (right y-axis). The dashed horizontal line marks the UK-LLW limit for total $\beta + \gamma$ activity. See the main text for details.

suggests the concentration limits for that element, e.g. Ni and Cu are both problematic but pure Cu has a shorter time-to-LLW and so could be allowable to higher concentrations. Of course, fully-detailed inventory simulations of a prototype material composition would be required to be more quantitative, but assessments like that shown in figure 9, which could also be performed for waste classification systems with nuclide-specific criteria, can be used as a guide.

4. Mitigation

A feature of the results in these studies is the strong geometry dependence on the predicted waste severity of certain components. For example, in figure 4(*b*), there are VV layers behind the HCPB blanket where SS316 is predicted to be unacceptable at the French CSA facility for more than 1000 years. Immediately adjacent (behind) these layers there is often the opposite extreme; of layers where the irradiated SS316 can be immediately (at DEMO EOL) disposed of as LLW. Similarly, in the radially sub-divided (ten equal layers in each module) HCPB blanket, figure 4(*a*) showed that some of the rear (furthest from the plasma) layers of the outboard breeder blanket zones could be UK-LLW-accepted after a few 100 years; a timescale on which the problematic long-lived ^{14}C cannot have begun to decay, and so it cannot be the cause of the LLW timescale in those cases. What is the fundamental difference between these blanket layers and the shallower (nearer the plasma) outboard layers where the UK-LLW-acceptance time is more than 1000 years (and thus governed by ^{14}C levels)?

Analysis of ^{14}C production reveals the answer. Figure 10 shows the β-activity from ^{14}C produced in Eurofer during (and after) exposure at three different layers (depths) of the outboard equatorial mid-plane of the HCPB breeder blanket. The left of the plot charts the growth (build-up) of ^{14}C during the ∼16 year phase two operational plan for DEMO [17], when no blanket replacement is foreseen. The right of the plot, beyond the DEMO EOL dashed line, shows the near-constant decay activity from the grown-in ^{14}C during the subsequent 1000 years of Eurofer decay. Since ^{14}C is a pure β-emitter (it has no other decay modes) the calculated activity is exactly proportional (via a simple $A = \lambda C$ relation between concentration C of ^{14}C per unit mass, activity A and ^{14}C decay constant $\lambda = \ln 2/T_{1/2}$) to the concentration, and so the latter is also quantified in the figure.

The figure clearly shows that in the rear layer of the HCPB blanket ^{14}C never reaches the necessary concentration to exceed the UK-LLW $\beta + \gamma$ activity limit. On the other hand, in both the front (closest to the plasma) and mid-blanket layers, the growth-rate of ^{14}C via (n,p) reactions on ^{14}C is sufficient for its activity to exceed the 12 MBq kg^{-1} limit. Once this limit is reached the ^{14}C activity will never fall below it on the 1000 year timescale because of the nuclide's long, 5715-year half-life. Notice, however, that the ^{14}C activity takes around 6 years of DEMO operation to reach the UK-LLW limit in the mid-blanket layer (fifth layer), raising the possibility of adjusting DEMO operational plans to reduce exposure of certain components and thus reducing higher-level waste production. Of course, for the near-plasma layer (layer one immediately behind the armour), the threshold is reached after only 2 years of operation, and it is unlikely that DEMO would be economically viable if the blanket were replaced on that timescale.

Eurofer, like SS316, also contains niobium, albeit at a lower concentration (at least 50% less by weight [6]), and it was already noted (section 3) that there is some threshold under the France CSA facility regulations that is exceeded in Eurofer at the near-plasma side of the divertor body that is not reached at the rear (bottom)—see figure 4(*b*). Figure 11 shows the evolution of ^{94}Nb activity and concentration in four (of ten) different radial layers of the divertor body, which is the bulk of the divertor shown in figure 1—there are three plasma facing layers around 5 cm thick in total and two shell layers around the body of 4 cm each, while the body itself is at least 30–40 cm thick. Also shown in the plot is the ^{94}Nb limit for the France LLW classification. In the bottom layer (10), furthest and most-shielded from the plasma, ^{94}Nb production

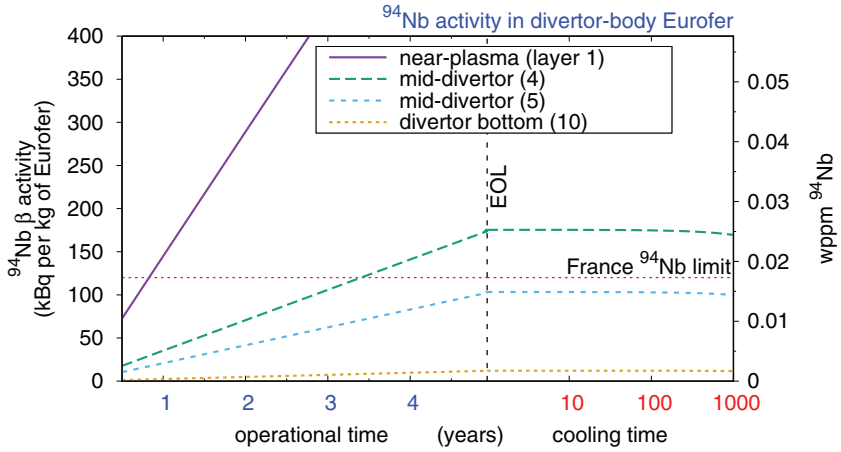

Figure 11. ^{94}Nb evolution in Eurofer during (DEMO phase-2c operation) and after exposure in four (out of ten) layers of the divertor body. The threshold between operational exposure on a linear timescale and decay cooling on a logarithmic scale is the vertical dashed line marking DEMO EOL. ^{94}Nb is plotted as absolute β activity (left y-axis) and wppm concentration (right y-axis). The dashed horizontal line marks the ^{94}Nb limit under the acceptance criteria for LLW at the French CSA facility. See the main text for details.

is far below the France-LLW limit and consequently Eurofer in this layer is predicted to be immediately disposable at the French CSA facility. Conversely, in the first layer, nearest the plasma, the rate of production of ^{94}Nb—via neutron capture (n,γ) reactions on the stable ^{93}Nb of niobium—is so high that Eurofer in this region is predicted to exceed the France-LLW limit within the first year of operation, and consequently would not be disposable as LLW under French regulations for more than 1000 years.

On the other hand, in the middle of the divertor there is a sharp interface between layers 4 and 5 (15–20 cm into the divertor), where ^{94}Nb either does or does not reach the French-LLW-limit during the planned operational life-cycle. As well as suggesting that changing (reducing) the planned operation lifetime of the divertor could produce an improvement in the waste disposal prospects of divertor-Eurofer (and hence VV-SS316 where the same slow, in-growth of ^{94}Nb is observed), the concentration axis in the plot suggests other possibilities.

The difference between falling below or above the limit is of the order of 0.01 wppm ^{94}Nb produced from 50 wppm (30 appm) elemental Nb in the assumed (typical) composition of Eurofer in these simulations [6]. Reducing the starting niobium content by half or more would produce a different result to that shown in figure 11—less of the divertor body would be activated above the French LLW limit. At the same time, it has already been noted that many near-surface low-level nuclear waste repositories around the world take into account the source of the waste when deciding whether it can be accepted. In particular, repositories are often designed to accept a specific waste form, and may have (nuclide) limits that reflect the expected activity in that type of nuclear waste. Rather than assuming that DEMO fusion waste must adhere to limits of current, often fission-orientated, waste repositories, it is feasible to expect a fusion-specific repository to be tailored to the specific type of waste expected from DEMO. Under such circumstances the very low ^{94}Nb limit shown in figure 11 could be relaxed upwards with potentially only minor changes to the shielding characteristics of a bespoke near-surface repository for DEMO waste.

5. Summary and outlook

This paper has presented recent results from the ongoing waste assessment efforts that form part of the European DEMO design studies. In particular, it has highlighted the potential issues surrounding the minor impurities in structural steels used for in-vessel (Eurofer) and vacuum-vessel/ex-vessel (mainly SS316) components in current design concepts. Two different waste categorization approaches—one based on global activity limits, the other on nuclide-specific criteria—to define low-level waste (LLW) have been applied to produce estimated predictions of the decay-timescale required before DEMO waste can be disposed of as LLW in near-surface repositories. The predictions show that under either classification scheme much of the in-vessel steel in current DEMO designs may exceed LLW limits for more than 1000 years. This is not an attractive prospect for fusion, which aims at leaving zero-amounts of higher-level waste, requiring deep geological disposal repositories, beyond 50–100 years after DEMO (or a commercial fusion plant) has finished operating. Relatively small amounts of minor elemental impurities, such as nitrogen and niobium in Eurofer, are the main culprits of this negative waste outlook because they lead to the production of problematic long-lived radionuclides. Even some functional materials, such as tungsten and beryllium contain manufacturing impurities that can delay (or prevent) convenient disposal.

The findings highlight the need for further research and engineering effort for DEMO so that it will meet the stated low-activation objective, or alternatively this goal should be relaxed. Several possible mitigation approaches are foreseen for the specific issue of minor compositional impurities leading to small concentrations of nuclides with high radiotoxicity:

(i) the planned DEMO operation schedule could be adjusted so that the maximum neutron dose received by materials in a particular component does not cause the growth of problem radionuclides to exceed LLW limits;
(ii) the material composition could be altered to reduce the minor impurity causing the issue and ensure that the long-lived radionuclide it produces remains below acceptable limits;
(iii) the planning (and costing) for DEMO could include the design of a specific repository to handle the radioactive waste it will produce. Many of the long-lived radionuclides are produced in tiny concentrations and relatively minor changes to existing near-surface repository designs may allow these to be accepted at higher levels;
(iv) the problematic long-lived radionuclides could be extracted via processing before waste material (steel) is released for long-term storage. For example, recent and ongoing work within the European DEMO design programme suggests that it could be possible to decarburize both SS316 and Eurofer using oxygen to levels below one part per million (ppm) [30]. Under such circumstances the equivalent reduction in ^{14}C (a small fraction of carbon but it would be reduced by the same relative amount) would decrease its corresponding Becquerel activity far below the UK-LLW limit. A similar method, if proven, for niobium reduction in steels could also mitigate the issues surrounding ^{94}Nb.

The work presented further exemplifies how the rigorous methodology used here (and previously [6, 7]) to assess the waste prospects for whole reactor designs can be rapidly applied to test new DEMO models, and/or to large databases of potential materials to provide guidance on acceptability of materials for future evolutions of DEMO design.

Acknowledgments

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