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ABSTRACT

W and W-alloys are among the primary candidate materials for plasma facing components in the design of Fusion reactors, particularly in high heat flux regions such as the divertor. Under neutron irradiation W, like all other elements, undergoes transmutation to its near-neighbours in the periodic table. Additionally He (and H) is produced as a by-product of certain threshold reactions, and this is particularly significant in fusion research since the presence of helium in a material can cause both swelling and a strong increase in brittleness. In this report we present the results of inventory calculations on pure W and give quantitative estimates for He production rates. We also investigate transmutation reactions in certain alloys, which are being considered in an attempt to reduce the brittleness of pure W.

1. INTRODUCTION

In the design of fusion power plants and fusion experimental devices, such as ITER, Tungsten (W) is being considered for various plasma facing components because of its high melting point, high thermal conductivity, and high resistance to sputtering and erosion [1, 2]. As a result, W is a primary candidate material for the divertor armour. Additionally, these qualities also make tungsten a desirable material for use in other parts of the first wall [3]. However, whereas the divertor armour need only have a lifetime of around two full-power years to make fusion commercially viable [4, 5], the first wall components must be typically able to withstand the fusion environment for at least five years [3, 4]. At these longer times, the irradiation-induced embrittlement of W, which shows significant brittleness to begin with, will become a very serious issue, and both modellers and experimentalists are seeking ways to make W more resilient to these effects.

The ductility of W can be improved through the addition of certain alloying elements such as Re, Tc, Ru, Os, Ti, and Co [6]. Some of these, such as Co [7], would be undesirable for fusion applications because of their unfavourable activation characteristics under neutron irradiation, but others have shown promising results. Re, in particular, has been found to reduce the ductile-brittle transition temperature (DBTT) and significantly improve ductility [6], and both mechanical resistance and hardness at room temperature [8], even in concentrations exceeding 25% [8, 9]. However, Nemoto *et al.* [1] have questioned the suitability of such a material under neutron irradiation. At the same time, it has been observed that alloying W with Re can dramatically improve the swelling of tungsten under high dose-rate irradiation [10]. More recently, specifically for structural applications in a fusion reactor, Ta, V, and Ti have also been proposed as possible alloying elements for W (for example, see [11–13]).

Of further significance in the design of W and W-alloy materials for fusion applications is the effect that the neutrons produced by the nuclear reaction have on the atoms of a material. Specifically, the capture of a neutron by an atomic nucleus, and the subsequent emission of other

particles (whose precise nature depends on the energy of the incident neutron), changes the atomic number (number of protons) and/or the mass number (total number of protons and neutrons in the nucleus) of the original atomic nuclide. This process, known as transmutation, may result in the accumulation of products that could be detrimental to the mechanical and engineering properties of the original material [14]. As was reported by Cottrell [2], and others [14–16], the fusion environment will cause W to undergo significant transmutation to first Re and then Os. Cottrell [2] suggested that a 5-year irradiation under first wall conditions leads to initially pure W transmuting into a W-OS-Re alloy with a composition close to that of the sigma-phase for the alloy, which is extremely brittle. Furthermore, the presence of Re as an alloying element may in fact induce the precipitation of a brittle sigma-phase [14].

It is worth realising that transmutation is not always undesirable. In fact, it is precisely this process that will be harnessed in the breeder-blankets of a fusion reactor to produce Tritium through the transmutation of Li. However, there are further problems associated with the transmutation of large atomic nuclei. Many of the possible neutron-capture reactions can lead to the production of protons (*i.e.*, Hydrogen) and α-particles (Helium). These small-nuclei elements can diffuse through the bulk material and cluster in existing cracks or in grain boundaries, leading to an increased brittleness (or hardening) of the material.

There have been previous studies on the transmutation of W, such as the irradiation experiments of Noda *et al.* [16], and the inventory calculations of Forty *et al.* [14] (using an early version of FISPACT - see below), but there is an urgent need to update and clarify the situation in W and its possible alloys. In this report, we re-evaluate the W-transmutation calculations introduced in [2], and consider the transmutation characteristics of various alloying elements - both as isolated pure samples and as part of a possible W-alloy. As highlighted above, He and H are important transmutation products and in this work we give them special attention.

For comparison, the burn-up of other fusion relevant materials, namely Fe, Fe-Cr, and SiC, are also presented.

2. CALCULATION METHOD

To characterise the transmutation properties of W and W-alloys in a fusion-like environment we have made use of the inventory code developed and maintained at United Kingdom Atomic Energy Authority for the last 20 years by Robin Forrest. This code, known as FISPACT, was created to perform calculations of the activation induced by neutrons (and more recently deuterons and protons [17]) bombarding materials in fusion devices. As well as calculating the activity in a material after a particular irradiation, or indeed after a period of post-irradiation cooling, it also keeps track of the number of atoms of each particular nuclide present in the system. As a result, it is possible to measure the burn-up (transmutation) of the starting materials during the irradiation calculation.

Together with external libraries of reaction cross-sections and decay data on all relevant nuclides, FISPACT forms the European Activation System (EASY) [18]. FISPACT uses these libraries to calculate the inventory of nuclides (atoms) produced during the irradiation of a starting material composition with a specified flux and spectrum of neutrons (or deuterons or protons). The current versions of the reference libraries are collectively known as the European Activation File EAF-2007, which, together with FISPACT-2007 [17], form EASY-2007 [18]. However, each version of FISPACT is backwards compatible with previous versions of EAF, and so in the present work we have used EAF-2003 in conjunction with FISPACT-2007 (although both FISPACT-2005 [19], and FISPACT-2003 [20] give identical results with these libraries). The older libraries have been used due to recent validity issues surrounding the newer versions, particularly concerning some of the important He production reactions (usually neutron capture followed by α-particle emission (n, α)) for W and its periodic-table neighbours. Further information on the 2003 libraries can be found in [21] for the cross-sections, and [22] for the decay data.

To perform calculations on a given set of starting materials, FISPACT requires a neutron spectrum for the incident particles. This spectrum, which is supplied by the user, is combined with the cross-section data to produce an effective 1-group cross-section library for all reactions available in EAF. This library is then used for all subsequent calculations with the given spectrum [17]. Following previous work to describe the activation characteristics of all naturally occurring elements [7, 23], in the present study we have used the most relevant first wall (of a fusion device) spectrum from the recent Power Plant Conceptual Study (PPCS) [3, 4]. Specifically, a neutron spectrum calculated for the first wall of Model B has been used, which has a total flux of 1.04×10^{15} n cm^{-2}s^{-1} [24]. In line with the expected life-time of first wall components, the total irradiation time was five years in all calculations.

Figure 1 shows the neutron spectra, plotted as neutron flux per lethargy interval against energy in eV. A lethargy interval is the standard measure for spectra of this type, and is equal to the natural logarithm of the current energy group's upper bound divided by its lower bound. In [24] the neutron flux was tallied using the 175-group "Vitamin J" structure (see [17] for more details), with the lowest group bounded above by 0.1 eV, whilst the highest energy group has an upper bound of 19.64 MeV. The energy profile of the source neutrons used in the calculations (performed with MCNP, version 4C3 - see [25, 26] for more details) followed a Gaussian distribution with a peak at the correct 14.1 MeV for the DT fusion reaction [24]. As a result there was no neutron flux associated with the highest energy-groups.

The total energy fluence produced by the spectrum at the given neutron flux was 4.39 MW m^{-2}. We can use this value in the graphs presented later to convert the irradiation time into MW·y m^{-2} (MegaWatt years per metre squared), which is perhaps a more useful quantity. Note that this total energy fluence is greater than the 2 MW m^{-2} for the 14.1 MeV neutron wall loading associated with PPCS Model B [3, 4] because it takes into account neutrons passing through the

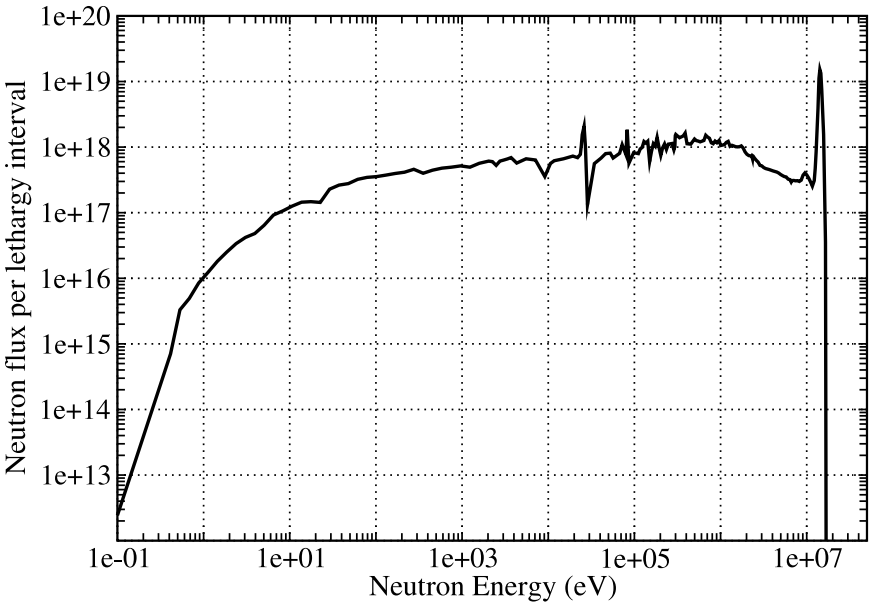

Figure 1: Neutron spectra for the first wall of the Model B conceptual power plant design [3, 4, 24].

first wall in all directions. Specifically, it includes neutrons that may have back-scattered from other parts of the reactor, as well as those coming directly from the plasma, because any neutron entering the first wall can cause transmutation.[1]

In Figure 1 we see that there is a pronounced peak in the flux at the 14.1 MeV mark, which is produced by neutrons hitting the wall directly from the plasma. There is, however, a broad spectrum of reduced energy neutrons at a lower level of flux. These neutrons are produced by the scattering of neutrons from other parts of the reactor.

Using the spectrum described above we have performed FISPACT calculations to measure the transmutation of 1 kilogram pure samples of W and possible alloying elements, including V, Ti, Ta, and Re, as well as specific alloy compositions. For comparison we have also considered 1 kg samples of Fe, Fe-Cr, and SiC. Note that in all cases the starting composition of a particular element follows the naturally occurring abundances of its nuclide isotopes. Table 1 shows the naturally occurring nuclide make-up of the elements considered in the present study. For example, pure W is made up of five different isotopes, one of which, W180, forms only a very small fraction (0.12%), while the other four are more abundant. W184 is the most common, making up 30.64% of naturally occurring W.

We also note that in the present calculations, the well-known phenomenon of self-shielding has not been considered. Self-shielding is the name given to the overestimation of the

[1]Note that the exact scaling relation between the wall loading directly from 14.1 MeV plasma neutrons and the total neutron energy fluence in the first wall will depend on the details of the first wall and blanket materials used in a particular reactor design, which will control the energy spectrum and direction of scattered neutrons.

Table 1: Isotropic Abundance of the naturally-occurring composition of elements considered in this study. Taken from [7]

Element	Atomic number	Atomic Mass	Isotopes (% Abundance)
Ti	22	47.867	Ti46 (8.25), Ti47 (7.44), Ti48 (73.72) Ti49 (5.41), Ti50 (5.18)
V	23	50.942	V50 (0.25), V51 (99.75)
Cr	24	51.996	Cr50 (4.345), Cr52 (83.789) Cr53 (9.501), Cr54 (2.365)
Fe	26	55.845	Fe54 (5.845), Fe56 (91.754) Fe57 (2.119), Fe58 (0.282)
Ta	73	180.948	Ta180m (0.012), Ta181 (99.988)
W	74	183.840	W180 (0.12), W182 (26.50), W183 (14.31) W184 (30.64), W186 (28.43)
Re	75	186.207	Re185 (37.4), Re187 (62.6)
Si	14	28.086	Si28 (92.23), Si29 (4.683), Si30 (3.087)
C	6	12.011	C12 (98.89), C13 (1.11), C14 (Trace)

transmutation rates under fusion neutron-irradiation of heavy elements, such as W, due to the large resonances in the (n, γ) reaction cross-sections [27]. Previously, Cottrell *et al.* [27] have considered this effect, and found that transmutation rates in W can be reduced by several percent. However, they only consider the self-shielding effect for the W186(n, γ)W187 reaction, while in reality all such reactions in this region of the periodic table should be investigated. Work is currently underway to correctly account for these self-shielding effects.

3. RESULTS

Figure 2 shows how W transmutes during the 5-year irradiation time. The graph shows the relative concentrations, in atomic parts per million (appm), of the elements present in the sample during the course of the irradiation. As well as the time in irradiation years, a secondary x-axis is included showing the total neutron fluence in MegaWatt years (MW·y) per unit area. Note that in the graph, and in the other transmutation graphs presented later, elements present in the starting composition (in this case just W) are underlined in the legend. In addition, only those elements whose concentration exceeds one appm after five years of irradiation are included. In practice, FISPACT calculates the production of many minor elements, and while there may be millions (or more) of atoms of such elements present in the system, the actual appm concentration is extremely small - a fact made clear by realising that in a one kilogram sample of pure W, there are of the order of 10^{24} atoms of W. Atomic parts per million (appm) are used for the concentrations shown in this graph (and all others) because this is the measure that is the most useful for the

material modellers, where they are interested in how many atoms of the base element should be replaced by atoms of an alloying element. Also, the alternative representation, weight parts per million (wppm), is misleading as far as the production of He and H is concerned because if wppm is used these light elements will always appear rare in the transmutation of a much heavier element like W.

At the start, before any irradiation takes place, W makes up 100% of the sample, and so in Figure 2 the curve for W starts from 1×10^6 appm. As the irradiation proceeds, the amount of W begins to decrease slightly, as some of the W atoms are transformed, through neutron capture, into other elements. However, even after five years of irradiation, the vast majority of the atoms present in the sample are still nuclides of W, and the concentration of W in total is still around 7.5×10^5 appm. In other elements, such as Ta (Figure 4) and Re (Figure 5), it is possible for the burn-up to be so great that the original element no-longer dominates the composition after the 5-year irradiation.

Along with W, six other elements from the third row of the transition metals in the periodic table are also shown in Figure 2. These are produced at various different levels of concentration, with Os and Re the commonest after five years at 1.3×10^5 and 1.2×10^5 appm, respectively. At the other extreme, Pt is only present at a level of 100 appm after five years, which is not surprising given its distance from W in the periodic table. The reaction pathways required to produce Pt from W are long, with, for example, a sequence of 4 (n, γ) reactions (neutron capture followed by gamma emission) and β^- decays needed to transform W186 into Pt190, which is the shortest possible pathway from a stable isotope of W to a stable (almost) isotope of Pt ((n, γ) and β^- are generally the commonest reactions that increase the neutron and/or proton number).

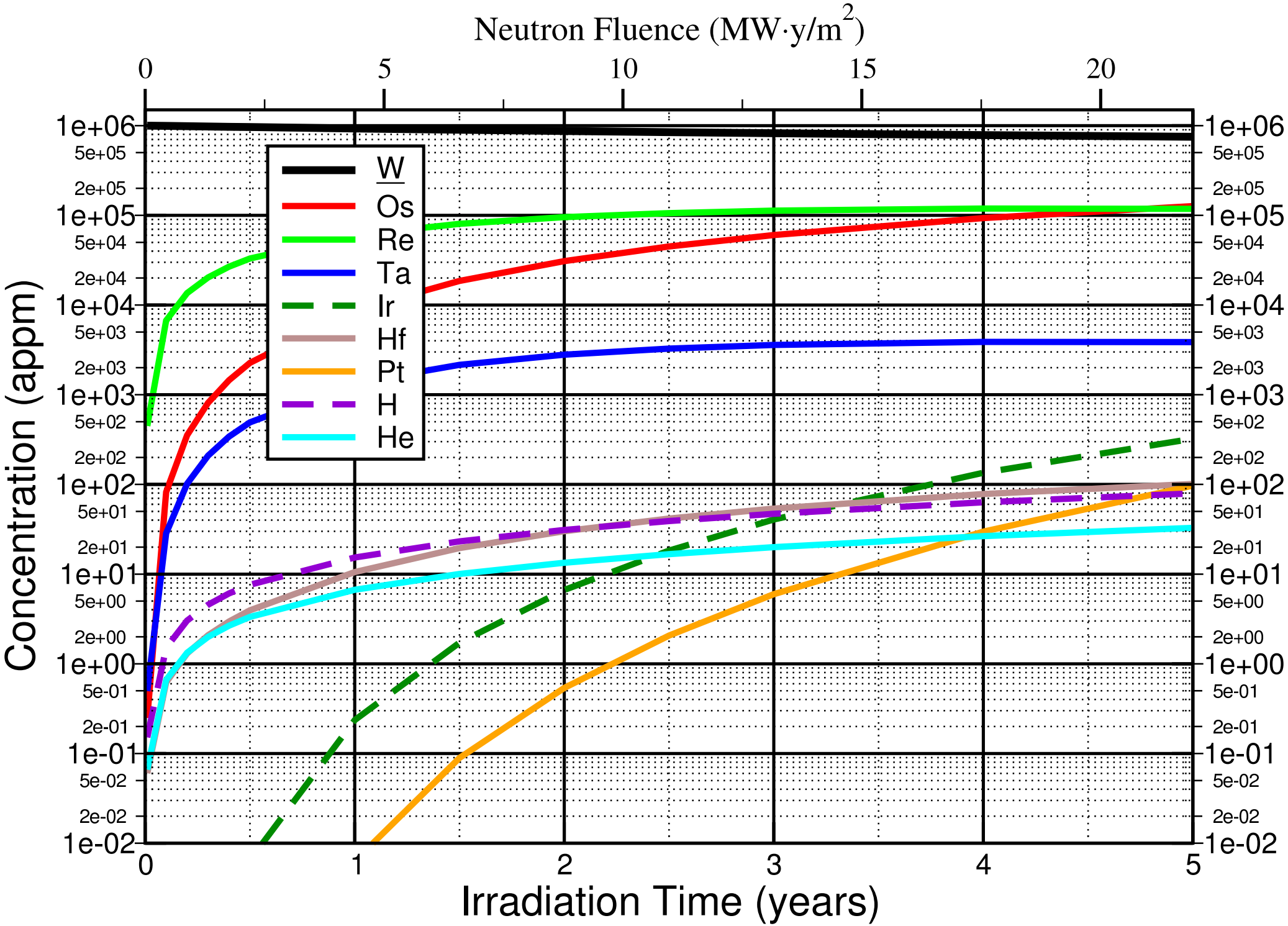

Figure 2: Transmutation of W during a 5-year irradiation under first wall conditions

One particular aspect of these transmutation results that maybe an issue for W-component lifetimes concerns the significant production of both Re and Os. Initially, the concentration of Re is much higher than Os because it is produced directly from W. However, as we shall see later in the transmutation results of pure Re, Re undergoes high levels of transmutation to Os, so that at the end of the 5-year irradiation Os has caught-up and over-taken Re to be the element with the highest concentration in the irradiated material. This finding agrees well with the calculations of Noda *et al.* [16], who found that Os became dominant after a neutron fluence of between 10 and 100 MW·y m^{-2}. However, they observe greater transmutation rates than those presented here because the concentration of W falls below that of Re after only 30 MW·y m^{-2}, which will certainly not be the case in an extrapolation of Figure 2.

As mentioned previously, Cottrell [2] noted that the W-Os-Re alloy composition (approx. 75%-13%-12% after five years) produced by the first-wall transmutation of pure W is very close to the σ-field of the equilibrium phase diagram after the 5-year irradiation. This may result in the the production of σ-phase precipitates, which would be both denser and more brittle that the original bcc (α-phase) structure. Subsequent high tensile stresses produced by both shrinkage and in-service mechanical working could lead to cracking and crumbling of these precipitates, causing a weakening of the material as a whole.

Curves for H and He are also shown in Figure 2. Hydrogen is typically produced during (n, p) reactions, whilst Helium originates mainly from (n, α) (neutron capture followed by α-particle (He^4 ion) emission). The graph shows that H increases to around 80 appm after five years, while He is produced at a slower rate, with its concentration only at around 30 appm at the end of the irradiation.

Whilst the level of He production found above is relatively low and may not, on its own, cause any significant structural changes to a W component, it is worth considering an extreme example. If the spectrum used in the main calculations (see Figure 1) was replaced by one in which only 14.1 MeV neutrons are present, then, with the same total flux, the transmutation results become significantly different. Figure 3 displays the results of such a calculation for pure W. Note that in this case, the total energy fluence is higher than that produced by the full spectrum under the same total flux (23.72 instead of 4.39 MW m^{-2}), and so the secondary x-axis in Figure 3 is different to that in Figure 2.

The first thing to notice in Figure 3 is that, compared to Figure 2, the production of Ta has increased dramatically, whereas the amount of Os in the inventory has almost fallen to zero. The main reason for this change is that with only high-energy neutrons there is an increase in the relative frequency of threshold reactions (*i.e.*, ones where there is a minimum incident-neutron energy required before the reaction cross section becomes significant) such as (n, α) and (n, p), which all produce a daughter nuclide with a reduced atomic mass. For W, this leads to Ta production being favoured, whilst Os creation, which will generally occur through a simple sequence of (n, γ) reactions and β^- decays, becomes relatively less common.

Another consequence of this change in behaviour under *only* high-energy neutron bombardment, is that He (and H) production is increased by an order of magnitude to 240 (and 530) appm after five years. Of course, in a real fusion reactor, the level of flux for 14.1 MeV neutrons would not be so high as the one we have used here (it would be more like the single group flux for that energy in the original spectrum), but this result does demonstrate that it is primarily the high-energy neutrons that are responsible for He production in not only W, but in materials in general. This is one of the reasons why He production is such a significant issue for fusion, as opposed to fission where neutron energies are much lower.

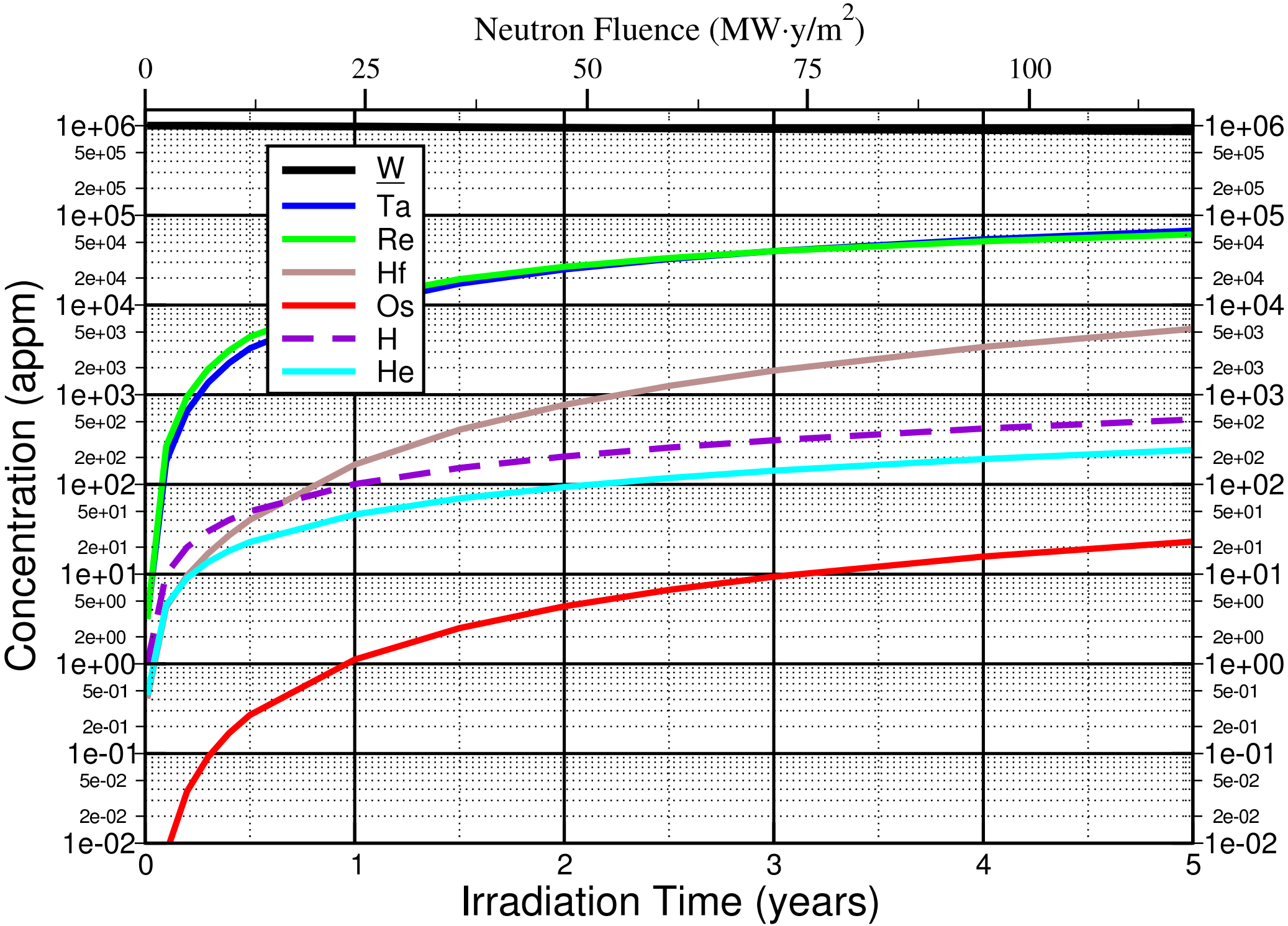

Figure 3: Transmutation of W during a 5-year irradiation under 14.1 MeV neutrons

3.1. TRANSMUTATION OF POSSIBLE ALLOYING ELEMENTS

As mentioned in the introduction, the extreme brittleness of pure W is a significant concern to those designing and planning future fusion experiments and reactors. One possible solution to this may be found by alloying W with certain other metals. In this report we have considered four possible alloying elements: Tantalum (Ta), Rhenium (Re), Titanium (Ti), and Vanadium (V). Figures 4, 5, 6, and 7 show the transmutation results for these four elements, respectively. In all cases the irradiation conditions used in FISPACT were the same as those used above for W under a full first-wall spectrum.

From Figure 4 it is clear that Ta undergoes large-scale transmutation into W. As was noted by Cottrell [2], after only 2.5 years, W overtakes Ta and becomes the element with the highest concentration in the material (76% after five years, compared to only 23% Ta). Calculations performed using an early version of FISPACT, also demonstrated that W overtook Ta on the order of 10^3 days [14]. It is interesting that, hypothetically, a plasma facing component made of Ta would become predominantly W during service, with W levels from pure Ta $\approx$ W levels after the 5-year irradiation of pure W. At the same time, it would not suffer from the same levels of Re and Os production expected in a purely W component (0.34% and 0.16% after five years, respectively, compared to 13% and 12% when starting from pure W). Therefore, the use of Ta as a plasma-facing material might solve any problems associated with the brittle σ-phase in W-Re-Os [2]. However, before such a possibility could be entertained, it would be necessary to perform a detailed investigation of Ta's behaviour in a fusion environment, considering, for example, its neutron-activation characteristics and H embrittlement. Note that He and H production levels during the transmutation of Ta are almost identical to those seen in W.

In contrast, if only 14.1 MeV neutrons are used in the spectrum, then the concentration of W falls dramatically, so that after five years, it only makes up 2.5% (2.5×10^4 appm) of the total. In this scenario, Ta remains dominant, with a concentration still at 8.3×10^5 appm after five years, while Hf makes up most of the remainder at a concentration of 1.4×10^5 appm. This is a further demonstration of the effect of threshold reactions, and reveals that the transmutation of Ta into W involves reactions that require only low-energy incident-neutrons. At the same time, the (n, α) and (n, p)-type reactions which produce He and H, respectively, require the higher energy neutrons, and so their production is greatly increased in Ta when irradiating with only 14.1 MeV neutrons.

The Re transmutation results in Figure 5 confirm that, as we saw in W, the conversion of Re into Os occurs at very high levels. After five years Os makes up over 75% of the material (7.6×10^5 appm), compared to only 22% Re (2.2×10^5 appm). Meanwhile, W is produced in pure Re at a modest rate, with the concentration only reaching 1.5×10^4 appm (1.5% of the total) after five years. The likely consequence of these transmutation results for Re is that any issues surrounding the precipitation of the brittle σ-phase for the W-Re-Os alloy would be magnified if Re was used to alloy W. Since Re would be present from the start in W-Re, Os would be produced

in much larger quantities in the early years of irradiation compared to starting from pure W.

However, if we consider results using only high-energy 14.1 MeV neutrons, then a completely different picture emerges. Os production is reduced dramatically to only 1.3×10^5 appm or 13% of the total, whilst W production increases from only 1.5% to more than 10% (1.0×10^5 appm) when only the high-energy neutrons are used (at the same flux level). As was the case with W and Ta, this indicates that the (n, γ) reactions that control the standard pathways from Re to Os, and other higher atomic number elements, are lower energy reactions. At higher neutron energies, these reactions become dominated by the threshold reactions such as (n, α) and (n, p), which, as well as increasing the production of reduced atomic number elements (*i.e.*, W from Re), also lead to an increase in the inventory of He and H.

Figure 5 shows that, under the broad spectrum of neutron energies associated with the first wall of a fusion power station, He production in Re is slightly lower than in W or Ta, with a concentration of only 22 appm after five years. On the other hand, H production is slightly higher, with a concentration of around 100 appm after the complete irradiation.

For Ti and V, the transmutation picture is somewhat different to those of the heavier elements discussed above. As Figures 6 and 7 show, the rate of burn-up of the starting element (either Ti or V) is much slower than for any of W, Ta, or Re. In the figures the concentration of the input material remains close to 1×10^6 appm (100%) for the entire duration of the 5-year irradiation. In both cases the concentration of Ti and V, respectively, is 9.9×10^5 appm at the conclusion of the irradiation calculation.

Other elements are only produced in (relatively) minor quantities. For Ti (Figure 6) there is some production of Sc (1.1×10^3 appm after five years), Ca (4.0×10^2 appm), and V (10 appm). In V (Figure 7), Cr is the main neighbour-element (in the periodic table) with a concentration of 6.6×10^3 appm after five years, while Ti is produced at a slightly slower rate (2.2×10^3 appm after five years). However, despite this low level of transmutation in general, the production of both He and H is significantly increased compared to the earlier results in pure W, Re, or Ta.

In Ti the amount of He produced after five years is around 910 appm, while in V the situation is better, with only 360 appm of He produced. H production is much higher at 3000 appm in Ti and 2600 appm in V. Thus, adding either of these two elements to W may lead to He production and retention becoming more of a problem than it would have otherwise been in pure W (increased H production may be less of an issue, as this highly reactive element is less likely to build up in materials over a long period of time).

Note that, as in W, Ta, and Re, transmutation calculations under *only* 14.1 MeV neutrons leads to around an order of magnitude increase in He and H production in both Ti and V, with similar increases in the appropriate lower atomic-number elements in each case. As before, this shows that the reactions producing He and H are threshold reactions that require a higher incident-neutron energy before they can take place on a regular basis.

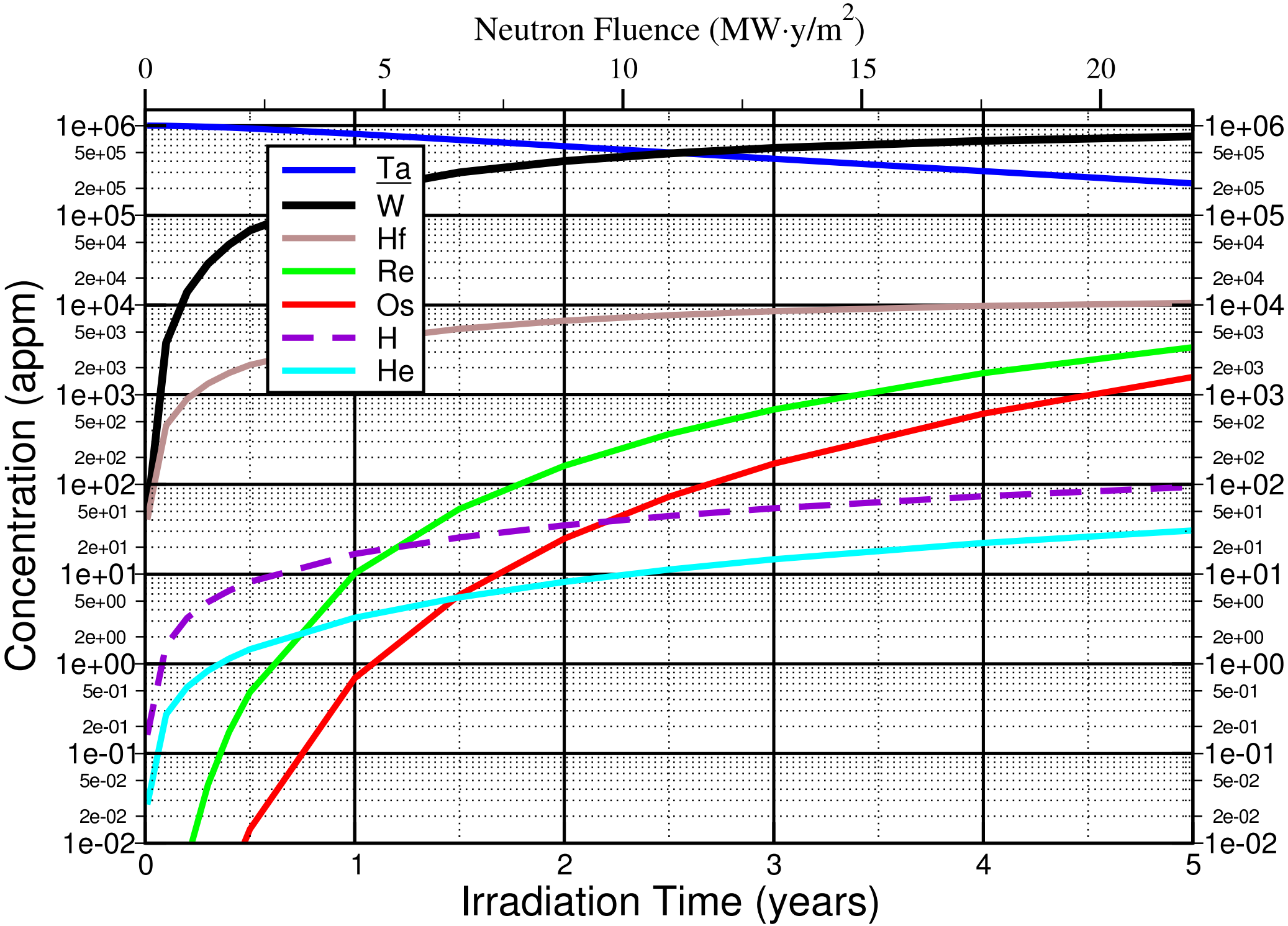

Figure 4: Transmutation of Ta during a 5-year irradiation under first wall conditions

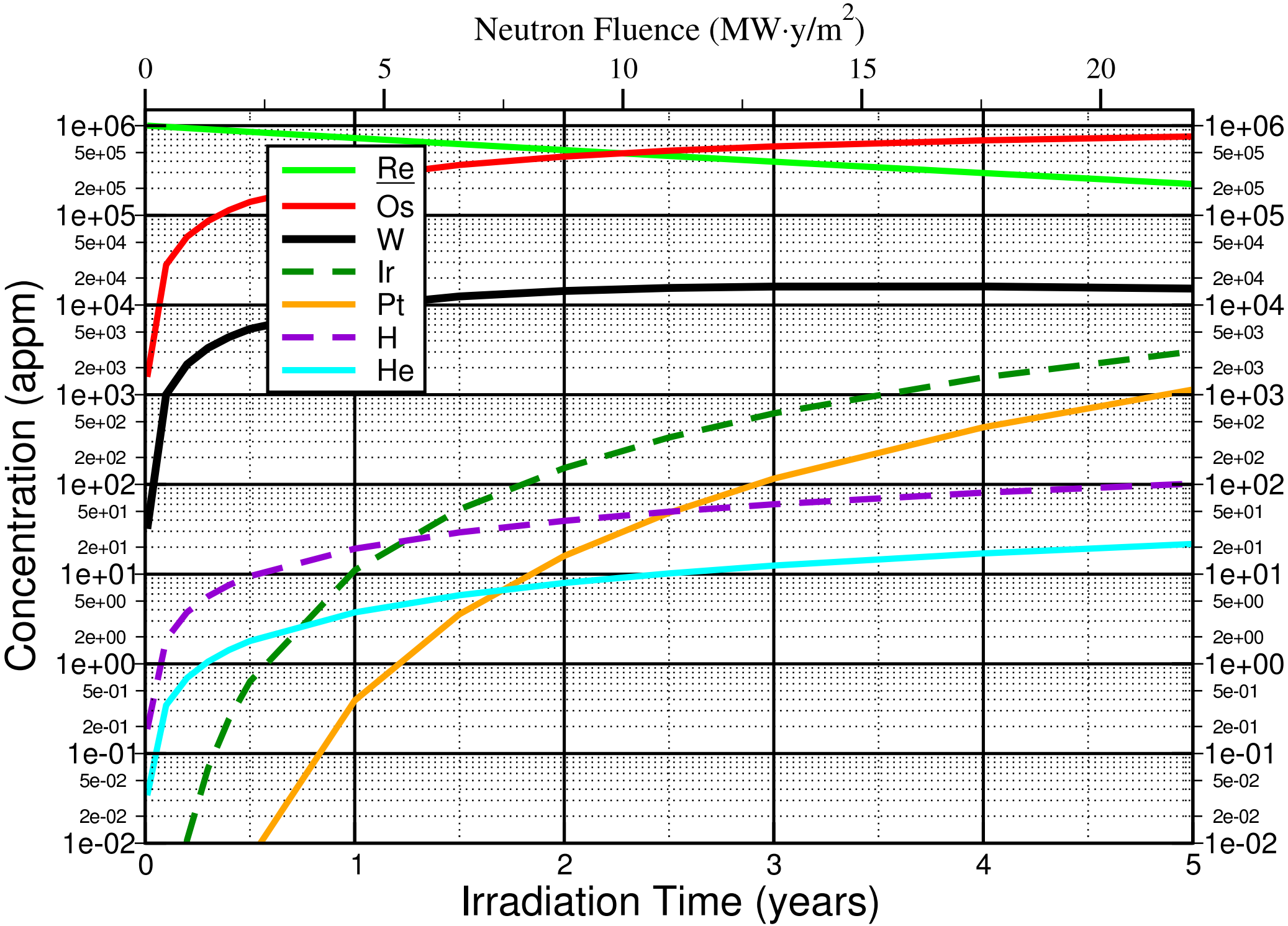

Figure 5: Transmutation of Re during a 5-year irradiation under first wall conditions

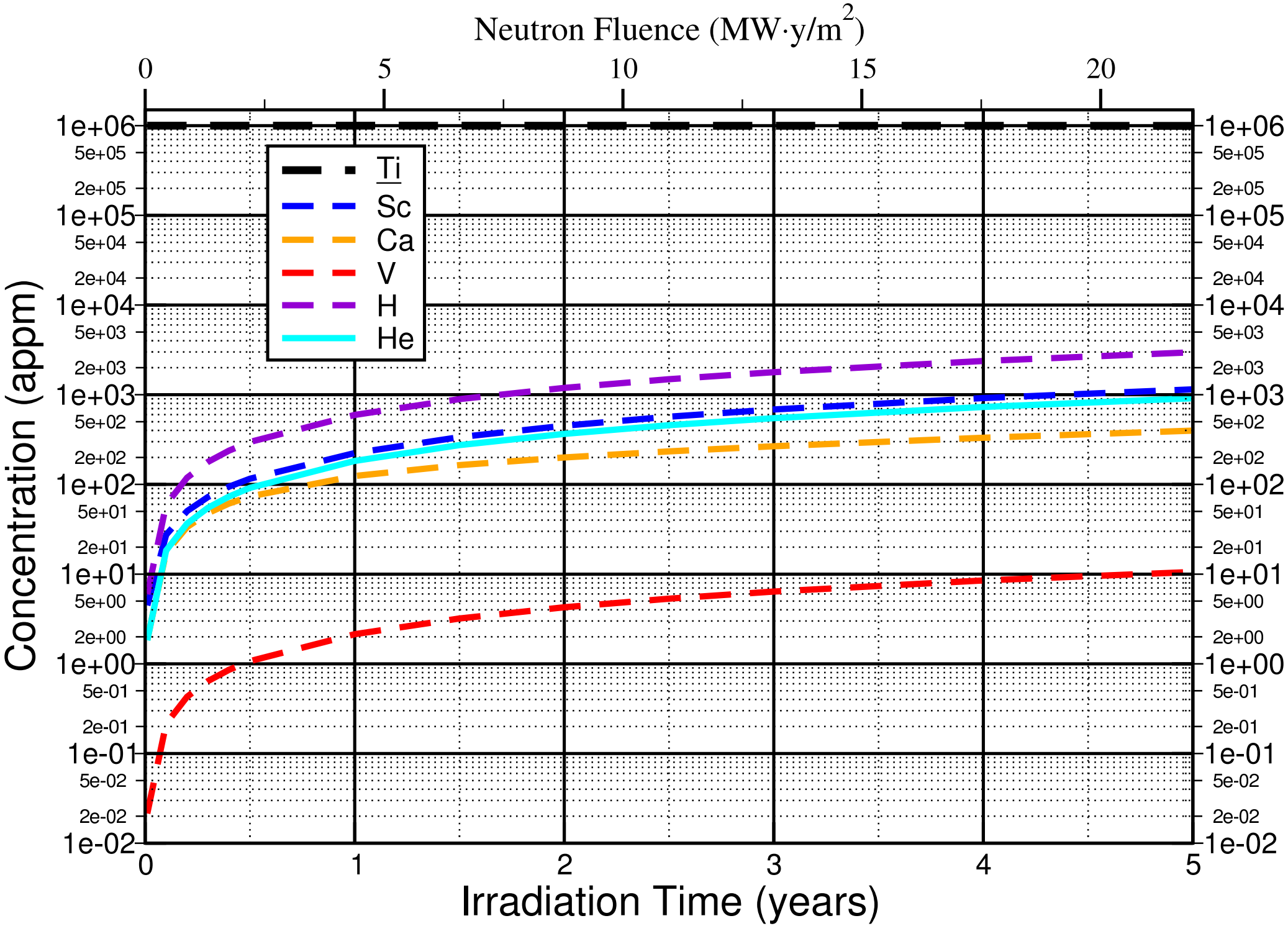

Figure 6: Transmutation of Ti during a 5-year irradiation under first wall conditions

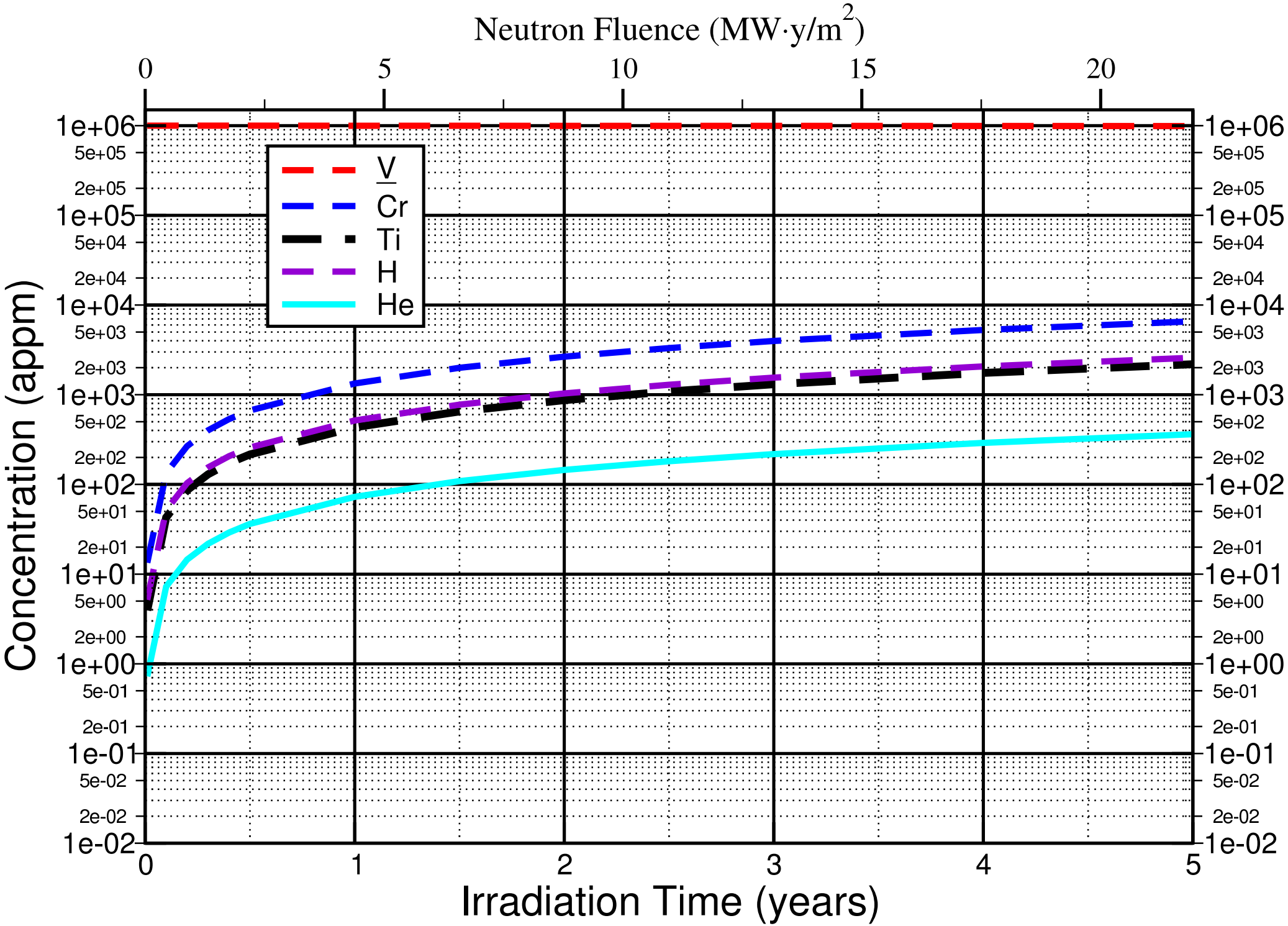

Figure 7: Transmutation of V during a 5-year irradiation under first wall conditions

3.2. COMBINED EFFECTS IN ALLOYS

In the previous section we considered the transmutation of elements that could be added to W to improve its ductility both before and after neutron irradiation. We observed that there may be some desirable and not-so-desirable transmutation consequences associated with these alloying elements. However, to fully appreciate these effects it is worth considering the burn-up of actual W-alloys. As mentioned in the introduction, Re has been considered as an alloying element of W in concentrations of up to 26% (by weight) [1, 8]. Since Re and W have similar atomic masses (see Table 1) this is roughly equivalent to an alloy containing 26% atoms of Re. To investigate the effects of Re addition to W at a 'worst-case-scenario' level, we have calculated the transmutation of a sample containing 30% Re and 70% W (by atoms), which, in standard notation, is W-30atm% Re. For a realistic comparison between the different alloying elements, this 30% by atoms level was also used for Ta, Ti, and V. Note that in the case of Ti and V, a 30% atomic percent composition means that the weight percentages are significantly lower for these elements in the starting material because of the greater disparity between their atomic weights and that of W. Using the atomic masses from Table 1 we can calculate the corresponding weight percentages for these 30% alloy compositions. 30% by atoms Re corresponds to 30.269% by weight (*i.e.*, slightly more than 30 because Re has a higher atomic mass than W), for Ta the value is 29.668% by weight (Ta is slightly lighter than W). In comparison, for Ti a 30% by atoms corresponds to only 10.039% by weight, while for V it is slightly higher at 10.615%.

To produce the transmutation results presented in the subsequent graphs (Figures 8, 9, 10, and 11) there are two options. Either the appropriate weight fractions can be fed into FISPACT and a new inventory calculation performed, or the transmutation results obtained with the pure elements can be utilised. The second of these is the more straightforward, but we have checked by performing the necessary FISPACT runs that both methods produce the same burn-up results. From pure element transmutation calculations the concentration C_X in appm of an element X in a combined alloy Y-Z after a total irradiation time t is given by

$$C_X = \frac{N_{(Y\to X)}W_Y^{\%} + N_{(Z\to X)}W_Z^{\%}}{\sum_S (N_{(Y\to S)}W_Y^{\%} + N_{(Z\to S)}W_Z^{\%})} 10^6, \tag{1}$$

where $W_Y^{\%}$ is the fractional percentage by weight of element Y in the starting alloy (similarly for Z). $N_{(Y\to X)}$ is the number of atoms of element X produced after an irradiation time t from 1 kg of element Y (similarly for Z, S, *etc.*). The denominator is essentially the total number of atoms present in the transmutation calculations of elements Y and Z, with contributions weighted according to $W_Y^{\%}$ and $W_Z^{\%}$, respectively.

For W-30atm% Re, the graph in Figure 8 shows that the production of Os is very significant. It reaches a concentration of 3.2×10^5 appm after five years, while the concentration of Re drops by half from 3×10^5 appm (30%) to 1.5×10^5 appm in the same period. W is reduced to 5.3×10^5 appm in the final composition. Other elements are produced in relatively minor

quantities, which is in agreement with the findings for pure W and Re. There is 0.29% Ta (2.9×10^3 appm) and 0.11% Ir (1.1×10^3 appm) after five years, with Pt and Hf present in even lower concentrations. Since He and H production were broadly similar in both W and Re it is not surprising that the production of these elements in the alloy is also similar, at 29 and 86 appm, respectively, after five years.

As we suggested in the previous section, the production of Os directly from the Re starting material would almost certainly lead to a greater potential for precipitation of the brittle σ-phase than in a purely W initial composition. Thus any brittleness induced in W by the combined presence of Re and Os is likely to be magnified if Os production is increase, which it is if Re is present from the start. It seems that alloying W with Re, whilst beneficial in non-irradiation environments, might have a detrimental effect on component lifetime under neutron irradiation. However, we note that there is still significant uncertainty about the phase diagram for W-Re-Os, particularly under non-equilibrium conditions, and there is an urgent need for further study to confirm (or not) the predictions made by Cottrell [2] and others.

Figure 9, which shows the burn-up of W-30atm% Ta under first-wall neutron irradiation over five years, suggests that alloying W with Ta might lead to a different set of problems to W-Re. It is true that the levels of both Re and Os remain below 10%, even after five years, reaching 8.4×10^4 appm and 8.9×10^4 appm, respectively, and hence there is a reduced chance of brittle-phase precipitates building-up in any significant quantity (certainly compared to W-Re, if not pure W as well). However if we observe the change in Ta concentration during the irradiation, we discover that it falls from the initial 30% level to around 7% (7.1×10^4 appm) after five years. This means that any benefit produced by the addition of Ta in the first place would have long been lost, and the material would most likely suffer from the same draw-backs associated with pure W, such as high DBTT.

On the other hand, the reduction in Ta is associated with an increase in W, which increases from the 70% level at the start to over 75% after five years in our 30% Ta example. This might lead to some benefit in a real material if W has significantly better irradiation characteristics compared to Ta. However, Ta is likely to be added at a much smaller concentration level than we have considered here, so any improvement caused by the increase in W is likely to be minimal and out-weighed by the negative effects associated with Ta reduction and Re/Os production. As was the case with W-Re, the He and H production levels in our W-Ta calculation are almost identical to those in pure W.

In W-30atm% Ti and W-30atm% V (Figures 10 and 11, respectively) the transmutation results are significantly different to those seen in W alloyed with its close neighbours in the periodic table. In both cases there is a much greater inventory of elements because Ti and V produce a completely different set of transmutation products to W, with He and H being the only overlap. Also, as we saw in the previous section, the burn-up of these lighter elements is much less than any of W, Re, or Ta, and so when they are added at a concentration of 30 atomic percent to W,

they essentially remain at this level throughout the whole 5-year, first-wall irradiation (see Figures 10 and 11). This also means that the minor transmutation products produced from Ti and V are largely dominated by those produced from W, even when taking into account the larger proportion of W in the starting composition.

As we saw with W-30atm% Ta, both Re and Os reach a concentration of around 10% during the irradiation of both W-Ti and W-V, which demonstrates that it was primarily the burn-up of W in W-Ta that led to the production of these elements (*i.e.*, Re and Os are not produced from either Ti or V, and so are only produced through the transmutation of W).

Perhaps the most critical change associated with alloying W using either Ti or V concerns the production of He and H under neutron irradiation. As was seen in the transmutation of pure Ti and V, the production of these very light elements is increased compared to transmutation of W, Re, or Ta. In 30% alloys, this leads to He (H) reaching a concentration of 300 appm (950 appm for H) in W-Ti (Figure 10) and 130 appm (830 appm for H) in W-V (Figure 11) during the 5-year irradiation. Even though the concentrations of Ti and V used in our alloys are higher than those likely in a real material, the increase in He (and H) caused by their addition should still be carefully considered when designing materials for advance fusion applications.

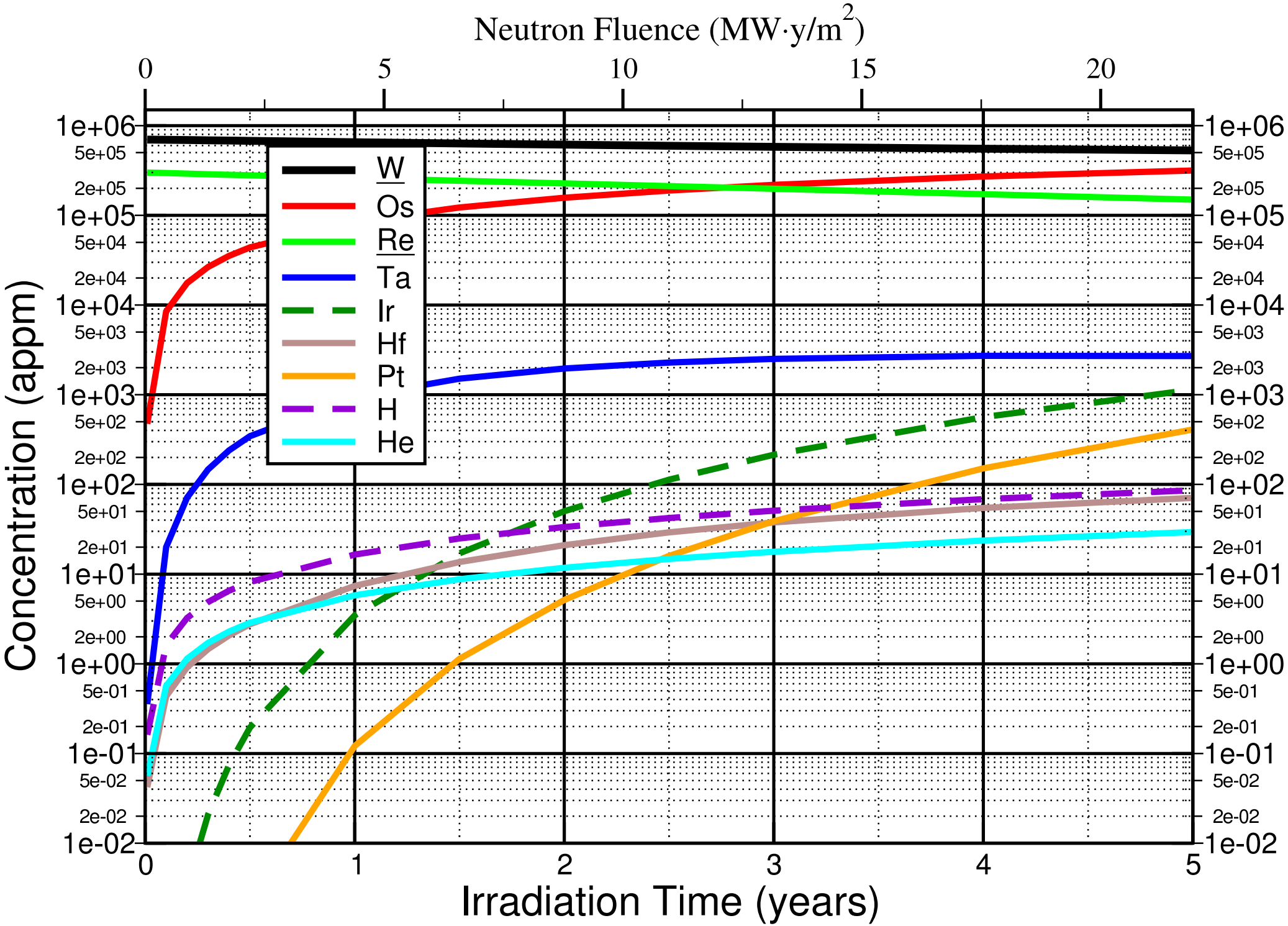

Figure 8: Transmutation of a W-30%Re (by atoms) alloy during a 5-year irradiation under first wall conditions

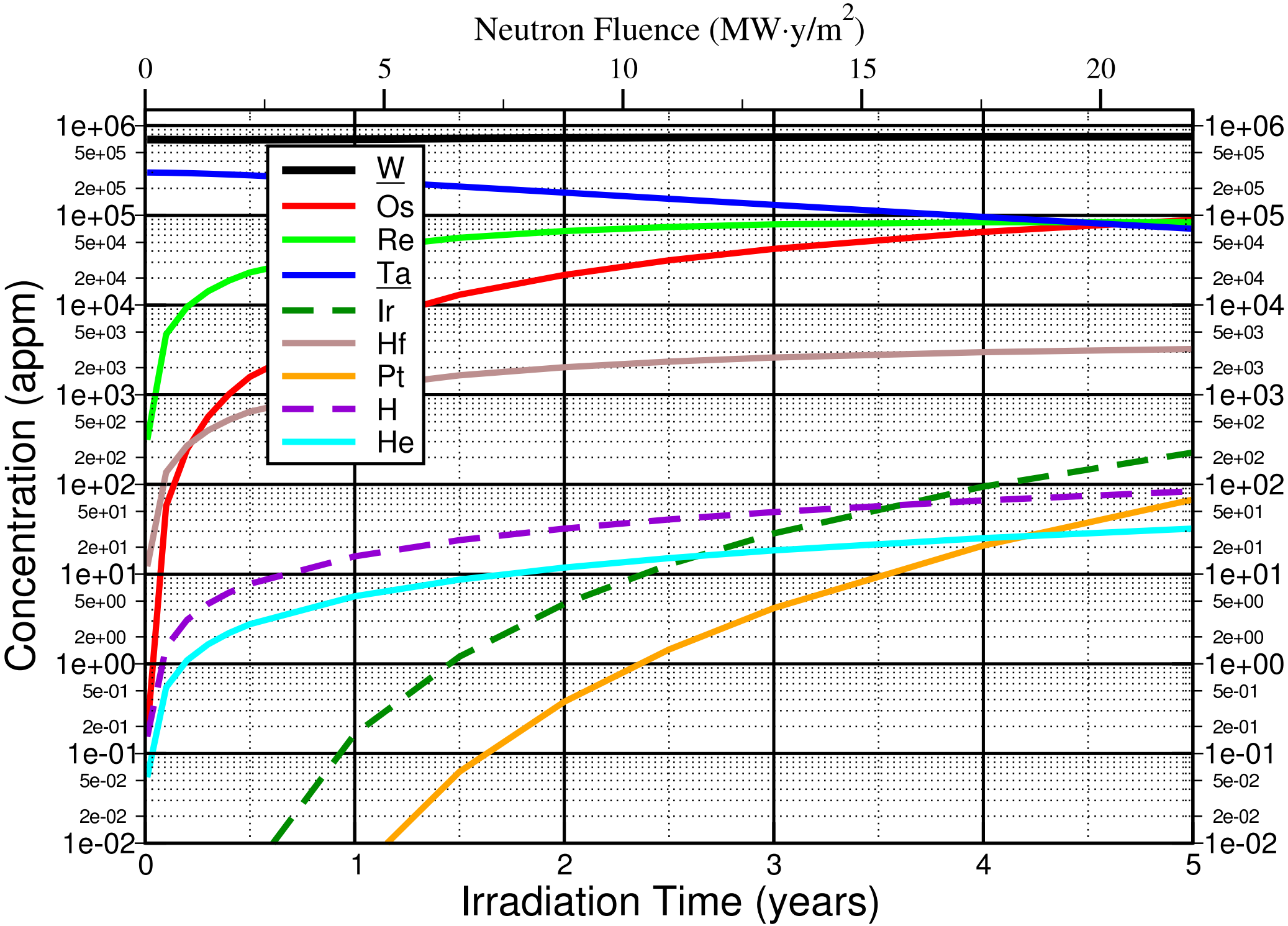

Figure 9: Transmutation of a W-30%Ta (by atoms) alloy during a 5-year irradiation under first wall conditions

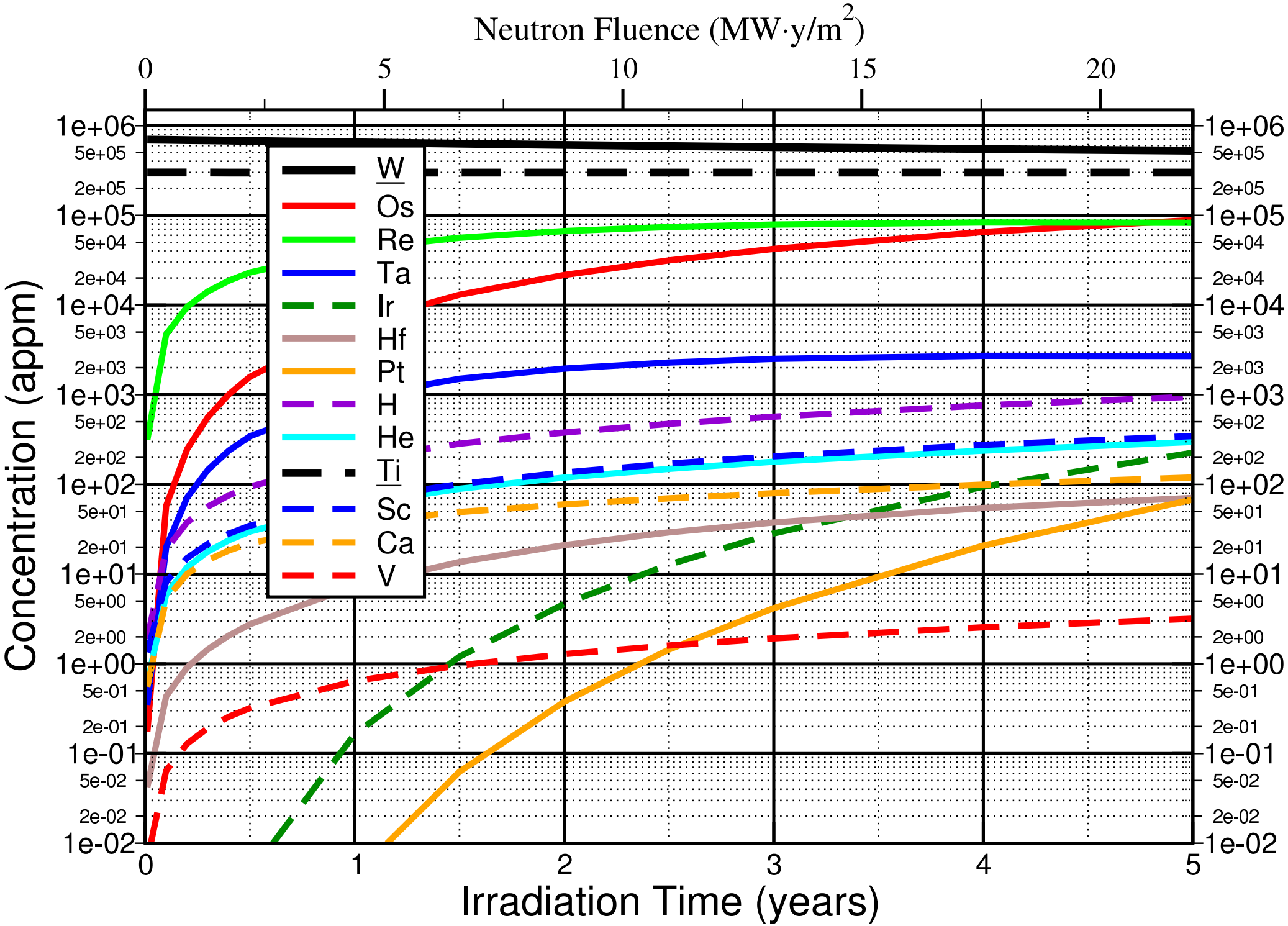

Figure 10: Transmutation of a W-30%Ti (by atoms) alloy during a 5-year irradiation under first wall conditions

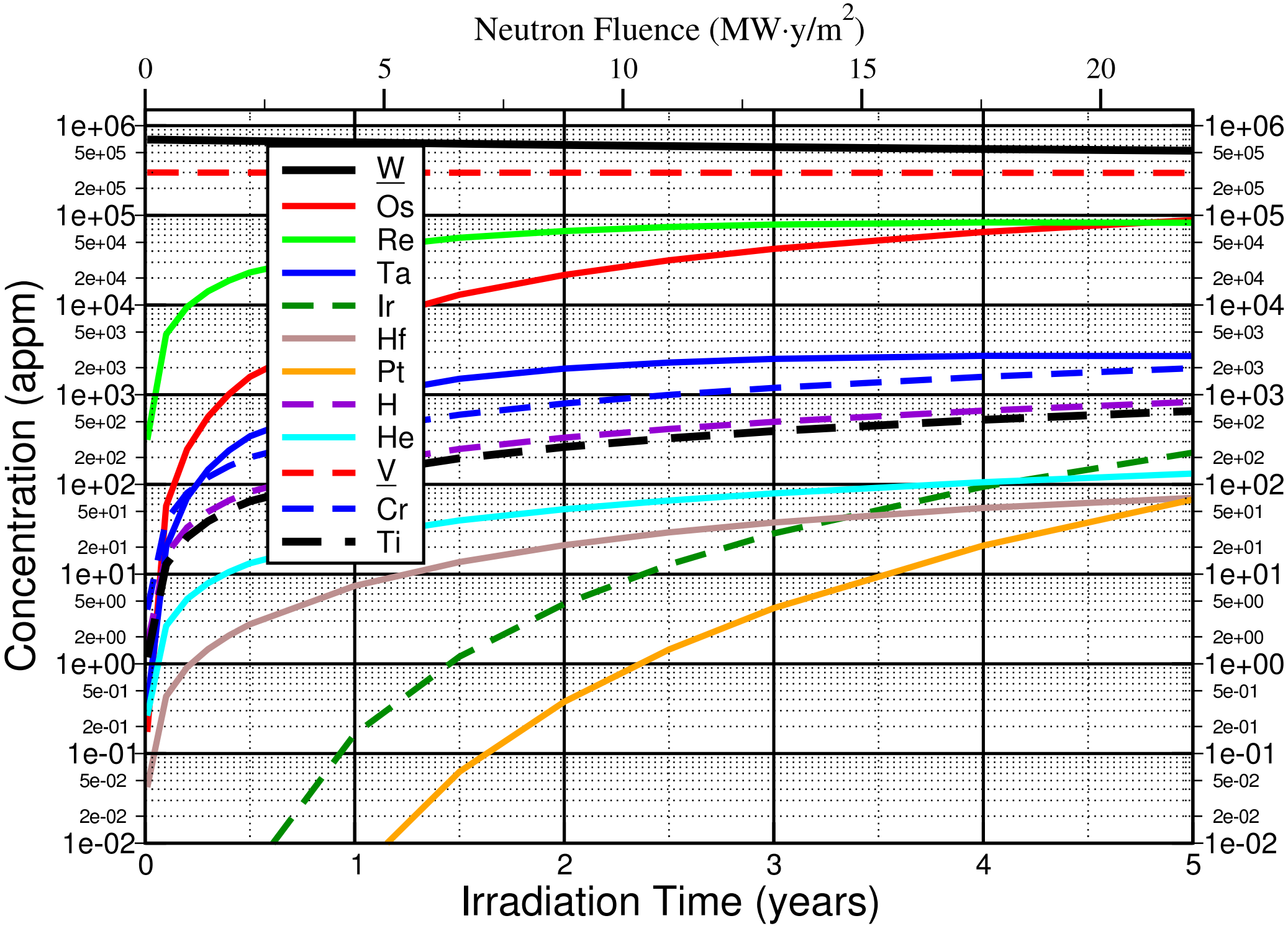

Figure 11: Transmutation of a W-30%V (by atoms) alloy during a 5-year irradiation under first wall conditions

3.3. COMPARISON WITH OTHER FUSION MATERIALS

To put the results for W and W-alloys into perspective we also present here transmutation calculations for some of the other primary candidate materials for in-vessel components of fusion reactors.

Steels are widely used in both experimental fusion devices and nuclear fission reactors, and will certainly be the main structural material used in future fusion devices and reactors. Fe is the primary constituent of steels, and in ferritic-martensitic steels (body-centred cubic structure or bcc) makes up more than 80 weight % of the total. Meanwhile, Cr is the main alloying element in reduced-activation Ferritic-Martensitic (RAFM) because it helps to reduce the radiation-induced swelling of the steel [28], which is a significant problem in the popular austenitic steels (face-centred cubic or fcc) [29, 30]. A general consensus has now been established for a Cr concentration of 9% (since the atomic mass of Cr is similar to Fe this can be considered as either weight or atomic percent). Above this level, at fusion operating temperatures and irradiation levels, Cr begins to segregate into clusters, which causes a loss of ductility and corrosion resistance [31, 32], while below 9% Cr remains in solute and exhibits short-range ordering [31, 33]. In this study we have considered the transmutation of both pure Fe (Figure 12) and Fe-9atm% Cr (Figure 13).

Alternatively, silicon carbide (SiC) composites are also considered as possible first wall structural materials because of their low activation characteristics under neutron irradiation [34]. They also show good fracture resistance and have excellent high temperature mechanical properties (see, for example, Raffray *et al.* [35], Jones *et al.* [36], and Riccardi *et al.* [37]). Here we consider a 50-50 mix (by atoms) of Si and C, which is usual in these materials. Note that in mass terms this corresponds to 70.045% by weight Si and 29.955% by weight C (*i.e.*, Si has a higher atomic mass).

Figure 12 shows the burn-up response of a 1 kg sample of pure Fe under the same 5-year first-wall irradiation conditions used elsewhere in this report. As with Ti and V, which are both in the same row of the periodic table (transmutation trends follow rows, rather than groups), the transmutation of Fe under the conditions applied here is fairly minor. The most common elements after five years are Mn and H, but these are only produced at concentrations of 5.5×10^3 and 5×10^3 appm, respectively, which, in percentage terms, are both around 0.5%. He is produced at concentrations reaching 1100 appm, which is higher than in Ti and V, and more than a factor of 10 greater than in W. This trend in He production under irradiation agrees qualitatively with previous studies. For example, Übeyli and Demir [38] have recently calculated He production in possible structural alloys in a fusion first wall as a function of the thickness of a thorium molten salt liquid shield layer, which has been proposed as a way to avoid frequent replacement of the first wall. They observed the greatest He production, at all liquid layer thicknesses, in Ferritic steels (*i.e.*, predominantly Fe), with V-alloys slightly better, and a W-Re alloy the best of all, with an order of magnitude less He produced compared to the other two.

Adding 9 atomic percent Cr to Fe has very little effect on the transmutation characteristics compared to pure Fe. The transmutation results for Fe-9atm% Cr are shown in Figure 13. The only differences in composition after five years, apart from the presence of 9% Cr, are that Ti now appears as a minor product and the amount of V has increased slightly, but these are present in very small amounts (around 0.01 and 0.1% of the total, respectively) and are not significant. He and H production levels are almost identical to those seen in pure Fe.

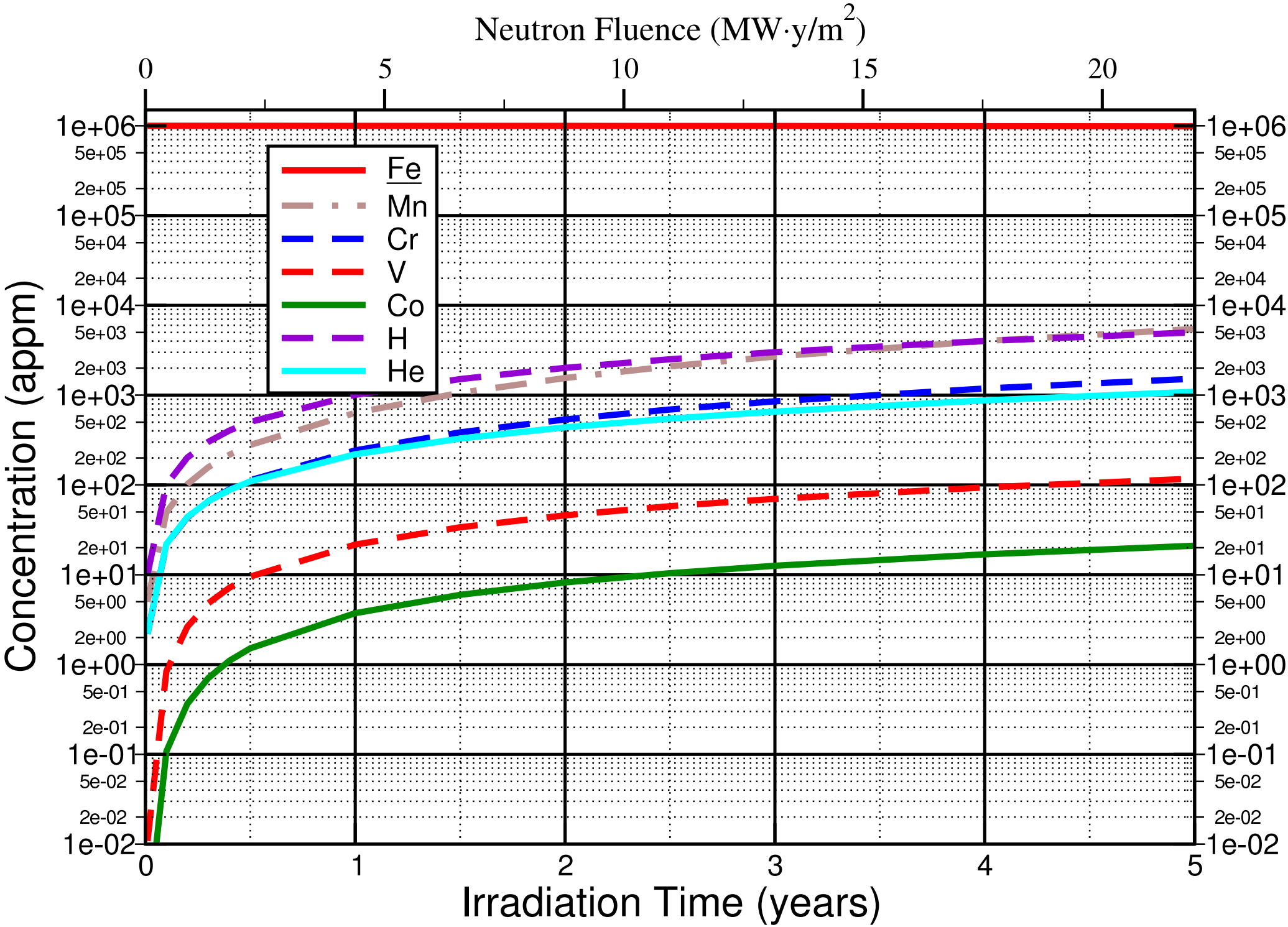

Figure 12: Transmutation of Fe during a 5-year irradiation under first wall conditions

In experimental studies of Fe-9% Cr martensite it has been found that bubbles of He can significantly increase brittleness when present at concentrations of around 5×10^3 appm [39, 40], although it is not clear if the same effect would be observed at the lower He concentrations observed in our calculations, and there is also H to consider. In any event, it is likely that ferritic/martensitic steels (austenitic steels are known to suffer from He embrittlement at concentrations as low as 10 appm [41]) can withstand slightly higher levels of He (and H) impurity than W and W-alloys because of the difficulties in manufacturing these latter materials. Whereas the melting point of Fe (and hence steels) is around 1500 K, W has a melting point of 3410 K, which makes the production of large quantities of homogeneous metal almost impossible with current technology. Current processes for W involve sintering of powder (or pellets), leading to an irregular and perhaps weakened microstructure in the finished product.

On the other hand, in the transmutation of SiC (Figure 14), the production of He is much greater than in any of the other materials we have studied so far. After five years of irradiation, the concentration of He is 1.1×10^4 appm or 1.1 % of the total. This is 10 times higher than in Fe and 100 times greater than in W. These results are in good agreement with previous calculations, where He production was estimated to be around 2000 appm per year and around 10000 appm during component lifetime [42, 43].

Threshold reactions, such as (n, α), still play a major role in the production of He from Si and C, which is illustrated by the fact that He concentrations rise by another factor of six to 6.6×10^4 appm in a purely 14.1 MeV neutron burn-up. However, unlike the situation with other elements, from C there are now pathways involving these reactions that can also result in the production of other isotopes of He (*i.e.*, other than He^4). This seems to be a contributing factor to the increase in He production. Indeed, in [7] it was found that He was the chief transmutation product from C (even in wppm), although He is still a significant product in Si transmutation. While the concentration of He in our calculation was higher than any other transmutation product, H was not far behind. After five years its concentration was found to reach 4.2×10^4 appm (0.44% of total). The remaining products are all produced at lower concentrations, with Mg the highest at around 0.26% of the total.

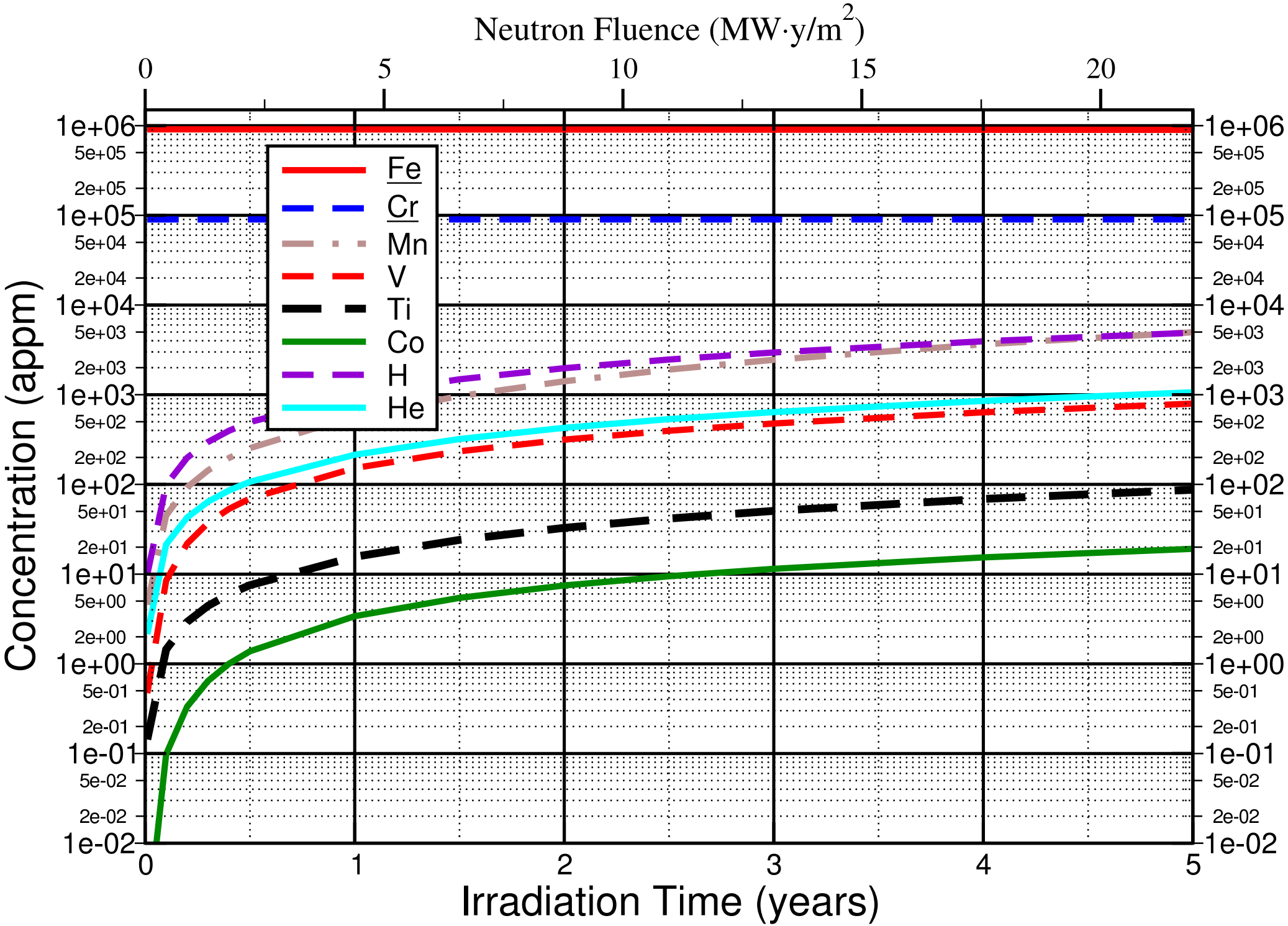

Figure 13: Transmutation of Fe-9%Cr during a 5-year irradiation under first wall conditions

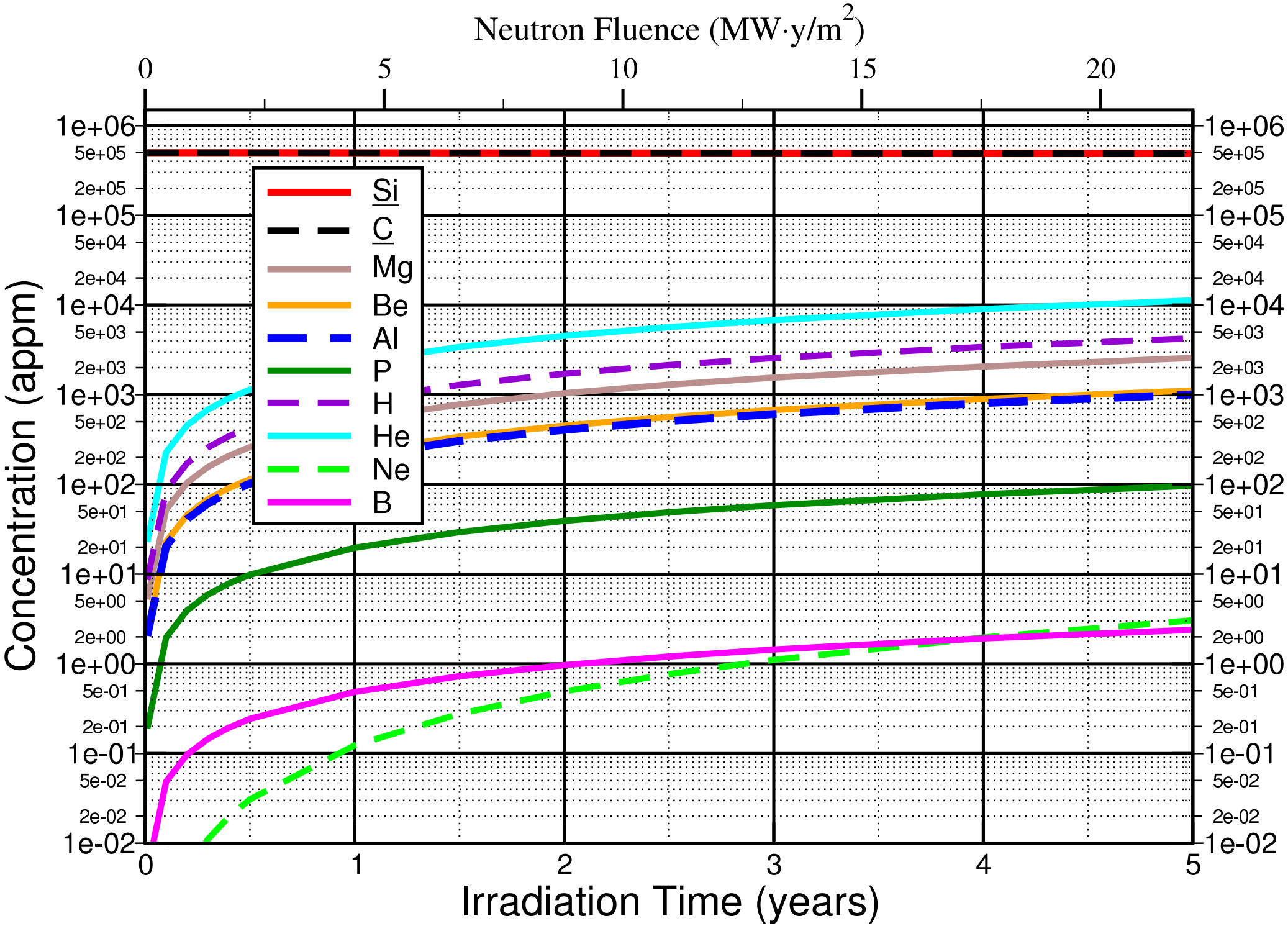

Figure 14: Transmutation of Si-C during a 5-year irradiation under first wall conditions

4. CONCLUSIONS

In this report we have investigated the transmutation properties of W and possible W-alloys. Calculations show that W and its near-neighbours in the periodic table undergo significant burn-up under first-wall fusion reactor conditions. In W our results agree with previous calculations, and demonstrate that there is an urgent need to understand the consequences associated with the significant production of Re and Os, particularly concerning the possibility of producing brittle σ-phase precipitates. If there is a problem, then using Re as an alloying element might not be appropriate in a neutron-irradiation environment because it experiences extreme levels of burn-up to Os. At the same time, Ta suffers from high transmutation into W, and so the benefits associated with using this element to alloy W may be quickly lost in a fusion reactor. It might be the case that the high burn-up rates in such heavy elements will make them undesirable from an alloying perspective, and may even raise issues with W itself [2].

However, He (and H) production during the transmutation of such heavy elements like W, Re, and Ta is comparatively low, with typical concentrations of only 30 appm (80 appm for H) after a full 5-year first wall irradiation. At the same time, lighter elements, such as Ti, V, and Fe, produce an order of magnitude more He and H, with C an order of magnitude worse again.

Ti and V may be more promising candidates for alloying elements of W because they show much slower rates of burn-up under neutron irradiation. Therefore, the initial alloying concentration levels would be fairly constant during the expected 5-year lifetime of first wall components. However, the He and H production rates from these materials are significantly higher than in W itself. Careful consideration will be needed when using such elements, and a maximum concentration limit is likely to be imposed by the need to reduce He and H production. It is also worth noting that Ti and V, as well as other elements from this row of the periodic table, have better activation characteristics compared to W and its neighbours [7].

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