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L. W. Packer, P. Batistoni, N. Bekris, S. C. Bradnam,
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Savva, I. E. Stamatelatos, T. Vasilopoulou, A. Wojcik-
Gargula, A. Zoharf, JET Contributors

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A. Zoharf, JET Contributors

WASTE PREDICTIONS AND NEUTRON ACTIVATION OF FUSION MATERIALS AT JET: SIMULATIONS, RADIOMETRIC OBSERVATIONS AND MEASUREMENT CHALLENGES

L.W. PACKER*, A. TURNER, S BRADNAM, H. CHOHAN, Z. GHANI, M.R. GILBERT, C.R. NOBS
United Kingdom Atomic Energy Authority, Culham Centre for Fusion Energy, United Kingdom
*Email: lee.packer@ukaea.uk

P. BATISTONI, M. PILLON, S. LORETI
ENEA - Department of Fusion and Technology for Nuclear Safety and Security via E. Fermi 45, 00044 Frascati (Rome), Italy

J. W. MIETELSKI, R. KIEREPKO, A. WOJCIK-GARGULA
Institute of Nuclear Physics, Polish Academy of Sciences, PL-31-342 Krakow, Poland

E. LASZYNSKA
Institute of Plasma Physics and Laser Microfusion, 01-497 Warsaw, Poland

M. FABBRI
Fusion for Energy, Josep Pla 2, Torres Diagonal Litoral B3, 08019 Barcelona, Spain

I. LENGAR, A. ZOHAR
Reactor Physics Department, Jozef Stefan Institute, Jamova cesta 39, SI-1000 Ljubljana, Slovenia

I.E. STAMATELATOS, M. I. SAVVA, T. VASILOPOULOU
Institute of Nuclear and Radiological Sciences, Technology, Energy and Safety, NCSR Demokritos, Athens, 15310, Greece

N. BEKRIS, D. LEICHTLE
Karlsruhe Institute of Technology (KIT), Hermann-von-Helmholtz Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

and JET Contributors. See the author list of "E. Joffrin *et al* 2019 *Nucl. Fusion* **59** 112".

Abstract

Experimental activities conducted at JET as part of a EUROfusion project have exposed ITER construction material samples to fusion neutrons. To date this has been during deuterium plasma operations, and presently, exposing further samples in the ongoing DT experimental campaign. Activation measurements and calculational predictions for the JET fusion environment acting on a range of supply chain ITER materials that were exposed to neutrons during deuterium fusion plasma operations. The ITER materials include: poloidal field (PF) coil jacket and toroidal field coil radial closure plate steels, EUROFER 97-2 steel, W and CuCrZr materials from the divertor, Inconel 718, CuCrZr, 316L stainless steel for blanket modules and vacuum vessel forging samples. The measurement of nuclides present in these samples using high resolution gamma spectrometry techniques is discussed. To date these are principally measurements for activation products of importance to remote handling in operations and early decommissioning phases.

Initial findings have revealed some variations in observed, short-lived nuclides relevant to maintenance and decommissioning operations, that are present in steels from different suppliers. Measurement techniques and strategies for long-lived nuclides, such as ^{94}Nb , that are relevant to long-term waste disposal timescales are discussed along with the outlook and radiometric challenges following the ongoing JET DT experimental campaign. The amounts of fusion generated waste arising at JET itself may be predicted using 3D neutronics and activation codes with underpinning modern nuclear data libraries. Details of the applied nuclear analysis methodology acting upon a detailed neutronics model of the JET machine are presented. Results show that effective detritiation of materials and modest decay times can be effective as part of an overall strategy to ensure that ILW volumes can be minimised.

1. INTRODUCTION

Nuclear fusion has the potential to provide a long-term, low carbon-generating, sustainable energy supply with the prospect of contributing to global energy generation in the second half of this century. The most promising fusion fuelling mechanism involves burning deuterium and tritium ions (the DT fusion reaction) in a super-hot, confined plasma in which energetic products, 14.1 MeV neutrons and 3.5 MeV alpha particles, are created. In magnetic confinement concepts such as tokamaks and stellarators the plasma fuel ions and electrons are confined for long periods of time (several seconds typically) via magnetic field lines, a so-called 'magnetic bottle'. The high energy neutrons borne in this process, being uncharged, readily escape the plasma and then interact with the structural

and functional materials surrounding it. The key utilisation of these neutrons in a power plant is to produce the otherwise scarce tritium fuel in a breeding blanket containing lithium compounds and neutron multiplying media such as Be. However, structural steels and various functional materials are necessarily present in the device and will become activated by the neutrons, particularly the first wall armour and divertor structures. In power plant operational conditions, the activation products that are generated within these largely solid metallic bulk materials are predicted to have significantly high levels of activity at short post-operational cooling times. Importantly, fusion wastes are significantly different from those generated in nuclear fission - they will not contain the high radiotoxicity, long lived actinides and transuranics that are associated with spent fission fuel. With the careful selection of fusion construction materials and minimisation of certain elemental impurities in supply chains and fabrication processes (such as Nb and Mo that lead to long-lived nuclides under neutron irradiation) the generated radionuclides are mostly short-lived (<100y). With recent advances in detritiation processing and other waste stream processing, the recycling of a large fraction of these materials (minimising the volume of waste stored long-term in repositories) seems to be a realistic proposition under a risk-based fusion regulatory framework that is distinct from the fission approach.

On the roadmap to fusion power, and critical to its success, is the scientific and technological learnings through tokamak-based experimental devices. A current focus of experimental effort is at JET [1], currently the largest and most powerful tokamak in the world - the only machine capable of operating with the deuterium-tritium fuel mix of future commercial reactors. Increasingly, in coming years, the shift of the international fusion scientific community will move towards ITER [2], currently being constructed in Cadarache in Aix-en-Provence in the South of France through international members. It is being designed to operate with a nominal 500 MW fusion power and a fusion gain of $Q = 10$. Once operational, ITER will test fusion technologies, materials, and physics regimes that guide the commercial pathway to fusion-generated electricity.

This paper provides some details on experimental activities at JET that exploit the JET neutron environment in support of ITER. Historical and future projections for JET operations are discussed and details relating to prediction of waste arisings according to the current England, Wales and Northern Ireland waste classification scheme using state-of-the-art computational neutronics and activation methodologies. Accurate predictions of the induced radioactivity associated with nuclear fusion is an essential input to the development of the required technologies and design; besides the prediction of nuclear waste arisings against classification schemes, the inherent residual nuclear fields themselves generated due to neutron activation of components present operational considerations that impact on the 'how and when' decisions around waste management and decommissioning. The experimental activities discussed in this paper related to the measured activity levels of post-irradiated ITER materials samples, sourced from a range of suppliers that have been selected for the construction of components within the device. Key measurement results obtained so far and the measurement techniques used are provided.

2. NEUTRON TRANSPORT AND ACTIVATION TOOLS: WASTE ARISING AND RADIATION ENVIRONMENT PREDICTIONS

Nuclear simulation codes such as MCNP6 [3] used in connection with nuclear data libraries such as FENDL-3.1 [4], together with inventory codes undertake an essential function in predicting the primary nuclear fields (neutron and photon) in which technologies operate. FIG 1 shows the spatial prediction of neutron flux distribution through an X-Z cross-section of an ITER model performed at 500 MW fusion power in DT operation using MCNP6.

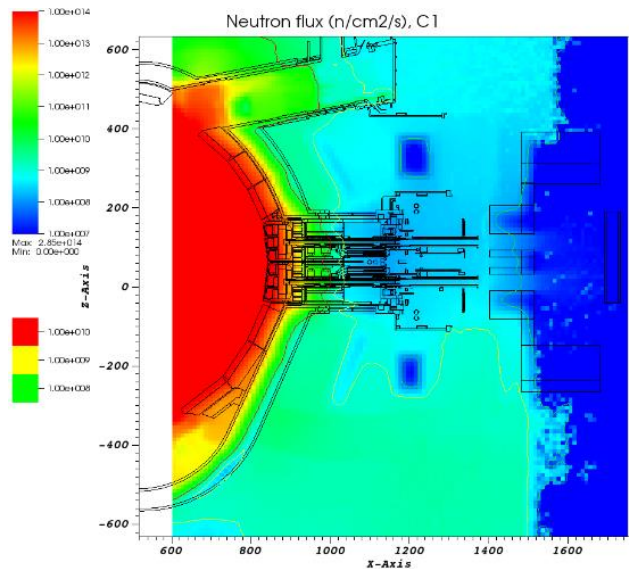


FIG 1: Simulated neutron flux map through an X-Z slice of an ITER model containing an ion cyclotron resonance heating system in an equatorial port. Calculations were performed using MCNP6 and show a neutron flux range spanning seven orders of magnitude. [image reproduced from IAEA IAEA-TECDOC-1935]

With such information, residual gamma fields, created due to neutron induced activation of the materials within the device itself, may be predicted in a temporal fashion using computational codes such as FISPACT-II [5],[6] with nuclear data libraries such as TENDL-2017 [8] or EAF-2010 [7]. Linking inventory codes together with radiation transport codes allow for time-dependent spatial distribution of the activation gamma fields to be determined. MCR2S [9], [10] is an example of such a code and has been applied in the illustrative JET waste analysis described in section 3.

3. JET WASTE ANALYSIS

Prior to the cessation of JET operations, expected in 2023, highly anticipated experimental campaigns will be performed using a mixture of deuterium and tritium (DT) to provide important data for ITER. The use of DT presents additional challenges for decommissioning due to the activation of materials by fusion neutrons.

As part of plans for the dismantling, repurposing and disposal of JET, it has been necessary to improve understanding of the activation of components within JET in detail and the expected shutdown dose rates when these components are removed by remote handling. Previous JET neutronics models that have been developed were relatively coarse in geometric detail and did not permit detailed shutdown dose rate analysis at the individual component level within the vacuum vessel. As such, efforts to develop a more detailed neutronics model of Octant 1 of JET was created containing detailed representations of the in-vessel components. Figure 2 illustrates the old and new JET model. The new model was created using SpaceClaim to perform geometry simplification (1) and converted using SuperMC [22] into a corresponding MCNP radiation transport model which has been used for recent nuclear analyses.

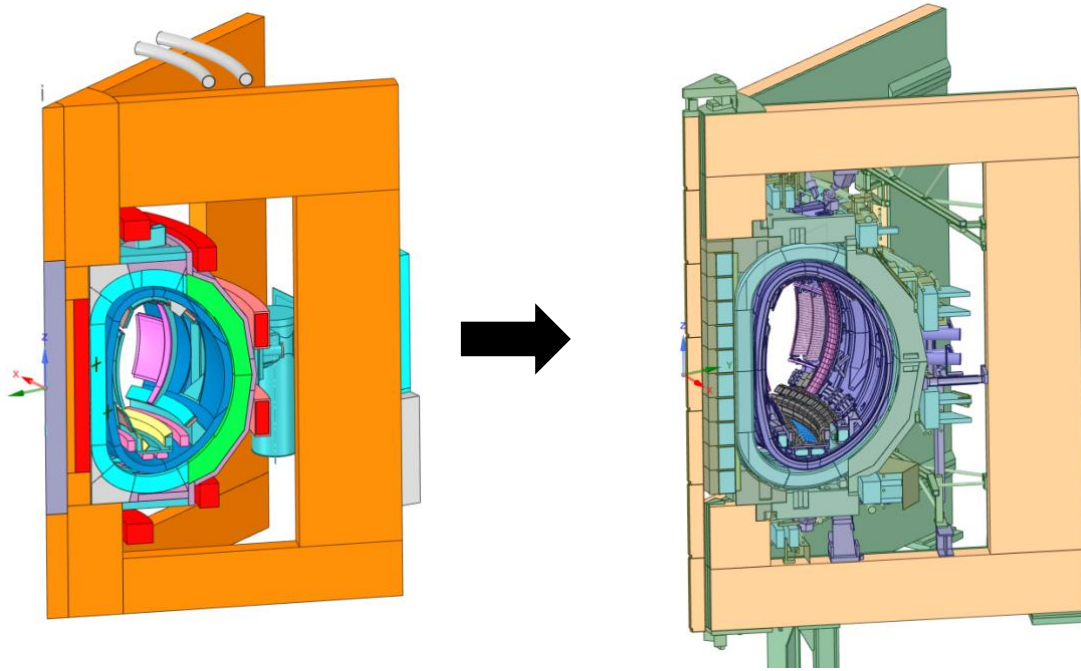


FIG 2: (LHS) previous JET model; (RHS) updated, detailed CAD model of JET which formed the basis of the neutronics analysis in this work

Material definitions were represented with impurity elements since these make a significant contribution to the levels of material activation. On-load neutron flux calculations were carried out using MCNP6, and the subsequent radionuclide inventory and decay gamma sources were determined using MCR2S which uses FISPACT-II for neutron activation. Cell-under-voxel techniques were employed in MCR2S to provide both high spatial accuracy as well as component-level segregation of the gamma source and radionuclide inventory.

A key input to the FISPACT-II activation code is the time variation in the fusion neutron yield. Data was used from actual, historical neutron yield data, with estimates being used for future campaigns, shown in Figure 3. It comprises the DTE2 campaign that began in late 2021 as well as an assumed DTE3 DT and D-D operations during the final 8 months of operation, finishing in December 2023. Shutdown dose rates were calculated for decay times of 1 year, 2 years and 5 years after the end of operations in December 2023, as well as immediately at shutdown (see reference cooling times in figure 4).

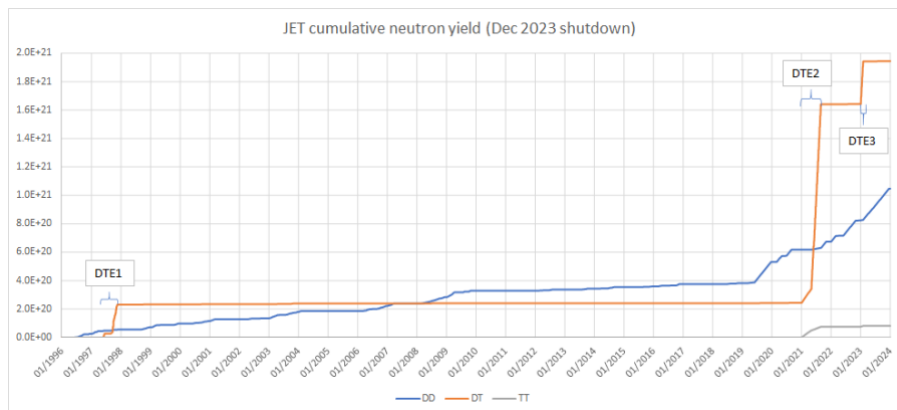


FIG 3: DD, TT and DT neutron yield history used for JET. The DT and TT neutron yield in 2021 onwards are based on assumptions relating to the maximum neutron budget.

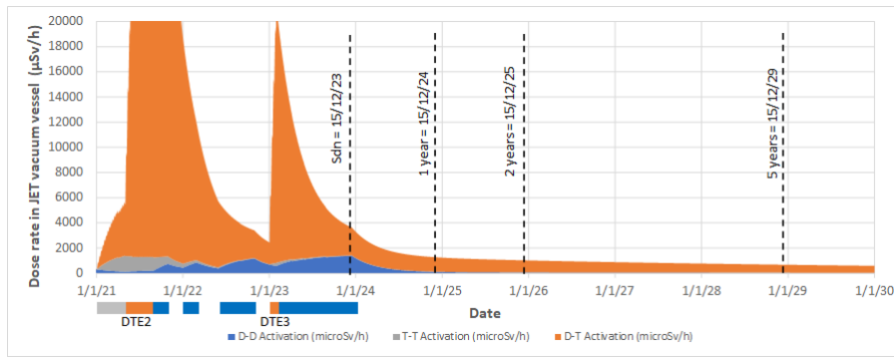


FIG 4: Reference decay cooling times used in this analysis (dashed lines) and approximate estimated in-vessel decay gamma dose rates.

The shutdown dose rate was obtained at various distances from isolated in-vessel components, to simulate the radiation fields expected when these components are removed from JET. Examples are shown in Figure 5 for both a divertor tile and an Inconel wall tile, for a cooling time of 1 year. Dose rate data is being used to inform the strategy for safe handling and processing of the various components and estimate the operator dose uptake.

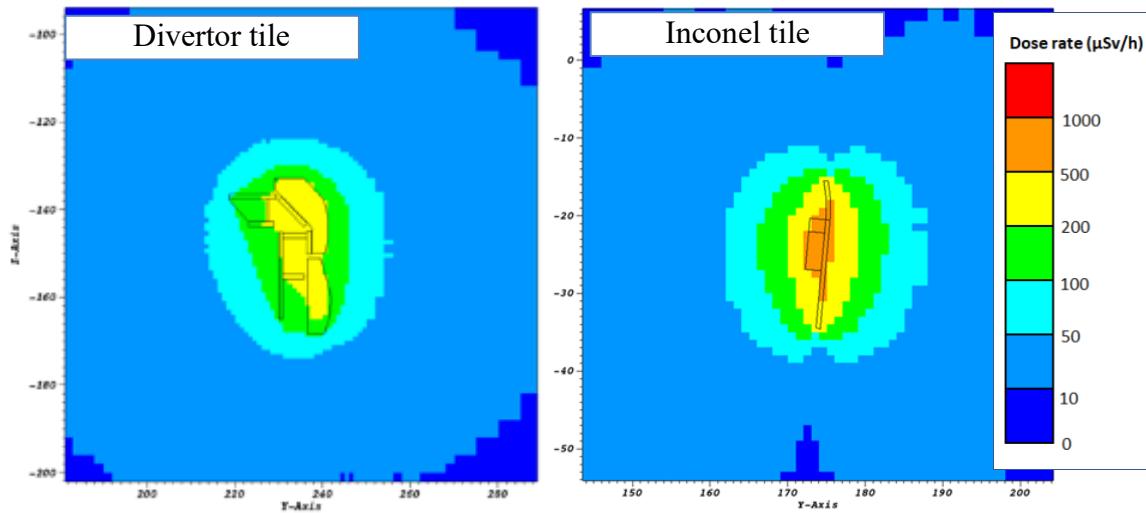


FIG 5: Shut-down gamma dose rate calculations calculated using MCR2S at a cooling time of 1 year: (LHS) a tungsten divertor tile, and (RHS) an Inconel tile.

A new waste categorisation feature of the MCR2S code was used to obtain the total mass of different waste types and its spatial distribution. The radioactive waste categories that were implemented in this assessment are shown in Table 1.

TABLE 1. RADIOACTIVE WASTE CLASSIFICATION SCHEME IN ENGLAND, WALES AND NORTHERN IRELAND

	HLW	ILW	LLW	LA-LLW	OSR
	High level waste	Intermediate level waste	Low level waste	Low activity low level waste ¹	Out of scope of regulations
Specific activity	> 12 GBq/t or > 4 GBq/t [α]		< 12 GBq/t and < 4 GBq[α]/t	< 200 Bq/g	See summation rule criteria for nuclides in [24]
Heat:	> 2 kW/m ³	< 2 kW/m ³			

¹As defined by LLWR, for disposal to suitable authorized landfill sites.

An example of the spatial waste categorisation of JET immediately at shutdown and at 5 years cooling time is shown in Figure 6 (assuming 100% detritiation of materials). Information on the distribution of ILW versus LLW is particularly useful to plan for the cutting and timing of larger components as part of a waste minimization strategy.

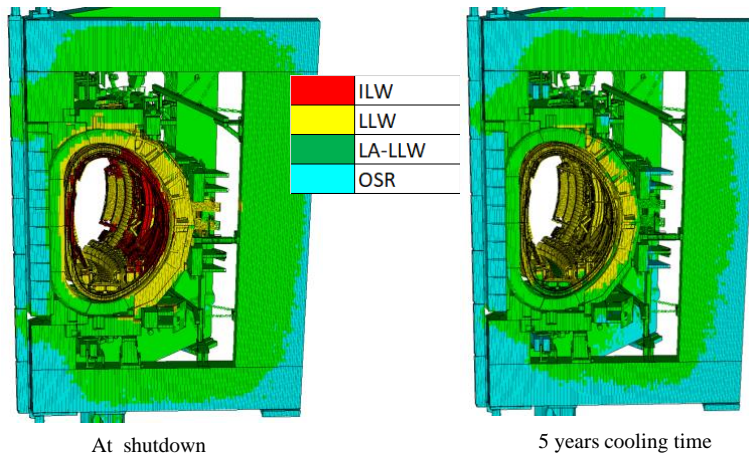


FIG 6: Spatial map of JET waste categorization at shutdown and 5 years cooling assuming 100% detritiation of materials.

The total waste masses of each type are shown in Figure 7 (the mass in each waste category due to DT neutron activation only and does not include impact from use of tritium other than routes produced through activation). These results clearly show that the quantity of ILW in JET falls markedly in the first two years after cessations of operations, and becomes negligible after 3 years cooling (left-hand side plot). Tritium fuel retention is not accounted for in the left-hand side plot. Work has been performed based on results obtained in [23] to supplement the neutron activation analysis with data on tritium retention from experimental measurements within the in-vessel components. It is clear that if significant quantities of tritium were to remain within the component, a significant volume of in-vessel materials would remain ILW for long time periods. Whilst significant work is still ongoing to refine analysis based on tritium loadings and on the associated detritiation techniques that can be applied, here our preliminary analysis in these two scenarios demonstrate that modest deferment/decay storage combined with efficient detritiation of the in-vessel components is highly beneficial as part of a strategy to minimize, or even eliminate, the ILW on disposal.

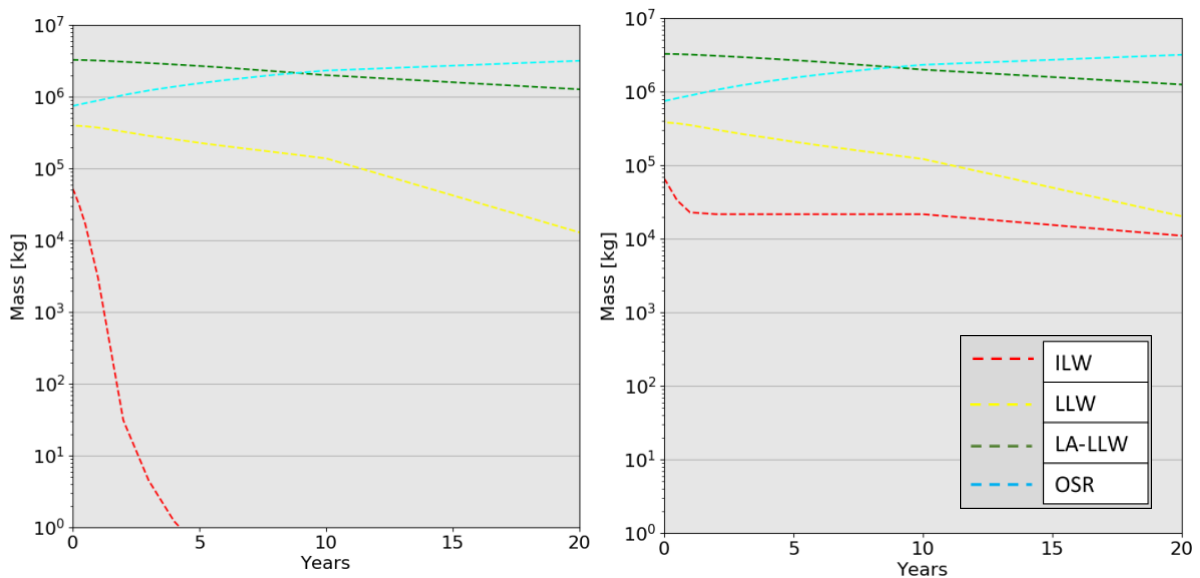


FIG 7: Prediction of JET global waste masses and categorisation at different cooling times; (LHS) assuming 100% detritiation of materials; (RHS) an example conservative scenario without any detritiation.

4. ITER MATERIALS ACTIVATION AT JET: HIGHLIGHTED RESULTS

Gamma spectrometry measurements and underpinning nuclear analysis is provided, obtained following part of the JET 2019 C38 deuterium campaign. In this campaign ITER material samples were irradiated in a 147 day period

in a long-term irradiation station (LTIS) assembly in a location outside of the JET vacuum boundary, but close to the vessel wall. The samples were retrieved post-irradiation and analysed at four EU laboratories to identify and quantify the nuclides present. [14], [15] provide detailed description of the preparatory work and full analysis. Here, new example results of the calculation methodology for an ITER XM-19 steel sample and the corresponding measurement are provided. Figure 8 shows the activation analysis per nuclide for this particular sample, both during and post-irradiation. A key input to the analysis, beside the irradiation history and neutron spectrum, was the material certificate which defined the elemental composition of this steel including impurities.

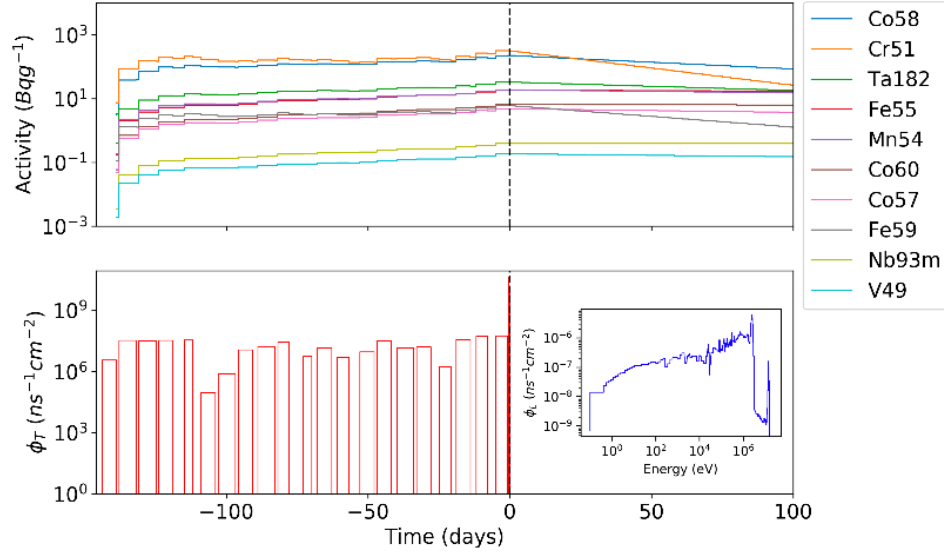


FIG 8: Top plot: specific activity prediction of dominant nuclides during and following JET irradiation of a ITER XM-19 steel sample (Aubert & Duval, Forgings for the ITER divertor cassette) sample (position reference 13-3). The dashed vertical line denotes the time at which the samples were removed from the JET LTIS. Bottom plot: weekly-average neutron fluence over the sample volume within the LTIS. The inset plot shows the neutron fluence per unit lethargy energy spectrum averaged over the sample volume within the LTIS, calculated using MCNP.

Figure 9 shows the gamma spectrum measured at UKAEA after the sample were retrieved. The presence of ^{182}Ta , ^{60}Co , ^{58}Co , ^{51}Cr , ^{57}Co , ^{54}Mn , ^{59}Fe , ^{65}Zn and ^{124}Sb are evident from the characteristic lines that are present. Figure 11 shows the calculated (C) over experimental (E) ratio for this sample. With the exception of ^{60}Co and ^{182}Ta (for the latter nuclide see Figure 11b) there is good agreement with values generally around 1.

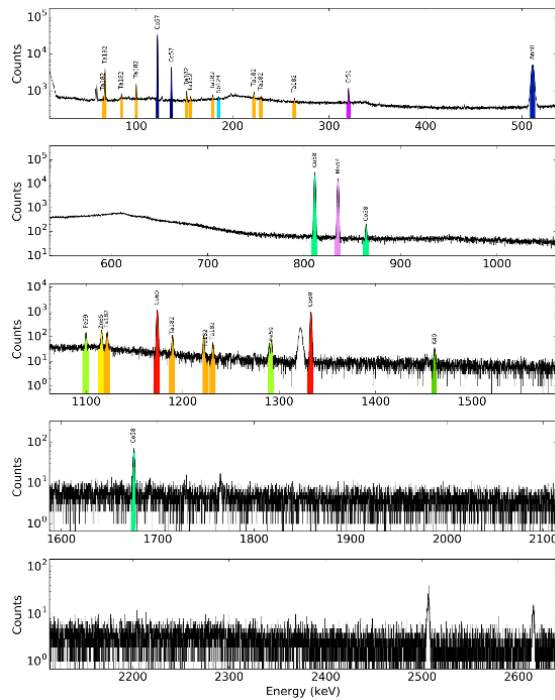


FIG 9: High energy resolution gamma spectrum measurement of an ITER XM-19 steel sample (Aubert & Duval, Forgings for the ITER divertor cassette) taken post-irradiation at JET using a UKAEA BEGe detection system.

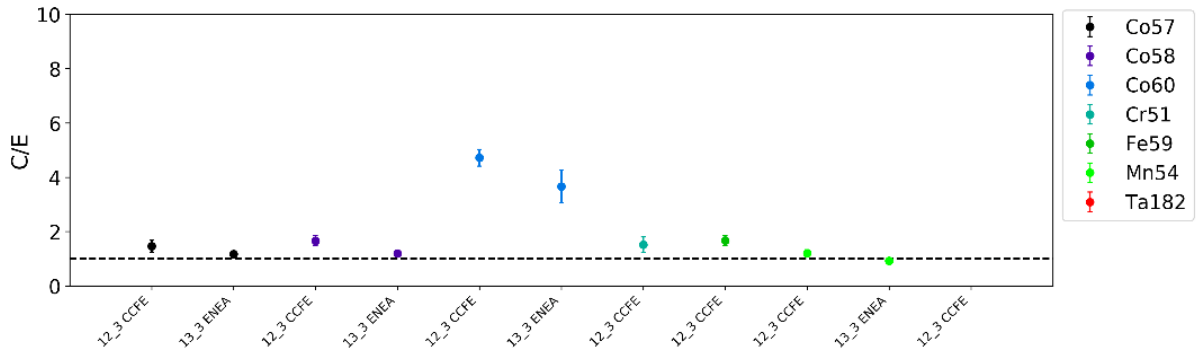


FIG 10: C/E values for measured and calculated XM-19 steel sample activities. Results are grouped by the measured isotope (note that isotope measurement results may be from different reaction pathways). ^{182}Ta is not shown on this scale but can be seen in the XM19 results in Figure 11b.

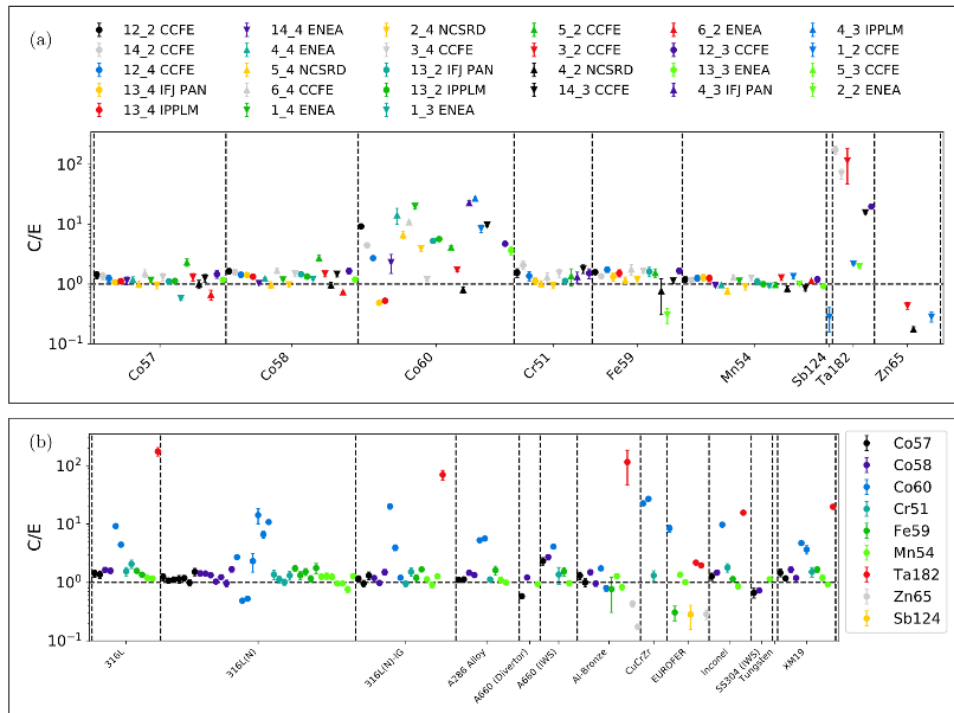


FIG 11: [figure reproduced from [14]] C/E values for all measured and calculated ITER materials from the JET C38 deuterium experimental campaign: (a) results grouped by measured isotope (isotope measurement results may be from different reaction pathways). The legend indicates the position–depth ID linked to the material description in Table 1 of [14] and the measurement laboratory; (b) results grouped by material type.

More generally, from the analysis discussed in [14] with C/E analysis for the full set of ITER samples that were irradiated, Figure 11 highlights discrepancies in C/E values for ^{60}Co , ^{65}Zn , ^{124}Sb and ^{182}Ta which require further studies to establish the exact reasons for high or low C/E values.

5. CONCLUSIONS AND CHALLENGES FOR MEASUREMENTS OF LONG-LIVED FUSION WASTE PRODUCTS

The accurate prediction and measurement of nuclides present in fusion generated wastes is essential to validate and develop the strategies for recycling, decommissioning and disposal. The potential for fusion as a low carbon generating, sustainable energy source is significant; it is important to ensure that these benefits are made as clear as possible, both to the public and to regulators, as well as the decommissioning approach and predicted waste arisings. Whilst there will be no high level waste (HLW) of the type that is associated with nuclear fission, some lower classification wastes (ILW, LLW and LA-LLW) will be generated unavoidably due to neutron activation and use of tritium as fuel. These strategies will need to work closely with the materials supply chain to minimise undesirable impurities, maximising the recycling of materials in future plants and, as our JET scenario nuclear analysis illustrates, to further advance detritiation technology in order to minimise the potential wastes.

Whilst simulated predictions are essential for estimates of waste arisings, it is important to learn and validate these as far as possible from experimental studies of the nuclear response of real construction materials that will be exposed in near-term tokamak based fusion experimental devices such as JET and ITER. In this work, more extensively covered in [14], the radiometric measurements of ITER materials that have been irradiated in the JET neutron environment have focused on the measurement of short- and medium-half-life nuclides. Next steps, for future irradiations of ITER materials in the ongoing JET DT experimental campaign and their retrieval post-irradiation, are to complement the existing measurement techniques with new approaches. The aim is to measure some of the more challenging long-lived nuclides that are expected to be present on long-timescales and thus are relevant to waste disposal. Some of these nuclides have been highlighted in previous work in [17] and [18] for example. One of the problematic long-lived nuclides is ^{94}Nb which has a half-life of 20,300 years. It can be produced through neutron capture in Nb impurities or through neutron energy threshold reactions with some Mo isotopes. One of the challenges in measuring such long-lived nuclides in activated materials using radiometric techniques will be to identify and quantify the nuclide in a high gamma background, present due to shorter-lived products with energetic gamma emissions, such as ^{60}Co for example. The decay scheme of ^{94}Nb is shown in Figure 12, which shows that the nuclide is a strongly cascade summing. This coincidence gamma signature can be exploited for quantification through use of a gamma-gamma coincidence measurement system (see right-hand side image of Figure 13 from R Britton, SnT proc. 2021), such as that detailed in the extensive work and new systems used within the comprehensive test-ban treaty organization network of laboratories [19], and also using a five-detector system in Japanese environmental radionuclide measurements [20]. The latter work indicates improvements in signal-to-background improvement factors of 205 for ^{94}Nb which is encouraging.

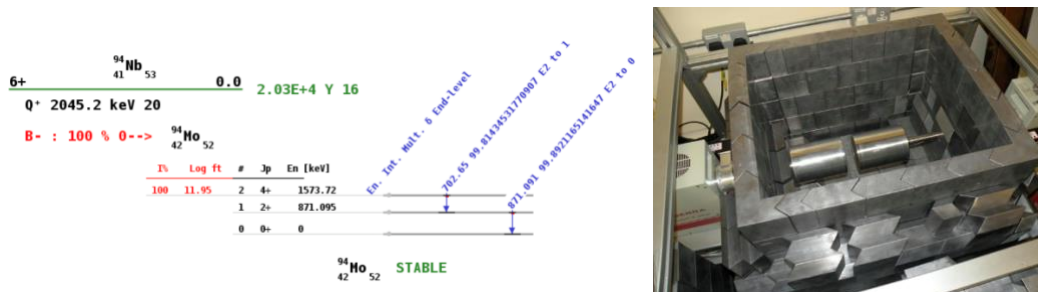


FIG 12: (LHS) Decay scheme for Nb-94 showing the 702.65 and 871.091 keV gamma cascade (IAEA livechart); (RHS) Example of a gamma-gamma coincidence system [Image - R. Britton et al., A Gamma-Gamma Coincidence System for Radionuclide Quantification, SnT2015 proc.] See [19].

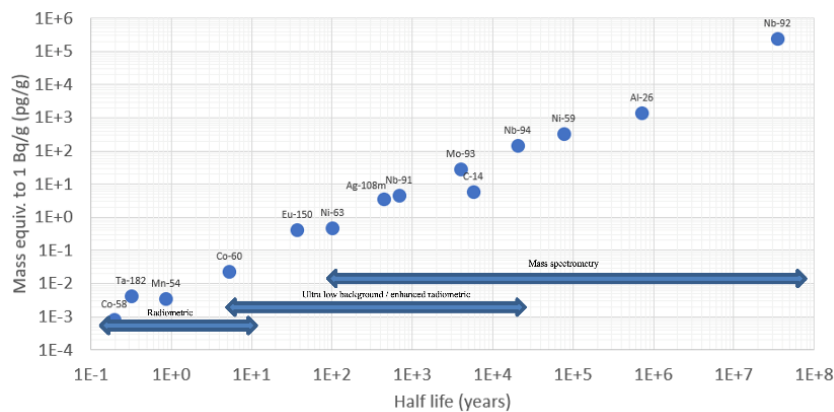


FIG 13: Half-life versus mass equivalent to 1 Bq/g (pg g^{-1}) for selected activation products.

The specific activity for a given radionuclide is inversely proportional to the half-life; for long lived radionuclides more sensitive measurements can be achieved by determining the atomic mass concentration rather than by measuring the activity of the nuclide [21]. Techniques such as inductively coupled plasma mass spectroscopy (ICP-MS) or accelerator-based mass spectroscopy (AMS) are potential alternatives to decay counting techniques. Whilst a drawback is that the techniques are destructive methods involving radiochemistry processing, they complement radiometric techniques and provide superior detection limits for some long-lived radionuclides that are present in ITER samples, such as Ni-59, Ni-63, Nb-94 and C-14. Figure 13 gives a simplistic illustration of the mass equivalent to 1 Bq/g in units of pico-grams per gram for some nuclides of potential interest plotted against their half-life. The plot 'loosely' indicates the temporal space where mass-based techniques may be advantageous in terms of detection limits over radiometric techniques.

ACKNOWLEDGEMENTS

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