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Abstract

A large number of materials exist which have been labeled as low activation structural materials (LAM). Most often, these materials have been designed in order to substitute-out or completely remove elements that become activated and contribute significantly to shut-down activity after being irradiated by neutrons in a reactor environment. To date, one of the fundamental principles from which LAMs have been developed is that natural elemental compositions are the building blocks of LAMs. Thus, elements such as Co, Al, Ni, Mo, Nb, N and Cu that produce long-lived decay products are significantly reduced or removed from the LAM composition. These elements have an important part to play in the composition of steels and the removal/substitution can have a negative impact on materials properties such as yield stress and fracture toughness. This paper looks in more detail at whether using isotopic selection of the more mechanically desirable, but prohibited due to activation, elements can improve matters. In particular, this paper focuses on the activation of Eurofer.

Carefully chosen isotopically enriched elements, which are normally considered to be on the prohibited element list, are added to EUROFER steel as potential alloying elements. The EUROFER activation results show that some prohibited elements can be used as alloying elements in LAM steels, providing the selected isotopes do not have a significant impact on waste disposal rating or shut-down dose. The economic implications of isotopically enriching elements and the potential implications for decommissioning are considered. It is shown that the addition of molybdenum and nickel in small concentrations (< 2 % mass) could have the potential to increase availability to such an extent that the capital investment associated with isotope enrichment is negated and profits from electricity sale increased.

Another important issue for materials exposed to neutron irradiation is the production of gases, in particular helium, as a result of nuclear interactions. Helium accumulation in materials can cause embrittlement and so mitigating the rate of production is an important consideration when selecting materials. The second part of this paper considers whether helium production can be reduced in CuCrZr by isotopic tailoring. CuCrZr is a candidate bonding material for tungsten at first wall locations, however it suffers from degradation due to helium production. Inventory calculations show that isotopically enriching the copper, in CuCrZr, can reduce helium production by approximately 50%. However, cost-benefit analysis suggests that the cost of enriching copper is not cost-effective due to the high price of electromagnetic enrichment that is required for copper.

Keywords: Activation, LAM, enrichment, shut-down activity, waste disposal, gas production.

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1. Introduction

The structural materials within the ITER experimental fusion reactor vessel will be exposed to unprecedented neutron irradiation. As a result, the incident neutrons will cause macroscopic defects and nuclear transmutations within the structural materials. One of the problems that is encountered when transmutations occur is the production of activated nuclides that decay with long half-lives, which have radiological and waste disposal implications. Next-step fusion devices, such as DEMO, will be subject to significantly higher neutron fluences than will be experienced by ITER, resulting in higher activation. Hence, the need to reduce the amount of activated nuclides produced in these fusion devices will become increasingly important. One principle, which has been adopted in order to reduce the amount of activated products is the substitution of elements, that are particularly susceptible to activation, with chemically similar elements with a lower susceptibility to activation. F82H and EUROFER steels are low activation structural materials (LAM), where niobium has been replaced by tantalum [1, 2], molybdenum has been replaced by tungsten [3], titanium has been replaced by vanadium [4] and manganese has been replaced by chromium[5].

The subject of LAMs is most often focused on the reduction of radioactive waste. This is, of course, very important. However, reducing the amount of long-lived nuclides produced is only one of the advantages of adopting the LAMs. Reducing the amount of short-lived nuclides that affect the shut-down dose during periods of maintenance is also important, as it can increase plant availability due to reduction of the cool-off period before maintenance. Very short-lived activation products could be problematic during an accident. Thus, a truly low activation material will have a low activation profile for short, medium and long-lived nuclides. Unfortunately, achieving this goal in entirety is likely to be impossible. However, a principle that has yet to be fully investigated, which could help move towards this goal, is the use of isotopically enriched elements within LAMs [6] in order to complement particular elemental reductions and substitutions. A commonly used method to produce LAMs is to eliminate the parent nuclides of alloying elements, such as Co, Al, Ni, Mo, Nb, N, B and Cu, which produce long-lived decay products [5, 7, 8]. In some cases, the replacement only works to a limited degree, for instance the replacement of manganese with chromium and the replacement of molybdenum with tungsten [4].

This method of producing LAMs assumes that the alloying elements have a natural isotopic composition. However, removing particular isotopes of the alloying elements may enable some elements to be used in LAMs, which had previously been eliminated from the composition. Isotopes of the same element have almost identical chemical properties but differ significantly in their nuclear properties. This method of careful isotope selec-

tion allows the bulk material to benefit from the elements chemical properties without suffering decremental effects due to an individual isotopes nuclear properties.

Other important issues for materials exposed to neutron irradiation are atomic displacements, resulting in lattice defects, and the production of gases, in particular helium, as a result of nuclear interactions. Helium accumulation in materials can cause swelling and embrittlement thus mitigating the rate of production is an important consideration when selecting materials. Gas production is especially problematic in the fusion environment due to the 14 MeV fusion neutrons being above many of the gas producing, energy threshold reactions which are not of concern in the fission environment, where neutron energies generally less than 2 MeV.

Isotopically enriching elements is likely to significantly increase the cost of the raw materials required for LAMs. Thus, isotopically enrichment is more likely to be adopted if there is a financial incentive for doing so. Financial incentives which may be able to offset the initial capital investment are:

- reducing cool-down period required for maintenance, thus increasing plant availability.
- reducing shielding required within the fusion vessel and within waste processing plants.
- reducing the amount of and classification of radioactive waste that would need to be sent for land burial.
- increased availability gained by adopting materials with increased longevity.
- reducing the cost of replacing components.
- increased thermodynamic efficiency.

Safety-based incentives include [9]:

- reduced radioactive impact to the environment during normal and accident scenarios.
- reduction of decay heat during loss of coolant accident.
- reduced gamma-ray dose during maintenance.

Other incentives such as legacy, responsibility and public perception should also be considered when deciding the extent of isotopic enrichment in LAMs. Only the benefit of increased availability has been included in the cost-benefit analysis in this paper.

Isotopic tailoring is quite prevalent in the literature and has a variety of applications that include ${}^6\text{Li}$ enrichment in tritium breeding blankets [10, 11, 12], minimizing activation [13, 6, 14, 15] and simulating increased gas production for damage studies [16, 17, 18, 19]. However, cost-benefit analysis of isotope enrichment has not been fully addressed in the literature.

This paper focuses on EUROFER and CuCrZr in order to ascertain if their performance can be improved and if any improvement is cost-effective:

1. The examination of the post-irradiation activity of EUROFER steel that has been modified to include isotopically enriched prohibited alloying elements molybdenum and nickel. Cost-benefit analysis is performed, considering cost of enrichment, cost of waste disposal, possible increased availability and possible increased thermodynamic efficiency.
2. The study of gas production in CuCrZr. Cost-benefit analysis is performed considering increased availability against cost of enrichment.

2. EUROFER

This section focuses on the selection of isotopes to be used in isotopically enriched low action materials (IELAM), the reduction of activity achieved by IELAMS and the economic case for IELAMS in relation to nuclear fusion devices.

The first step in ascertaining the financial viability of isotopic enrichment involves defining the susceptibility of constituent steel isotopes to activation. Figures 1 and 2 show nuclide maps [20] illustrate the magnitude of the dose (after 12 days shutdown) and activity (after 100 years shutdown) for nuclides produced as a result of 5 full power years (FPY), 1.25GW DT fusion power. The dose, at 12 days, is dominated by ^{54}Mn ($\sim 92\%$ from $^{54}\text{Fe}(n,p)$) and ^{182}Ta ($\sim 100\%$ from $^{181}\text{Ta}(n,\gamma)$). After 100 years shut-down, the activity is dominated by ^{63}Ni ($\sim 100\%$ from $^{64}\text{Ni}(n,2n)$), closely followed by ^{53}Mn ($\sim 92\%$ from $^{54}\text{Fe}(n,p)$), ^3H ($\sim 99\%$ from $^{10}\text{B}(n,t)$) and ^{14}C ($\sim 100\%$ from $^{13}\text{C}(n,\gamma)$). These EUROFER results will help steer the isotopic design of an IELAM.

2.1. Enrichment

A commonly used method to calculate the costs associated with centrifuge enrichment involves the calculation of the energy required to reduce the entropy of a system of mixed isotopes to a system of separated isotopes. The separative work (SWU) required to reach the prescribed level of enrichment has been calculated using [21]:

$$SWU = W\phi(x_w) + P\phi(x_p) - F\phi(x_f)$$

where P, W and F are the masses of product, waste and feed, respectively. Mass fractions of the waste, product and feed are specified as x_w , x_p and x_f .

The separation potential, $\phi(x_i)$, is defined as:

$$\phi(x_i) = (2x_i - 1) \ln \left(\frac{x_i}{1 - x_i} \right)$$

The mass of the product required can be used to specify the waste and feed mass using:

$$\frac{F}{P} = \frac{x_p - x_w}{x_f - x_w}$$

$$\frac{W}{P} = \frac{x_f - x_p}{x_w - x_f}$$

The waste enrichment has been calculated for each element based on minimizing the total cost associated with procuring the raw elements and isotopically enriching the element. An overall enrichment cost has been calculated by multiplying the SWU by C_{SWU} , the cost per separative work unit, which has taken to be £160/SWU, which is in-line with the cost associated with gaseous diffusion enrichment [22]. Some isotopes, such as copper, cannot be enriched via the centrifuge method and require other methods, such as electromagnetic separation (EM). The EM method is several orders of magnitude more expensive than centrifuge and diffusion methods. Although, this cost may reduce significantly by the expected date for the first DEMO devices to be built (2050's), due to advancements of enrichment technologies such as laser enrichment [23].

2.2. Electricity cost

One of the findings of the Power Plant Conceptual Study [24] is the empirical formula used to define the dependence of cost of electricity (COE) on availability (A), thermodynamic efficiency (η_{th}), net electric power (P_e), normalized plasma pressure (β_N) and Greenwald plasma density (N).

$$COE \propto \left(\frac{1}{A} \right)^{0.6} \frac{1}{\eta_{th}^{0.5} P_e^{0.4} \beta_N^{0.4} N^{0.3}} \quad (1)$$

The most influential variable in the COE equation (equation 1) is the availability. Hence, increasing the longevity of structural components could increase the availability and potentially help to reduce the cost of electricity.

One of the potential drawbacks of isotopic enrichment is the additional capital expenditure required. However, one of the findings of the Power Plant Conceptual Study was that structure contributes approximately 20% to the blanket cost and the blanket accounts for approximately 5% of capital. However, the blanket is a replaceable component so it accounts for approximately 15% of cost of electricity overall. Thus, the structural components account for just 3% of the cost of electricity. Equation 1

Time: 12.00 days (cooling)
 Total Dose Rate (Sv/hr): 1.359E+04

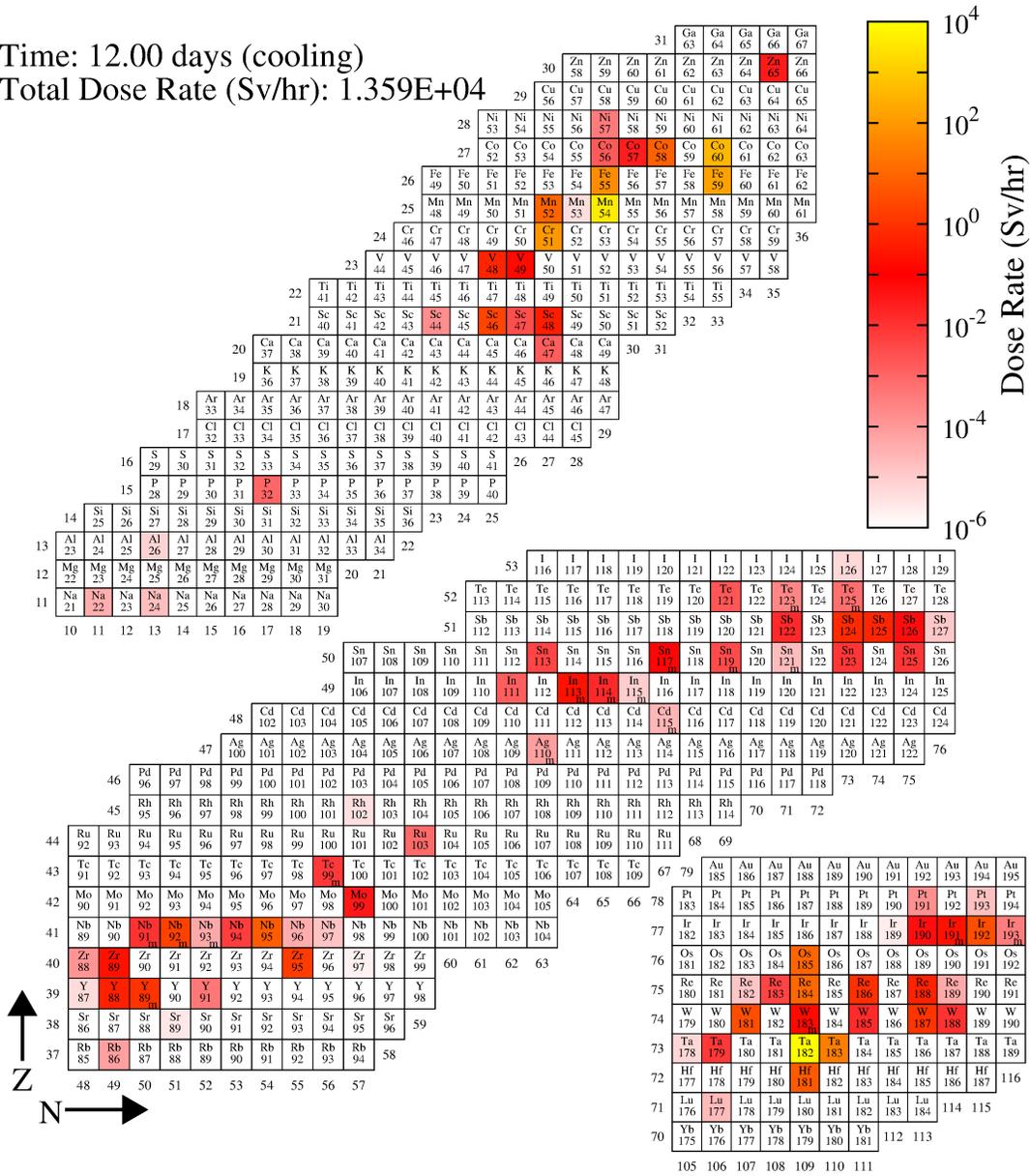


Figure 1: Nuclide dose map for EUROFER, 12 days after shutdown.

can be used to show that an increase in availability of just 5% would be required in order to counter balance the doubling of the cost of structural components.

In addition to the potential reduction of the COE as a result of isotopic enrichment increasing availability, enrichment may also increase η_{th} . The operating temperature of the blanket dictates the thermodynamic efficiency [25], thus the reintroduction of previously prohibited elements, such as nickel, could improve a LAM susceptibility to temperature induced degradation.

2.3. Method

The FISPACT-II inventory code [26] has been used to calculate the evolution of the EUROFER [27](table 1) nuclide inventory as a function of time. In addition to this, the code is used to calculate the activity and reaction/decay pathways between the parent and daughter nuclides. The reaction rates used within FISPACT-II have been calculated using the multi-group method (CCFE-709 group and TENDL-2012 library) in conjunction with neutron spectra calculated within several regions of a HCPB-DEMO model (figure 3) created by the HERCULES code [28]. Materials in this model are broadly homogenized, with only the major structural components being heterogeneously specified. The results presented are only applicable to the HCPB-DEMO model and no claims are made regarding the suitability of these results to other DEMO concepts, such as HCLL.

Neutron transport was modeled using the MCNP6 code [29] and an in-house plasma source routine. A DEMO relevant irradiation scheme, followed by 200 year cooling period has been used to assess the activity of the materials and waste disposal route.

Cost-benefit analysis has been conducted for two possible benefits from isotopic enrichment of specific constituents of EUROFER; reduction of shut-down activity and improvement of materials properties.

2.4. Results

2.4.1. Shut-down activity

Figure 4 shows a dose rate importance diagram [20] for EUROFER that illustrates the isotopic contributors, which have a dose that contributes more than 50 %, to shut-down dose rate over a period of 200 years as a function of neutron irradiation energy. Neutron energy spectra for positions A (Blanket) and B (First wall), of the HCPB DEMO model (figure 3), is shown in figure 5 in order to guide the interpretation of the importance diagram.

A reduction of activity/dose could be achieved in the short and long term by removing manganese. However, isotopic enrichment of manganese is impossible due to it being monoisotopic and the total removal would have

detrimental effect on materials properties, such as fracture toughness, due to its austenite stabilization properties. Conventional LAMs substitute nickel with manganese, due to its lower activity/dose profile. Replacing manganese with isotopically enriched nickel could improve materials properties , while having an insignificant effect on shut-down dose and activity.

Tungsten is a common substitute for molybdenum in LAMs and is used in EUROFER (see table 1), due to it having a reduced impact on activity/dose when compared to molybdenum. However, tungsten is not an ideal substitute for molybdenum, as it produces many undesirable daughter products that include isotopes of tungsten, iridium, osmium and rhenium. In addition to the neutronic properties, the material properties of tungsten fall short of molybdenum, due to Laves phase formation[5].

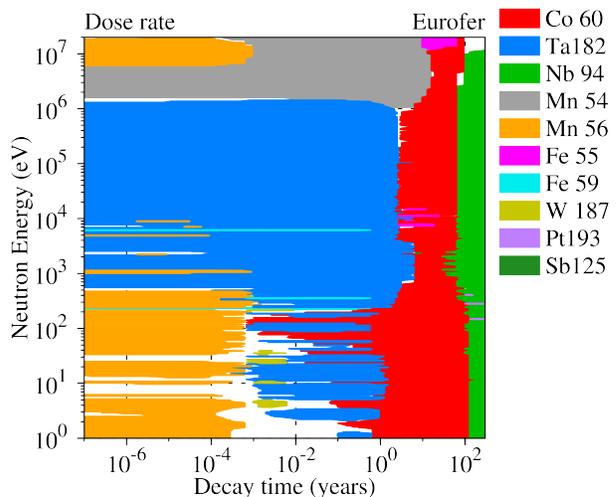


Figure 4: Dose importance diagram for EUROFER.

2.4.2. Additional alloying elements

The addition of nickel has been shown to improve materials properties of steel in a radiation environment [30]. Thus, this section investigates the potential capital costs and revenue implications of adding alloying elements to EUROFER. However, this study is performed purely from a neutronics perspective and the possible materials benefits from the addition of these alloying elements is speculative.

Figure 6 shows the activity of EUROFER and variants of EUROFER after 5 years of irradiation at position A of the DEMO HCPB model shown in figure 3. Adding 5% (mass) natural molybdenum or nickel to the EUROFER increases the activity in the long term. However, isotopically enriched nickel (^{60}Ni , ^{61}Ni & ^{64}Ni)

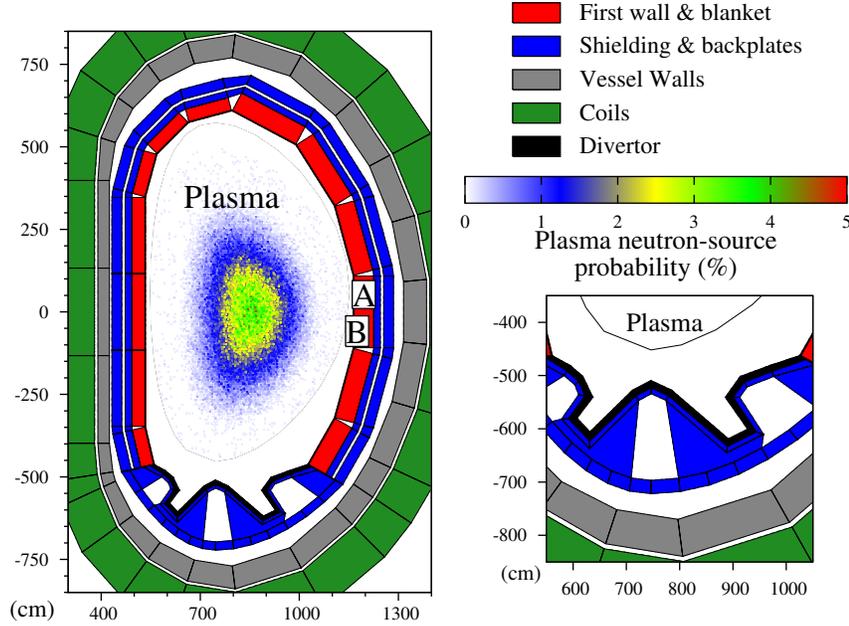


Figure 3: HCPB DEMO model used to irradiate isotopically enriched materials. Blanket and first wall positions used to determine neutron spectra are labeled as A and B, respectively.

Element	Fe	C	Si	Mn	Cr	W	V	Ta	N	B
Mass %	89.47	0.11	0.05	0.5	8.5	1.0	0.25	0.08	0.03	0.005

Table 1: EUROFER composition [5] (Not including impurities).

and molybdenum (^{97}Mo) have a negligible effect on the long-term activity, when compared to EUROFER with no additional alloying elements. Figure 7 shows that ^{64}Ni is the best candidate for enrichment, although other isotopes may be less expensive to enrich.

In order to assess if adding enriched nickel or molybdenum would be cost-effective the following assumptions have been made:

- Nickel and molybdenum compositions in EUROFER contribute 2% mass.
- Approximately 200 tonnes of EUROFER is suitable for alloying ($\sim 10\%$ of blanket EUROFER).
- 1 GW electricity plant.
- £160/SWU for gas centrifuge enrichment (Ni & Mo).
- Each percentage point of increased availability would add 100 GWh extra electricity each year, which is worth £5-10M per year. Assuming a blanket has a lifetime of 5 years, the extra revenue gained from nickel enrichment would be £25-50M per percentage point of availability increase, over 5 years.

- Alloying EUROFER with Mo or Ni could increase availability and/or plant thermodynamic efficiency that could generate extra income that would act as remuneration for the extra initial capital costs of enrichment.

Nickel. Item 1 implies that approximately £360K (45 SWU/Kg) of enriched nickel is required per tonne of EUROFER. Thus, an extra £72M of capital investment would be needed to cover the costs of enriched nickel for a typical DEMO device. Item 6 suggests that if the addition of enriched nickel increases the availability by 1.5 - 3 % the capital costs would be recovered and availability increases of more than 1.5 - 3 % would lead to profit.

Molybdenum. Item 1 implies that approximately £80K (8 SWU/Kg, £1300/Kg) of enriched molybdenum per tonne of EUROFER. Thus, for approximately 200 tonnes of EUROFER, an extra £5.2M of capital investment would be needed to cover the costs of enriched molybdenum for a typical DEMO device. Item 6 suggests that if the addition of enriched molybdenum increases the availability by 1% the capital costs would be recovered and availability increases of more than 0.2 % would lead to profit.

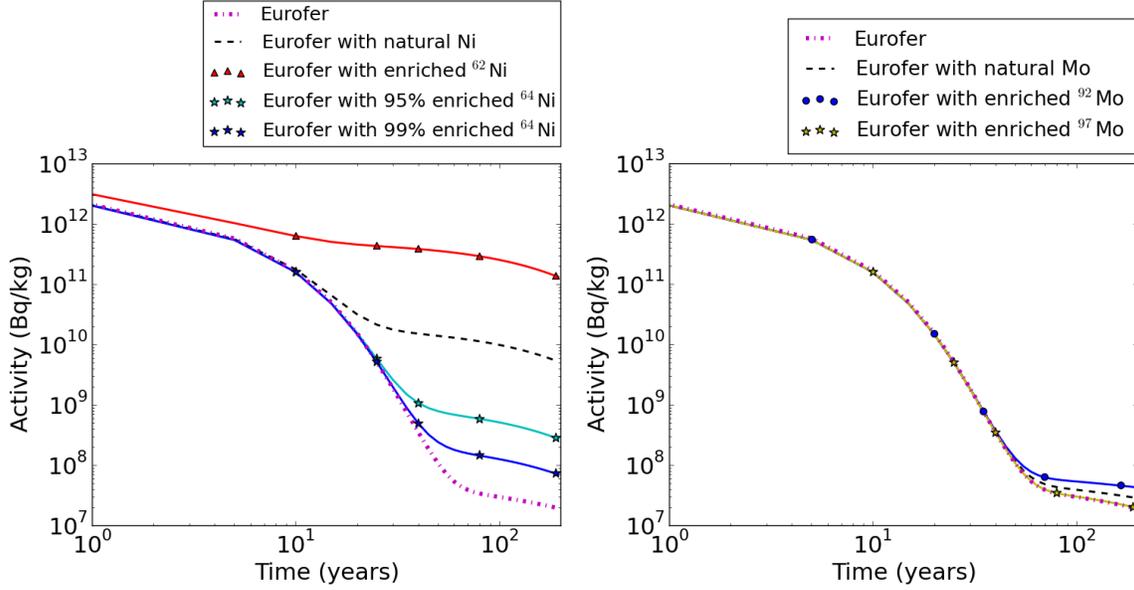


Figure 6: Shutdown activity of EUROFER and variants of EUROFER with natural and isotopically enriched (by mass) nickel and molybdenum.

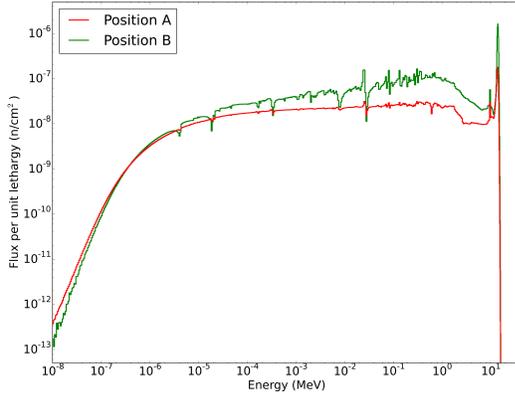


Figure 5: Neutron spectra, in CCFE 709 energy group format, at positions A and B in the HCPB DEMO model shown in figure 3.

A cost-benefit analysis of the effect of using isotopically enriched nickel and molybdenum is part of on-going research. However, an in-depth analysis of the effect of isotopically enriched materials on waste disposal can be found in reference [13].

3. Helium embrittlement in copper

This section outlines how the enrichment of copper in the CuCrZr alloy, a candidate for high-temperature applications, such as first wall structures, can reduce helium embrittlement. A life-limiting factor of CuCrZr is gas

production via $(n, x\alpha)$ reactions, which causes embrittlement due to helium build-up at the grain boundaries. A recent study [32] found that the embrittlement lifetime of molybdenum, with a first wall (FW) location and a crystal grain size of $5 \mu m$, is only 2 years. This might prevent the use of molybdenum at FW location, despite its otherwise good mechanical and thermal properties.

3.1. Method

Helium production in copper has been studied in the blanket and first wall of the DEMO model shown in figure 3. The first step involved calculating the helium production values, as a function of time, with copper having natural isotopic abundance. These single group cross-sections were calculated by FISPACT-II using neutron fluxes, binned in the CCFE 709 energy group structure, at a poloidally equatorial first wall location (position A in figure 3). In order to demonstrate the maximum reduction in gas production, repeat calculations were then performed using isotopically enriched copper.

3.2. Results

^{65}Cu has both the largest $(n, x\alpha)$ cross-section and the largest contribution to the CuCrZr composition. Thus, enriching chromium and/or zirconium would make little difference to the gas production.

Table 2 shows the $(n, x\alpha)$ cross-sections for all natural isotopes of copper, chromium and zirconium. This table clearly shows that reducing the ^{63}Cu fraction and

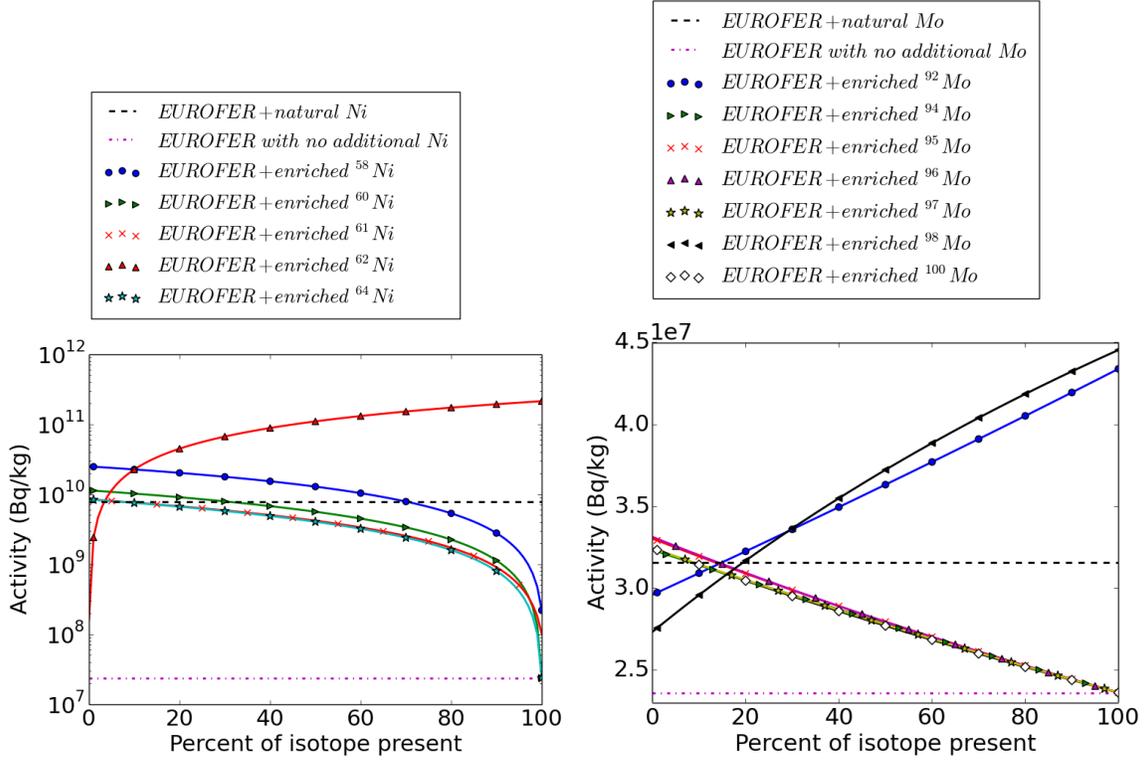


Figure 7: Activity, 100 years after shutdown, of nickel and molybdenum isotopes (100% enrichment, by mass) following a 5 year irradiation at position A in the DEMO model.

increasing the fraction of ^{65}Cu will reduce the production of ^4He in CuCrZr. Incidentally, enriching to increase ^{65}Cu also reduces the production of the long-lived activation product ^{60}Co via the $^{63}\text{Cu}(n,\alpha)$ reaction. The critical bulk concentration of helium, G_{He}^c , for copper has been calculated using the methodology developed by Gilbert et al [32]. These G_{He}^c calculations imply that CuCrZr would maintain structural integrity for 9.61 years. This time could be increased to 32.4 years if copper was to be enriched to 95% ^{65}Cu , as a result of reducing the $He_{\text{APPM}}^{\text{total}}(\text{FPY}^{-1})$ to 19.0. Based on enriching ^{65}Cu to 95% the additional cost of enrichment is approximately £71K/Kg of product. This is based on 3.57 SWU/Kg product, £20K/SWU, 20 cubic meters and resale of waste copper from the enrichment process being sold with a value equal to natural copper. Thus, the total cost for enriching copper is approximately £15B. Thus, an availability of greater than unity would need to be achieved in order to make enriching copper, as a bulk material, cost-effective based on the current cost of enrichment.

4. Summary

This paper has shown that isotopically tailoring elements could improve the longevity of structural materials such as EUROFER and the CuCrZr alloy. However,

some elements either cannot be isotopically enriched as they are monoisotopic (Mn, Nb) or are highly unlikely to be enriched to due technological/practical reasons (Fe).

Section 2 deduced that there there might be scope to reduce the shut-down-activity, -dose-rate and -heating of EUROFER via isotopic tailoring of molybdenum and nickel, rather than replacing them with tungsten and manganese. The capital costs associated with enrichment is large, but could be offset via modest increases of availability, plant thermodynamic efficiency or net power provided by improvements in materials properties.

Section 3 showed that isotopic enrichment can reduce helium production in copper, which could extend the life of CuCrZr alloys at first wall locations. However, the present costs associated with copper enrichment far outweigh the benefits gained via extended lifetime of components.

In summary, isotopic enrichment:

- could be cost-effective for alloying elements in steel, if the enriched alloying elements help to increase availability (via increasing the longevity of replaceable materials) and/or thermodynamic efficiency (via increasing the temperature at which the materials can be operated at. i.e. increasing the ductile-to-brittle transition temperature)
- is unlikely to be cost-effective (at present) for mate-

Material	Element	Composition (%)	Isotope	Abundance (%)	$\sigma_{(n,\alpha)}^{FW}$	$He_{Contrib}$	G_{He}^c	He_{APPM}^{total} (FPY ⁻¹)	T_C (Y)
CuCrZr	Cu	98.95 - 99.33	63	69.15	7.141E-3	5.86E2	615	64.0	9.61
			65	30.85	1.307E-3	4.82E0			
	Cr	0.6 - 0.9	50	4.345	1.223E-2	6.13E-2			
			52	83.789	4.485E-3	4.34E-1			
			53	9.501	6.324E-3	6.93E-2			
			54	2.365	1.700E-3	4.65E-3			
	Zr	0.07 - 0.15	90	51.45	3.909E-4	1.85E-3			
			91	11.22	7.028E-4	7.23E-4			
			92	17.15	8.041E-4	1.26E-3			
			94	17.38	2.611E-4	4.16E-5			
			96	2.8	5.784E-4	1.56E-5			

Table 2: Isotopic compositions for CuCrZr[31], (n, α) cross-sections for all natural isotopes of copper, plus critical bulk concentrations of helium, G_{He}^c . The critical bulk concentrations of helium quoted for CuCrZr are based solely on copper.

rials that require electromagnetic enrichment, such as copper.

- need not be cost-effective in order to be used in the composition of LAMs, as safety, legacy, responsibility and public perception should also be considered (some of these concepts cannot be analysed in terms of economic cost).

Other applications of isotopic enrichment, which can not be cost-benefit analyzed and are not studied in this paper, include:

- the enrichment of the tungsten first wall in order to reduce activated dust production through sputtering and plasma disruptions. This dust may pose a risk during maintenance.
- the enrichment of pipe coatings in order to prevent activated corrosion products in the cooling system.
- reducing the amount of and disposal rating of radioactive waste.
- enhanced materials properties of reactor pressure vessel steels.
- reduced heating and DPA (via selecting isotopes with lower heating and DPA-related cross-sections).
- improving materials properties of RPV steels.

Cost-benefit analysis of enrichment in order to reduce waste disposal has been performed as part of this work, and another programme of work [32], but has not been included in this papers due the the complexity of pricing mechanism for waste disposal. A follow-up paper will focus on the cost benefits of reducing of activity in the long-term and the possible gains to be made from reducing dose levels during maintenance periods.

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