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# **LOW ACTIVATION VANADIUM ALLOYS**

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## ABSTRACT

The properties and general characteristics of vanadium-base alloys are reviewed in terms of the materials requirements for fusion reactor first wall and blanket structures. In this review attention is focussed on radiation response including induced radioactivity, mechanical properties, compatibility with potential coolants, physical and thermal properties, fabricability and resources. Where possible, properties are compared to those of other leading candidate structural materials, e.g. austenitic and ferritic/martensitic steels.

Vanadium alloys appear to offer advantages in the areas of long-term activation, mechanical properties at temperatures above 600 °C, radiation resistance and thermo-hydraulic design, due to superior physical and thermal properties. They also have a potential for higher temperature operation in liquid lithium systems.

Disadvantages are associated with their ability to retain high concentrations of hydrogen isotopes, higher cost, more difficult fabrication and welding. A particular concern regarding use of vanadium alloys relates to their reactivity with non-metallic elements, such as oxygen and nitrogen.

## INTRODUCTION

The radioactivity induced in first wall and blanket structures of fusion reactors, raises concerns regarding safety, maintenance and waste disposal, e.g [1]. These concerns have reached a stage now where availability of low activation materials is considered to be of crucial importance for fusion power to become reality. Public acceptance of fusion power may be difficult to obtain, unless it can be demonstrated that it does not lead to production of large quantities of long-lived radioactive waste [2].

Radioactivity in D-T fusion reactors is primarily induced in surrounding materials by interaction of 14 MeV neutrons emerging from the fusion process with those materials. Since each element has its own specific radiological response, the composition of the irradiated materials determine activation levels and decay characteristics. The obvious approach, therefore, would be to select structural materials which are composed of elements with low induced and rapidly decaying radioactivity. Naturally, these materials should in addition have the required physical, mechanical and chemical properties.

Vanadium-base alloys are considered to be strong candidates for the first wall/blanket structure of a post-NET/ITER fusion reactor, e.g. [3]. These alloys have the potential of being structural materials with the lowest long-term activation, depending on the alloying elements and impurities, e.g. [4]. Vanadium alloys also compare favourably with other candidate materials, such as austenitic and martensitic/ferritic steels, as far as a number of additional important properties is concerned.

Vanadium alloys were evaluated as fast breeder fuel cladding material during the 1960's. Alloy development programmes were conducted at Argonne National Laboratory, Westinghouse Electric Company and Kernforschungszentrum Karlsruhe. Many of the data generated during these programmes are reviewed in [5] and [6].

In this paper some general radioactivity aspects of materials for the first wall/blanket structure of fusion reactors are briefly discussed. Published activation data are shown [4], which point to vanadium, chromium and titanium as being of prime interest. In the remainder of this paper a number of relevant properties and characteristics of vanadium alloys are summarized and, where possible, compared to those of austenitic and martensitic/ferritic steels.

## ACTIVATION CHARACTERISTICS

Different elements, and even isotopes of the same element, exhibit different nuclear response when bombarded by neutrons, resulting in a variety of radioactive products, activation levels and decay times. Activation calculations are performed by means of sophisticated computer codes using comprehensive nuclear data libraries, e.g. [7]. Fig. 1 shows activity levels and subsequent decay after shutdown for several relevant elements, as calculated by Cierjacks and Hino [4]. These activities were calculated for the first wall of a liquid lithium cooled tokamak-type fusion reactor. The target material is assumed to be a pure element in its natural isotopic composition. The first wall is that of the conceptual Culham Demo reactor [8] with a first wall loading of  $5 \text{ MW}\cdot\text{m}^{-2}$ . The irradiation period before shutdown is 2.5 years.

Activation levels and decay times depend on the neutron flux and spectrum and on the exposure time. This explains differences in calculated levels of activation as reported for various conceptual reactor designs. Also, the nuclear data base is updated when new experimental information becomes available. The more recent activation data, however, agree on a number of important aspects. The activation levels at shutdown vary by about three orders of magnitude, while differences in residual activity after periods in the order of 100 years are very much larger. The elements with lowest activity at shutdown, e.g. Si, do not necessarily have the shortest decay times.

Dose rates associated with the activation levels are shown in Fig. 2. Also indicated in this figure is the dose rate below which materials may be handled without protection in a controlled environment [9]. The time scale of decay times can be divided into four regions of radiological interest: 1. from shutdown to one day, relevant to accident situations, 2. from one day to one month, relevant to maintenance and repair operations, 3. from a few years to tens of years, relevant to possible recycling or short-term storage prior to permanent disposal and 4. larger than 100 years, relevant to eventual release of radioactive materials to the environment, for example due to leaching by groundwater, etc.

Potential benefits to be derived from the use of lower activating materials, clearly, would not include the possibility of hands-on maintenance. Shielding will certainly be required, but could be minimized when materials consist of elements as Si, Al, V and Cr. For the longer term, after decay periods of tens of years, a number of elements such as V, Cr, Mn, Ti and Fe have strongly reduced dose rates. Low activation materials have recently been defined as "materials that under neutron irradiation do not generate intensely radioactive, long-lived radioactive isotopes" [2]. According to this definition only alloys composed of these elements would qualify as low activation materials.

Since Mn is relatively volatile at high temperatures and Ti produces a volatile radioactive Ca isotope, application of both elements should be limited because of short-term safety considerations. Chromium-based alloys are not favoured because of very poor ductility and associated problems of fabrication and joining. This leaves only V and Fe as base elements for low activation materials. Presently, the main approaches to low activation materials are indeed represented by modification of steels, with strongly activating elements as Nb, Mo and Ni replaced by less activating elements, and by development of vanadium base alloys.

## STRUCTURAL MATERIALS REQUIREMENTS AND CONSIDERATIONS

Requirements for the first wall and blanket structural materials are obviously not limited to low activation. The first wall/blanket structure must operate for acceptable lifetimes in the severe radiation, thermal and stress environment as it exists in the core of a fusion reactor. Structural materials allowing for longer lifetimes will have a beneficial impact not only on the economics, but also on the safety and environmental aspects of fusion power. If vanadium-based alloys are considered to be most desirable materials from the point of view of long-term low activation, then an important question is: can these alloys fulfil all further requirements. It is difficult at the present time to establish whether vanadium alloys have the properties required for post-NET/ITER fusion reactors, because there are no designs that go beyond the conceptual stage. Also, the data base for vanadium alloys is limited. It is, however, possible to compare properties of existing vanadium alloys with those of other structural material candidates, as done in this paper. Some of the data used for this purpose were taken from the Blanket Comparison and Selection Study, carried out in the USA about half a decade ago [10].

### Structural material candidates

Three classes of alloys are currently considered as leading candidates for the first wall/blanket structure of a commercial fusion reactor: austenitic stainless steels, martensitic/ferritic steels and vanadium-base alloys.

Austenitic steels have been used extensively in fission reactor applications. For this reason, the austenitic steels are generally considered as a reference to which other alloys are compared. The primary candidate austenitic steel is AISI Type 316, which contains the elements Ni (10-14%) and Mo (2-3%). These elements are highly undesirable from the activation point of view. The low-activation counterpart to 316 is a manganese-stabilised steel with very low Ni and Mo content to reduce activation. Disadvantages of this type of steel are corrosion and safety because of volatility of Mn. Other properties are assumed to be similar to those of 316.

The high chromium martensitic/ferritic steels are more resistant to radiation swelling than austenitic steels. MANET, a one-time candidate first wall material for NET, contains almost 1% Mo and Ni each, as well as about 0.2% Nb. The US ferritic steel HT-9, with Nb replaced by V, would be slightly less activating than MANET, but does contain Mo and Ni. Less activating ferritic steels are obtained by elimination of Ni and substitution of Mo by W. These Fe-Cr-V-W steels are being studied in the US and Japan.

The class of materials having the potential of being the lowest long-term activation materials are the vanadium alloys. The V-15Cr-5Ti alloy, originally developed for fast breeder reactor applications [11] and with a larger data base than other vanadium alloys, is usually regarded as reference vanadium-base alloy.

### Relevant properties

In the remainder of this chapter a number of properties of vanadium-base alloys will be described, which in addition to induced radioactivity are of importance for fusion reactor application. Where possible comparison will be made with properties of austenitic and martensitic/ferritic steels. The data shown should be considered as indicative for each group of candidate structural materials.

### Physical properties

The physical properties of vanadium and some of its alloys are reasonably well known. Some relevant properties for V-15Cr-5Ti are listed in Table 1 [10]. Since most physical properties are not sensitive to moderate compositional variations, data for V-15Cr-5Ti can be regarded as being representative for similar types of vanadium. Also listed in Table 1 are property data for austenitic steel (Type 316) and ferritic steel (HT-9). An important advantage of vanadium alloys compared to the steels, is the potential for higher temperature operation, which allows for higher thermal efficiency. Several temperature limiting properties, such as radiation induced swelling, tensile strength and phase stability, correlate with the absolute melting temperature of the structural material. The melting temperature for vanadium alloys is more than 450 °C higher than for the steels, which effectively means an operational temperature advantage of 150-200 °C.

Because of the higher thermal conductivity and melting temperature and lower thermal expansion coefficients compared to the steels, vanadium alloys can accommodate higher thermal heat fluxes than the steels. The low density of vanadium alloys is another advantage. Lower weight to strength ratios are especially attractive for large structures.

### Mechanical properties

The strength of alloys, including vanadium-base alloys, depends on the type and amount of alloying additions, as well as on thermo-mechanical treatment. At temperatures up to about 600 °C ultimate and yield strength levels can be attained that are comparable to, or even higher than those of austenitic steel 316. This applies for example to the alloy V-15Cr-5Ti. An advantage of vanadium-alloys is that, while steels become weaker at temperatures above 600 °C, the vanadium alloys keep their strength up to at least 700 °C [12]. Ductility of vanadium alloys also depends on composition and material condition. Ductility generally decreases with increasing alloying additions. Also the impurities O, N, C and H, for which vanadium has a large affinity, decrease the ductility. Total elongations of 25-30% have been determined for the alloy V-15Cr-5Ti in the temperature range 25-700 °C [12]. The tensile properties and other mechanical properties of vanadium alloys have been reviewed extensively in [5].

The first wall and blanket structure will be subject to a combination of stresses and high temperatures over extended periods of time, which will make creep properties of vital concern. The available data indicate that the creep properties of the investigated vanadium alloys are superior to those of steels, e.g. [13]. Creep behaviour of vanadium alloys also depends on the composition. For example, the creep strength increases with Ti content up to about 2-3% Ti and then decreases with further increase in Ti content [14].

The importance of fatigue properties is questionable, because the eventual aim for fusion reactors is steady-state operation. At any rate, the few available results suggest that also the fatigue properties, at least of the alloy V-15Cr-5Ti, are better than those of austenitic steel 316 [15].

A concern which is specific for metals with the bcc lattice structure to which vanadium belongs, is the ductile-brittle transition temperature (DBTT), which is characterized by a change in fracture mechanism from shear (ductile behaviour) above this temperature to cleavage (brittle behaviour) at lower temperatures. The DBTT can range from -200 °C to above room temperature, again depending on alloy composition and impurity content. Particularly the impurities O, N, C and H can raise the DBTT considerably. An oxygen content of ~ 2000 ppm is enough to raise the DBTT from ~ 150 °C to above room temperature [16]. The effect of hydrogen is even more pronounced. Recent results of Charpy impact loading experiments on a number of vanadium alloys, showed that for alloys containing Cr, Ti and Si additions, the DBTT is lowest when the total alloying addition is in the range 3-9% [17].

## Radiation damage

The primary causes of neutron induced radiation damage are atomic displacements and transmutations, especially those producing helium. Combined or separately they may lead to dimensional instability, resulting from swelling, and to effects on mechanical properties such as embrittlement and changes in strength. The current data base for vanadium alloys is more limited than for austenitic and ferritic steels. However, existing data are sufficient to indicate trends and severity of dominant effects. Since a high flux 14 MeV neutron source is not available, the radiation data for all materials of interest are based predominantly on fission neutron and ion irradiations.

Swelling, which results mainly from cavity formation, has been observed in most neutron and ion irradiated metallic systems, including vanadium alloys. Cavity formation and swelling are dependent on dose and temperature, but also on the composition of the alloys. Experiments have shown that Ti concentrations of only a few percent strongly suppress swelling [18,19]. Recently reported swelling data for a variety of vanadium alloys irradiated to fluence levels of 84 dpa (displacements per atom) indicated that addition of Cr has just the opposite effect by increasing swelling substantially [20]. There are no neutron irradiation data for higher displacement levels, while a level of 100-150 dpa may already be reached after one year of operation in a power producing fusion reactor. Helium apparently slightly enhances swelling of some neutron irradiated V-alloys [21]. Comparison of experimental data indicates in general lower swelling for vanadium alloys and ferritic steels than for austenitic steels, particularly at high dpa level [3,22].

Increases in ultimate tensile strength and yield strength following neutron or ion irradiation have been reported for several vanadium alloys, the extent again being dependent on irradiation temperature, dose level and composition of the alloys. Most recent data for various vanadium alloys, after neutron irradiation to 90 dpa [23], show the yield strength to reach a maximum at about 50 dpa. Increase in strength due to irradiation is not considered to be a problem. On the other hand, loss of ductility caused by radiation hardening, helium embrittlement or increase in the DBTT, is a major concern. The available data indicate that although substantial reductions in ductility are observed in V-Cr-Ti, V-Cr and V-Ti alloys after neutron irradiation, significant residual ductility is retained [24,25]. As far as the combined effect of helium and displacement damage is concerned, reported results are not consistent. Pre-injection with helium led to significant reduction following neutron irradiation of V-20Ti at about 700 °C, not at lower temperatures [26]. These results and metallographic examination of the specimens suggested that the ductility loss was caused by helium embrittlement. On the other hand, similar experiments on the alloys V-15Cr-5Ti [27], V-5Ti and V-3Ti-1-Si [28] did not indicate any effect of helium on the ductility. Whether or not vanadium alloys are as sensitive to helium embrittlement as for example austenitic steel, is therefore still open to question. Based on helium embrittlement, which is com-



monly observed in metals at temperatures  $\geq 0.5 T_m$  ( $T_m$  is the absolute melting temperature), a maximum design temperature of around 700 °C seems appropriate for vanadium alloys, compared to 550 °C for austenitic and ferritic steels.

Neutron irradiation increases the DBTT. For vanadium alloys with a large fraction of alloying additions the increase can be substantial. The DBTT of neutron irradiated vanadium alloys containing Cr, Ti and Si, remains lowest ( $< -50^\circ\text{C}$ ) when the combined alloying addition is in the range 3-9% [17].

### Corrosion and compatibility

Corrosion and compatibility issues are also a major consideration in selecting first wall and blanket materials. Aspects of importance are compatibility with coolants (e.g. liquid lithium, helium), interactions with hydrogen isotopes and reactivity with air.

Problems concerning compatibility with liquid lithium arise from dissolution of wall material and from interaction with non-metallic impurities leading to deterioration of mechanical properties. Absorption of oxygen from liquid sodium and the resulting embrittlement was one of the main reasons for rejection of vanadium alloys for application in sodium cooled fission reactors (LMFBR). However, because the affinity of oxygen for lithium is even larger than it is for vanadium, vanadium alloys actually release oxygen when in contact with liquid lithium, leading to lithium penetration and weakening of the alloys [29]. This type of corrosion can be avoided by keeping the oxygen concentration of the alloys at a low level ( $< 1000$  ppm). Furthermore, the concentration of C and N impurities in liquid lithium, which in contrast to O will be absorbed by vanadium alloys, must be maintained at a low level to avoid embrittlement [23].

The rate of wall thinning due to dissolution puts an upper limit to the operating temperature. Based on only few data, this temperature limit for vanadium alloys, with lithium as coolant, is estimated to be at least 750 °C, compared to  $\sim 450$  °C for austenitic steels and  $\sim 550$  °C for ferritic steels [10].

Helium, being an inert gas, is not corrosive. The "corrosive" character of helium as coolant is due to, again, non-metallic impurities reacting with vanadium at high temperatures. In order to avoid contamination of vanadium alloys during prolonged exposure at 600 °C or above, residual impurities will have to be maintained at concentration levels which are currently impossible to maintain in helium circuits ( $< 1$  ppb) [30]. Helium as coolant in combination with a vanadium first wall/blanket is therefore not considered feasible.

Concerns related to compatibility of hydrogen isotopes with vanadium alloys are hydrogen embrittlement, excessive tritium inventory and tritium permeation. Hydrogen permeation rates for vanadium are high compared to steel [31]. Containment of tritium, being a radioactive hydrogen isotope, is a necessity.

Since vanadium will react with elements such as oxygen and nitrogen in air at elevated temperatures, a vanadium alloy structure must be protected from air during operation. Possible effects of accidental exposure to air must also be considered.

### Fabricability

Fabrication constraints exist for vanadium and its alloys due to their low tolerance for oxygen and nitrogen impurities. Fabrication can be achieved by current techniques for refractory metals, provided that all melting and annealing is done in vacuum. Vanadium and most of its alloys have excellent cold working properties and can be forged, rolled and swaged at room temperature. High speed steel and carbide tools may be used for machining. Vanadium can be welded using electron beam welding and by tungsten inert gas (TIG) welding [32]. Different vanadium alloys V-20Ti/V-15Cr and even V-15Cr/Type 304 stainless steel were welded successfully [3].

### Availability, production capacity and cost

Since relatively large quantities of material will be required for large-scale fusion application, resources and availability of vanadium need to be considered as well. Vanadium is a moderately abundant material in comparison with refractory metals as W, Nb and Mo, as can be seen in Table 2. Crude petroleum, uranium ores and phosphate deposits usually contain vanadium in recoverable quantities. Vanadium is a common by-product from processing these materials [33].

Production capacity for fusion application does not appear to be a problem. Present capacity of 35,000 kg per year can be doubled with a few weeks notice and increased 20-fold in about 2 years, according to one of the main producers of vanadium [33].

Although the cost of vanadium, estimated at about \$ 500 per kg at present, is significantly higher than of austenitic and ferritic steels, this is not considered to be a major factor in the first wall material choice. Compensation for the higher material cost will come from a longer wall life and a higher operation temperature.

## SUMMARY AND CONCLUSION

Vanadium alloys currently appear to show considerable promise as eventual replacement of ferrous alloys for the first wall/blanket structure of fusion reactors. They offer an excellent combination of low long-term activation, high temperature capability, corrosion resistance in liquid lithium and resistance to radiation damage. Particular concerns regarding use of vanadium alloys relate to their reactivity with non-metallic elements, tritium permeation and tritium inventory. Availability, production capacity, fabricability and cost are not expected to present major obstacles.

Vanadium alloys are not likely to be used for near-term fusion reactors because the data base is too limited. A period as long as 20 years may be needed to optimize these alloys for commercial fusion application.

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Table 1. Selected physical properties of three reference alloys

	316SS	HT-9	VCrTi
Melting temperature (°C)	1400	1420	1890
Density (g/cm <sup>3</sup> )	8.0	7.8	6.1
Linear thermal expansion (10 <sup>-6</sup> /K)			
400 °C	17.6	11.8	10.2
500 °C	18.0	12.3	10.3
600 °C	18.3	12.6	10.5
Thermal conductivity (W/m K)			
400 °C	19.5	26.8	26.8
500 °C	21.0	27.3	28.0
600 °C	22.5	27.7	29.5
Thermal stress factor *(MW/m <sup>2</sup> ,mm)			
500 °C	3.2	4.8	9.8

\* Thermal stress factor, which is a measure of the allowable heat load, is defined by:

$$\eta = (1 - \nu) k S_u / E\alpha,$$

where E = modulus of elasticity  
 $\alpha$  = coefficient of thermal expansion  
 $\nu$  = Poisson ratio  
k = thermal conductivity  
 $S_u$  = ultimate tensile strength.

Table 2. Relative abundance of vanadium in the earth's crust [33]

	Al	Fe	Mg	Ti	Mn	Zr	Cr
ppm	81,300	50,000	20,800	4,400	1,000	220	200
	V	Zn	Ni	Cu	W	Nb	Mo
ppm	150	130	80	70	69	24	15

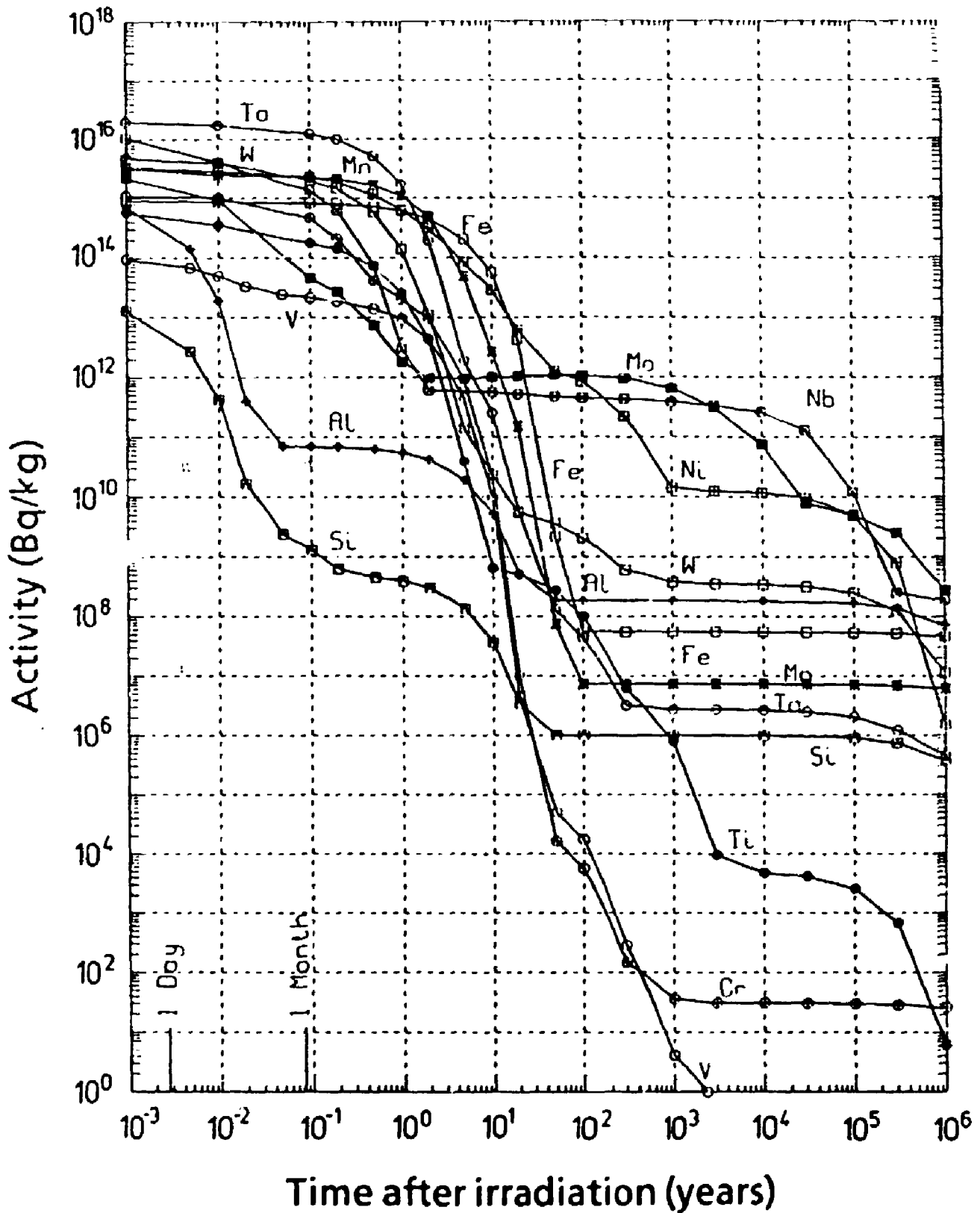


Figure 1. Radioactivities following shutdown of various elements [4]. They are assumed to be activated at the first wall of the conceptual Culham Demo reactor [8] after 2,5 years of operation at  $5 \text{ MW}\cdot\text{m}^{-2}$  neutron wall loading.

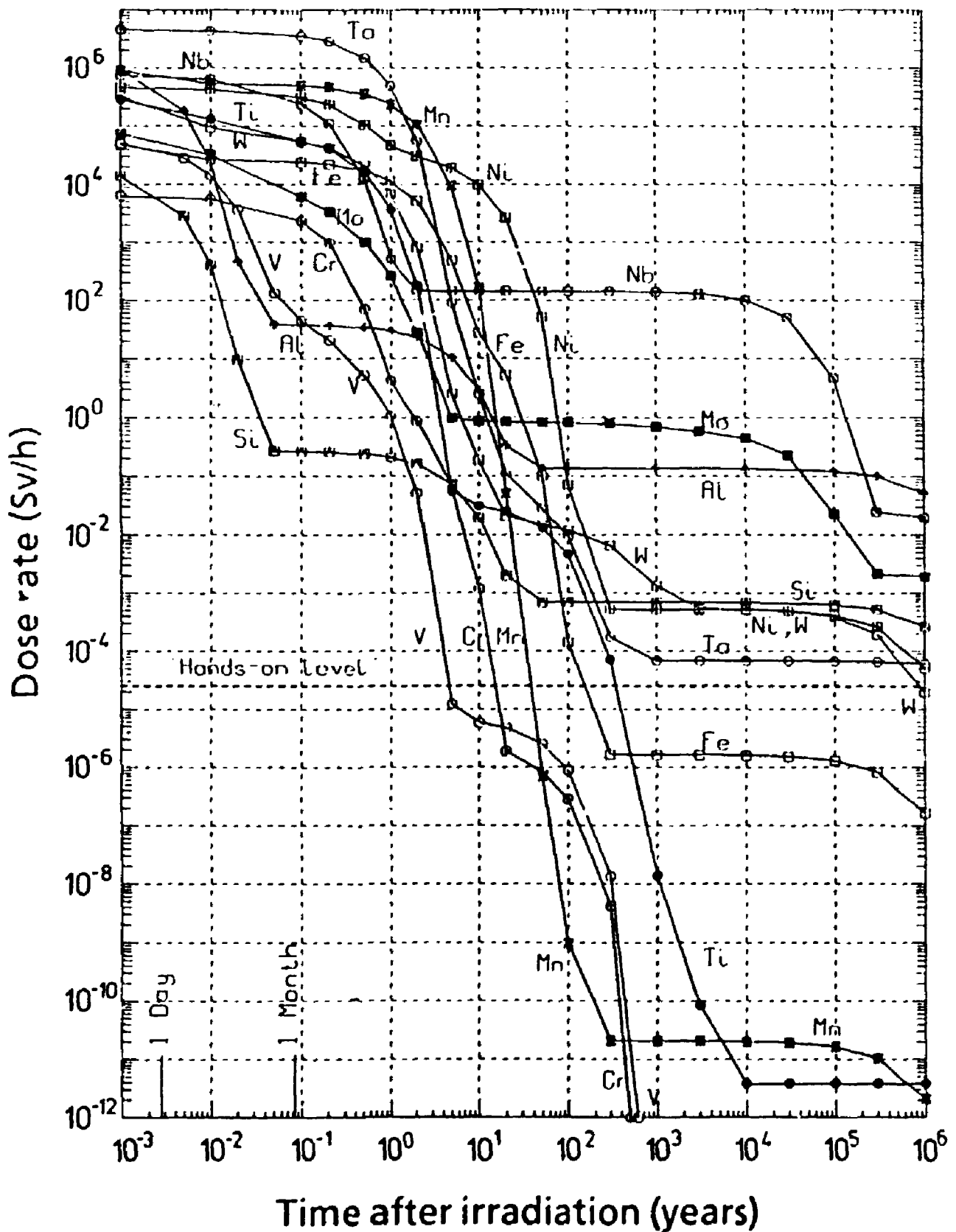


Figure 2. Dose rates derived from radioactivity levels of various elements indicated in Fig. 1 [4].