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ISOTOPICALLY TAILORED MATERIALS FOR FISSION AND FUSION REACTORS

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The use of isotopically enriched fuels in nuclear reactors is well known. In addition, because many structural components of a reactor are subjected to the neutron flux, the isotopic composition of these components can also affect the reactor performance. In many cases, the chemical compositions of these structural materials have been altered to either reduce their neutronic interactions or improve their radiation stability. Recently, several reactor specialists have suggested that the isotopic composition of some components could also be adjusted to improve their performance. The reactor characteristics that may be affected by such isotopic adjustments are: (1) neutron economy, (2) operating efficiency (life time and operating temperature), and (3) environmental impact (activation and transmutation products). We shall give some examples of isotopic modifications which illustrate the benefits of isotopic tailoring of materials in fission reactors and in proposed fusion reactors. Let us look at two fission reactor applications, one for thermal reactors and the other for fast neutron, breeder reactors.

Thermal Fusion Reactors

Zircaloy (98% zirconium, ~2% tin) is presently used as the fuel cladding in the heavy-water-moderated power reactors, known as CANDU reactors, which are manufactured in Canada. Neutron absorption by this cladding is the dominant neutron loss mechanism. Of the neutron absorption in the Zircaloy, nearly 60% occurs in the isotope zirconium-91, and 10% occurs in the impurity isotope hafnium-177. Critoph[1] has suggested that optimizing the zirconium isotopes 90 and 94 in the Zircaloy

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cladding could improve the neutron economy, resulting in a reduction in the fuel loading of approximately 6% and increasing the conversion ratio in a uranium/thorium breeder by 5-6%. He has calculated the allowable cost for isotopically tailored Zircaloy containing less than 5% zirconium-91 could be as high as $1,000/kg in a non-breeder reactor (nearly 25 times the present cost) and up to $3,000/kg in a U/Th breeder.

Fast Fission Reactors

In the early fast breeder reactors, oxide fuels are being utilized; however, advanced LMFBR fuels will be U/Pu carbides or nitrides because of their superior breeding and higher power performance capabilities. Nitride fuels made with natural nitrogen (99.6% ^{14}N, 0.4% ^{15}N) have a rather high neutron absorption cross section because of the nitrogen-14. Nitrogen-15 has a negligible neutron absorption cross section so the neutron absorption in the fuel can be nearly eliminated by using fuel from which the nitrogen-14 has been removed. In addition, the deletion of nitrogen-14 eliminates the formation of carbon-14 which is produced by the (n, p) reaction with nitrogen-14. A preliminary economic analysis by Tennery of ORNL[2] indicates that the allowable cost of 99.9% nitrogen-15 could be from $438 to $2500/kg depending upon the fuel parameters. It is estimated that 10^4 kg/year up to 1993 and 3×10^5 kg/year thereafter of enriched nitrogen-15 may be required. The present cost of nitrogen-15 is $68,000/kg. For this case, an engineering study was made for an isotopic enrichment plant for the production of 300 metric tons per year of nitrogen-15. The plant utilized the isotopic separation factor for the process involving chemical exchange between gaseous NO and counter-currently flowing aqueous nitric acid in a packed column (Table I). This study indicates that nitrogen-15 could be produced at $167/kg, which is well below the minimum allowable cost.

Fusion Reactors

Radiation damage has been called the second most serious obstacle to the commercialization of fusion power. The first obstacle, of course, is still the problem of containing the plasma. The first wall, i.e., the interior surface of the vacuum vessel that contains the plasma, is the part of a fusion reactor that suffers the greatest damage. In a D-T-fueled Tokamak, the first wall is subject to a 14 MeV neutron flux of the order of 10^{15} n/cm^2·sec, as well as

Table I

CHEMICAL EXCHANGE NO-HNO_3

Capital Costs (By Use of "Lang Factors" and the Addition of a 50% Contingency)	\$115.(10^6)
Production Cost	
(1) Direct Cost	
Operations (555 MY/YR)(\$23,000/MY)	12.8(10^6)
Utilities (6.6 x 10^8 KWHR/YR)(\$0.02/KWHR)	13.2(10^6)
Feed Cost	~0
(2) Fixed Charges	
Depreciation	11.5(10^6)
Taxes	2.3(10^6)
Insurance	1.2(10^6)
(3) Interest	9.2(10^6)
Total Production Cost	50.2(10^6)
Unit Cost \$50.2(10^6)/3.(10^5) kg =	167.33/kg

high energy photons, α's, protons, deuterons, and tritons. We shall discuss only neutron effects here.

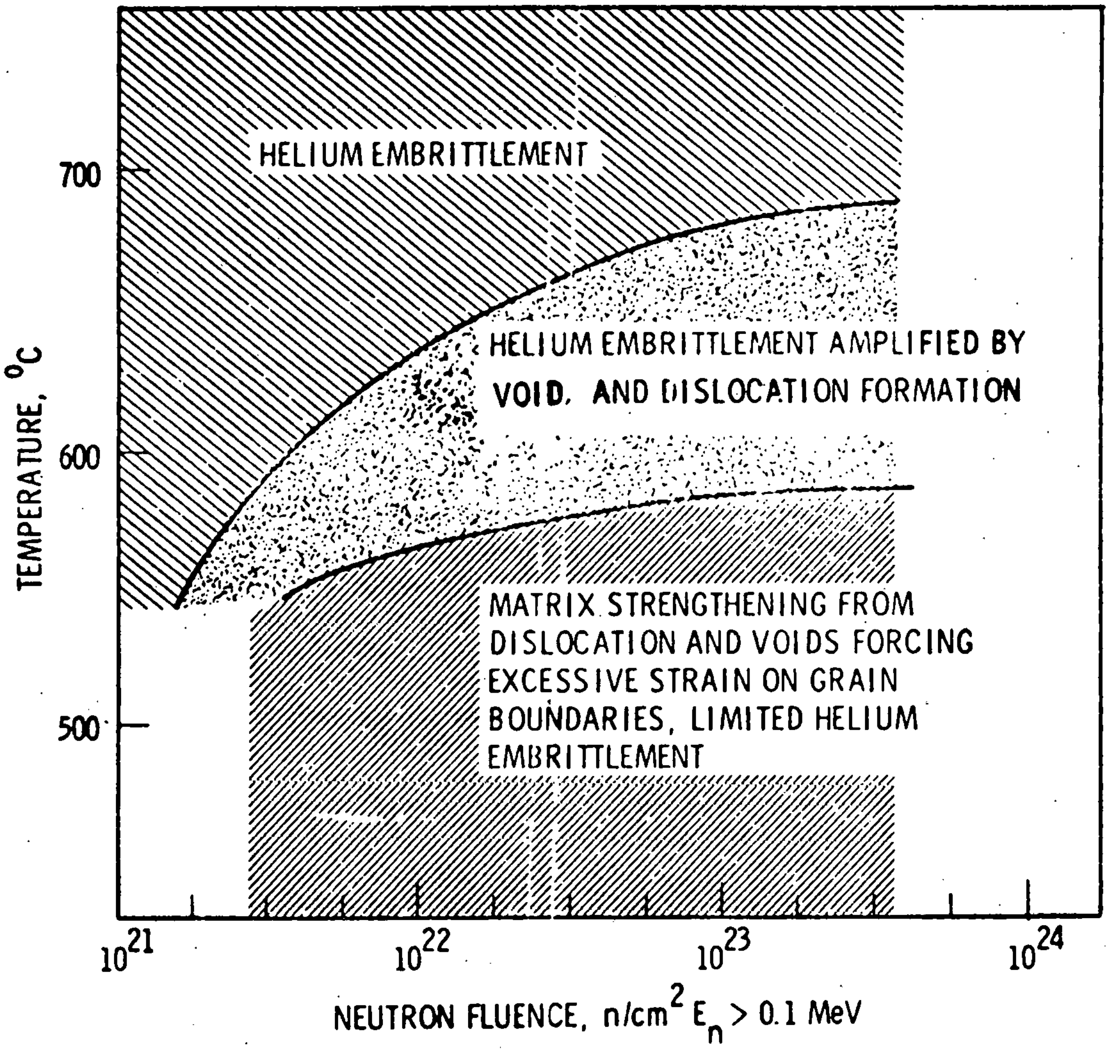
There are three principal aspects of the neutron radiation damage: (1) atomic displacements, caused by collisions between neutrons and lattice atoms; (2) helium, created by (n, α) reactions; and (3) transmutations, which alter the chemical composition of the structural material and/or make the structure radioactive. Atomic displacements (as many as a few hundred displacements per year for every atom in the first wall) create lattice imperfections which make the metal swell and reduce its elasticity. These effects are most important below about 600°C (in stainless steel) because they tend to be removed by annealing at higher temperatures. Reactions which create alpha particles cause an accumulation of helium in the metal. The helium cannot diffuse through the metal; consequently, it remains in the lattice, causing a high internal pressure which makes the metal swell. This effect is most pronounced at temperatures above 500-600°C (Figure 1).

At the probable operating temperature of the first wall, ~450-500°C, the displacement and helium ingrowth effects operate synergistically to degrade the metal wall.[3] Because (n, α) reactions are sensitive to nuclear properties, it has been proposed[4] that the first wall material could be isotopically tailored to reduce the (n, α) cross section and thereby reduce the amount of helium trapped in the metal. This would permit an increased operating temperature which would permit some of the displacement damage to be removed by annealing. The increased temperature would also increase the thermal efficiency of the reactor.

The most likely first wall material in early reactors is 316 stainless steel. Iron comprises 65% of this metal, and 5.9% of the iron is iron-54, which has a relatively high cross section for the (n, α) reaction at 14 MeV. It has been estimated that reducing the iron-54 concentration to less than 0.1% of the iron could eliminate half the (n, α) reactions. This could permit, depending upon other factors such as the frequency of atomic displacements, a doubling of the useful life of the first wall. The quantity of depleted iron required would be of the order of tons per year,and the potential cost savings are of the order of tens of millions of dollars per year.

Even if the first wall life is doubled, it will need to be replaced every few years. This is a major task, involving thousands of kilograms of material.

Figure 1


HELIUM EMBRITTLEMENT
HELIUM EMBRITTLEMENT AMPLIFIED BY VOID, AND DISLOCATION FORMATION
MATRIX STRENGTHENING FROM DISLOCATION AND VOIDS FORCING EXCESSIVE STRAIN ON GRAIN BOUNDARIES, LIMITED HELIUM EMBRITTLEMENT
700
600
500
TEMPERATURE, °C
10^{21}
10^{22}
10^{23}
10^{24}
NEUTRON FLUENCE, n/cm^2 $E_n > 0.1$ MeV

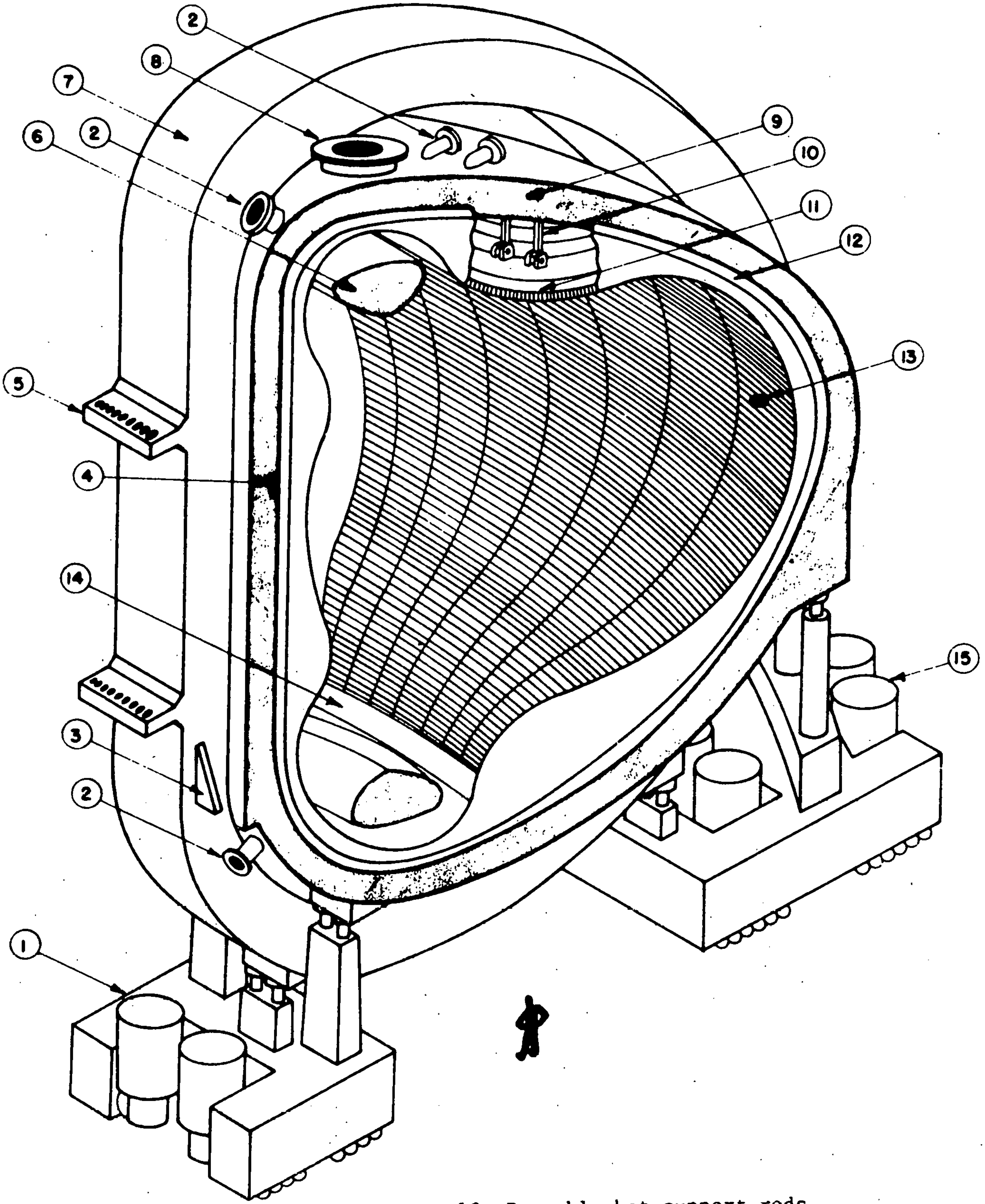
A section of a fusion reactor (UWMAK 1)[3] is shown in Figure 2. The job is complicated by the fact that the entire structure is now radioactive. This radioactivity builds up quickly, reaching a steady state in a matter of hours, but it decays more slowly, with half lives from seconds to years. Figures 3 and 4 indicate the predicted radioactivity as a function of time in the stainless steel structure of a fusion reactor.

The table of activation products in stainless steel (Table II) indicates that isotopic tailoring can be used to reduce the amount of induced radioactivity in a fusion reactor. Isotopes which have half lives less than one day are not included here because they do not contribute significantly to the handling problem. Activation products from chromium, iron, and molybdenum can be eliminated by removing all their respective isotopes except chromium-53, chromium-54, iron-57, and molybdenum-97. No nickel isotope is free of activation products, but by using only nickel-64, all the gamma emitting products of nickel can be eliminated. This reduces the hazard of handling the radioactive material because only light shielding is required for beta particles. The remaining gamma emitting product, manganese-54, can be eliminated by removing all the manganese in the steel and making chemical changes to compensate for the missing manganese.[5]

This analysis oversimplifies the problem because it considers only the elimination of radioactive products. A more complete analysis would include the half-life associated with each activation product, the relative cross section of each of the many activation reactions and their isotopic abundances. Figure 5 indicates the relative contribution of various isotopes to the radioactivity in the first wall after shutdown.

Of even more practical importance is the relative biological hazard of the radioactive products. This is shown in Figure 6. Note that iron-55, for example, contributes less to the total hazard than does the less active manganese-54. For example, if only chromium-50, chromium-52, manganese-55, iron-54, nickel-58, and the four lightest molybdenum isotopes were removed from 316 stainless steel, half the radioactivity would be eliminated and the gamma intensity would be reduced more than 90%. Almost all the remaining activity comes from iron-55 which decays by electron capture with no gamma ray. Isotopes with half lives greater than 100 years would be entirely eliminated. This would be considerably easier and probably cheaper than removing all but one each of the iron, nickel, and molybdenum isotopes.

Figure 2



1- Front motorised caterpillar
2- Lithium inlet or outlet
3- Front magnet dewar support
4- Front blanket support bar
5- Magnet support shear beam
6- Vacuum port shield
7- Toroidal magnet in its dewar
8- Vacuum connection
9- Shield
10- Rear blanket support rods
11- Heat removal cells
12- Blanket seal flange
13- Neutral beam injection port
14- Particle collection plate
15- Rear motorised caterpilar

Figure 3

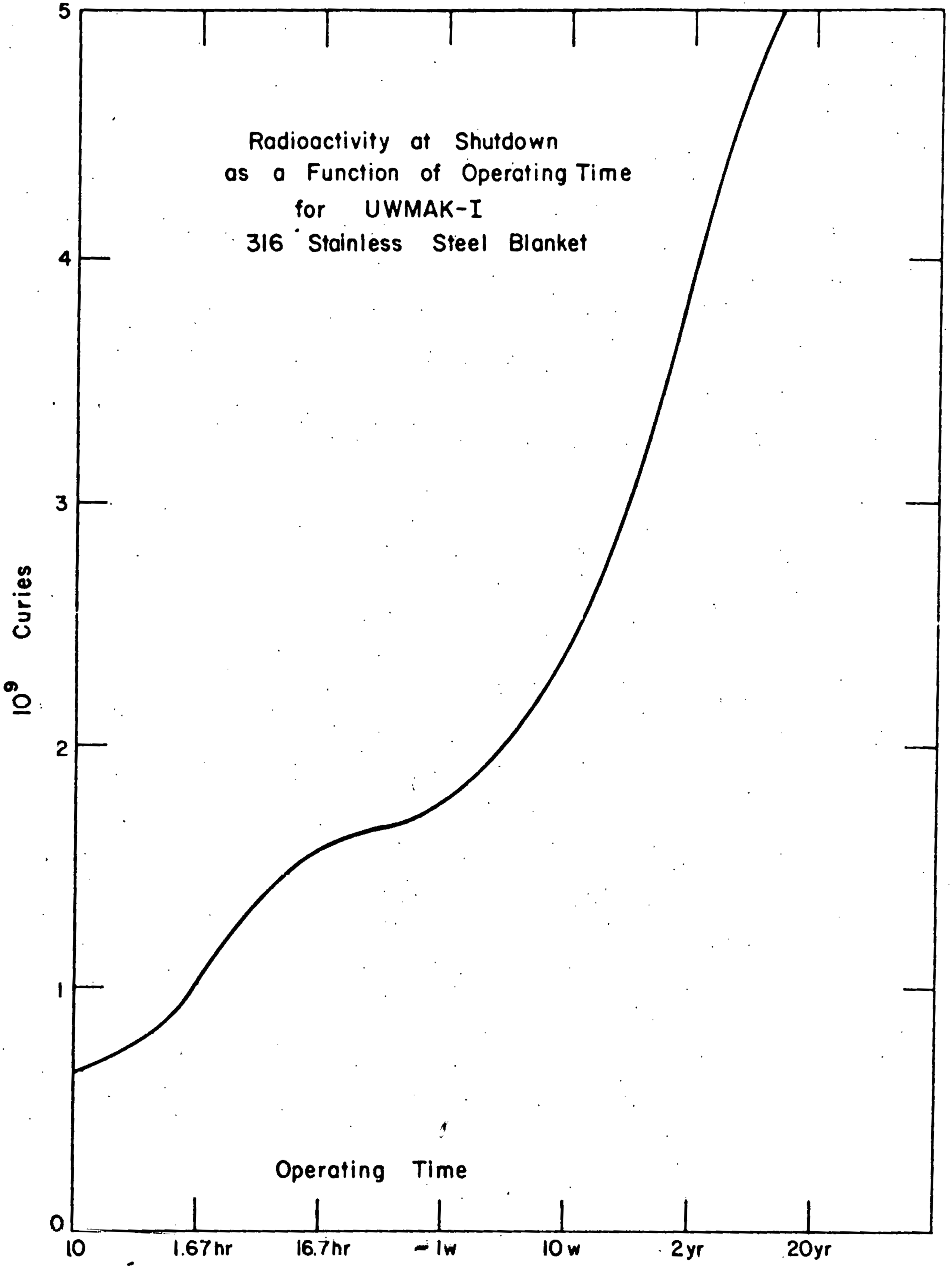

Radioactivity at Shutdown
as a Function of Operating Time
for UWMAK-I
316 Stainless Steel Blanket
10^9 Curies
5
4
3
2
1
0
Operating Time
10
1.67 hr
16.7 hr
~ 1w
10 w
2 yr
20 yr

Figure 4

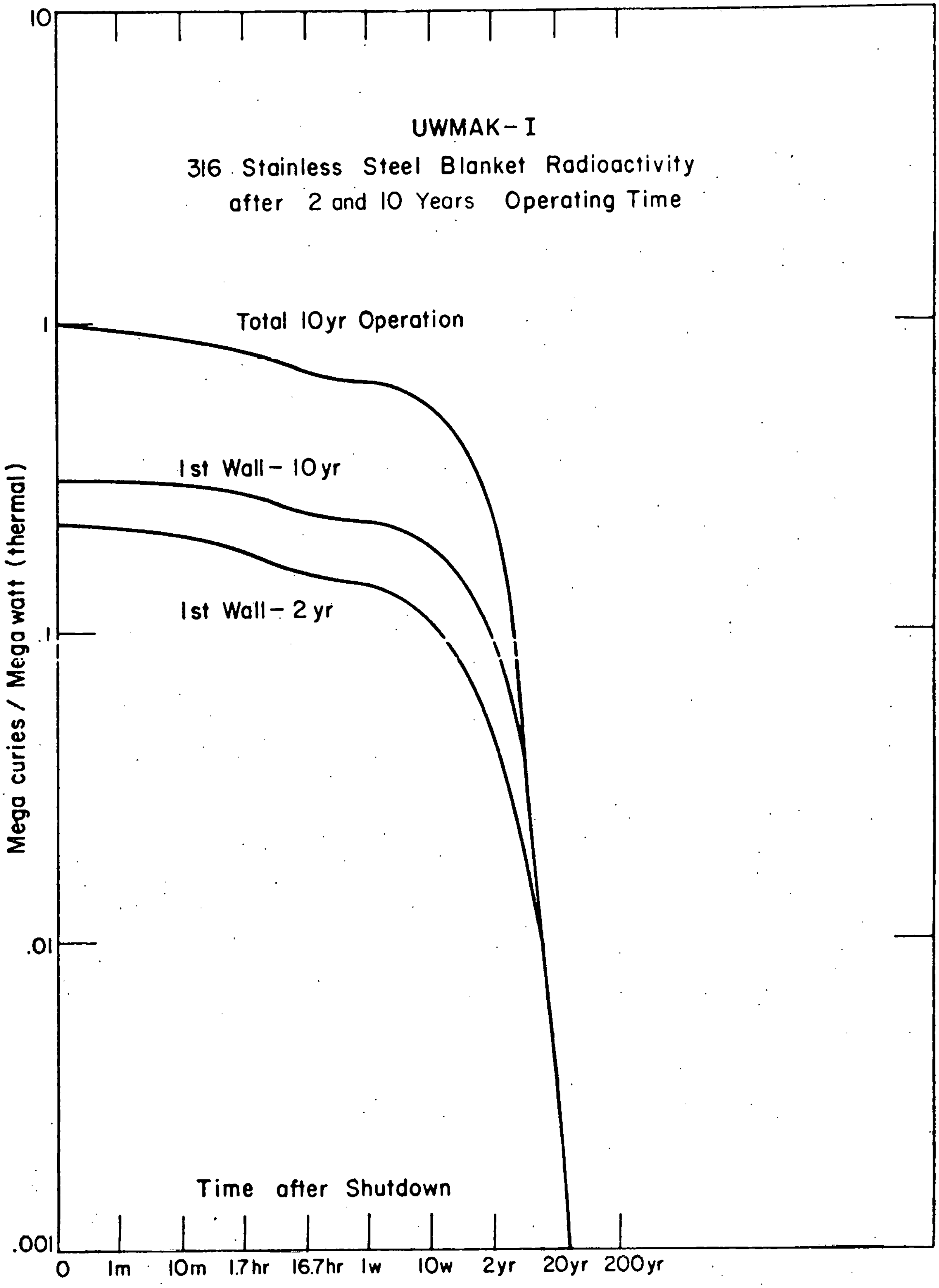
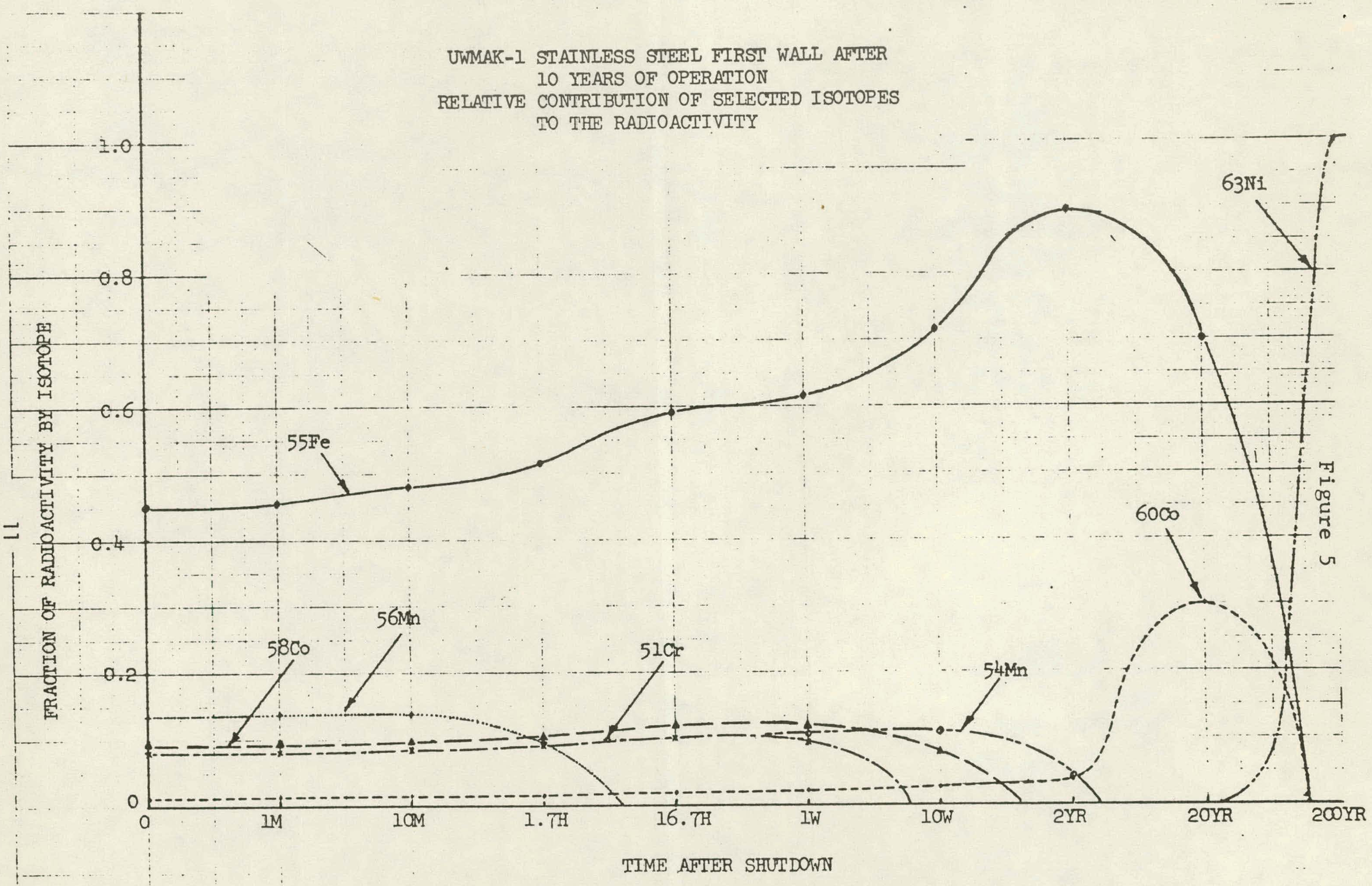

UWMAK-I
316 Stainless Steel Blanket Radioactivity
after 2 and 10 Years Operating Time
Total 10yr Operation
1st Wall - 10 yr
1st Wall - 2 yr
Mega curies / Megawatt (thermal)
Time after Shutdown
10
1
.1
.01
.001
0
1m
10m
1.7hr
16.7hr
1w
10w
2yr
20yr
200yr

Table II

PRINCIPAL RADIOACTIVE ACTIVATION PRODUCTS IN STAINLESS STEEL*

% of 316 SS	% Natural Abundance	Source	Product
18% Cr			
	4.4	$^{50}\mathrm{Cr}$	$\underline{^{51}\mathrm{Cr}}$, $^{49}\mathrm{V}$
	83.5	$^{52}\mathrm{Cr}$	$\underline{^{51}\mathrm{Cr}}$
	9.5	$^{53}\mathrm{Cr}$	
	2.6	$^{54}\mathrm{Cr}$	
2% Mn			
	100	$^{55}\mathrm{Mn}$	$\underline{^{54}\mathrm{Mn}}$
65% Fe			
	5.9	$^{54}\mathrm{Fe}$	$^{55}\mathrm{Fe}$ $\underline{^{54}\mathrm{Mn}}$ $\underline{^{51}\mathrm{Cr}}$
	91.7	$^{56}\mathrm{Fe}$	$\underline{^{55}\mathrm{Fe}}$
	2.2	$^{57}\mathrm{Fe}$	
	0.3	$^{58}\mathrm{Fe}$	$\underline{^{59}\mathrm{Fe}}$
11% Ni			
	67.8	$^{58}\mathrm{Ni}$	$^{59}\mathrm{Ni}$ $\underline{^{58}\mathrm{Co}}$ $\underline{^{57}\mathrm{Ni}}$ $\underline{^{57}\mathrm{Co}}$ $^{55}\mathrm{Fe}$
	26.2	$^{60}\mathrm{Ni}$	$\underline{^{60}\mathrm{Co}}$ $^{59}\mathrm{Ni}$
	1.2	$^{61}\mathrm{Ni}$	$\underline{^{60}\mathrm{Co}}$
	3.7	$^{62}\mathrm{Ni}$	$^{63}\mathrm{Ni}$ $\underline{^{59}\mathrm{Fe}}$
	1.2	$^{64}\mathrm{Ni}$	$^{63}\mathrm{Ni}$
1.5% Mo			
	15.8	$^{92}\mathrm{Mo}$	$\underline{^{93}\mathrm{Mo}}$ $\underline{^{92}\mathrm{Nb}^{m}}$ $\underline{^{91}\mathrm{Nb}}$ $\underline{^{89}\mathrm{zr}}$
	9.1	$^{94}\mathrm{Mo}$	$\underline{^{94}\mathrm{Nb}}$ $\underline{^{93}\mathrm{Mo}}$
	15.7	$^{95}\mathrm{Mo}$	$\underline{^{95}\mathrm{Nb}}$ $\underline{^{94}\mathrm{Nb}}$
	16.5	$^{96}\mathrm{Mo}$	$\underline{^{95}\mathrm{Nb}}$
	9.4	$^{97}\mathrm{Mo}$	
	23.8	$^{98}\mathrm{Mo}$	$\underline{^{99}\mathrm{Mo}}$ $\underline{^{95}\mathrm{Zr}}$
	9.6	$^{100}\mathrm{Mo}$	$\underline{^{99}\mathrm{Mo}}$

*Only isotopes with half lives greater than one day are included. Underlined isotopes are gamma emitters; all listed isotopes decay by beta emission or internal conversion.



UWMAK-1 STAINLESS STEEL FIRST WALL AFTER
10 YEARS OF OPERATION
RELATIVE CONTRIBUTION OF SELECTED ISOTOPES
TO THE RADIOACTIVITY

Figure 5

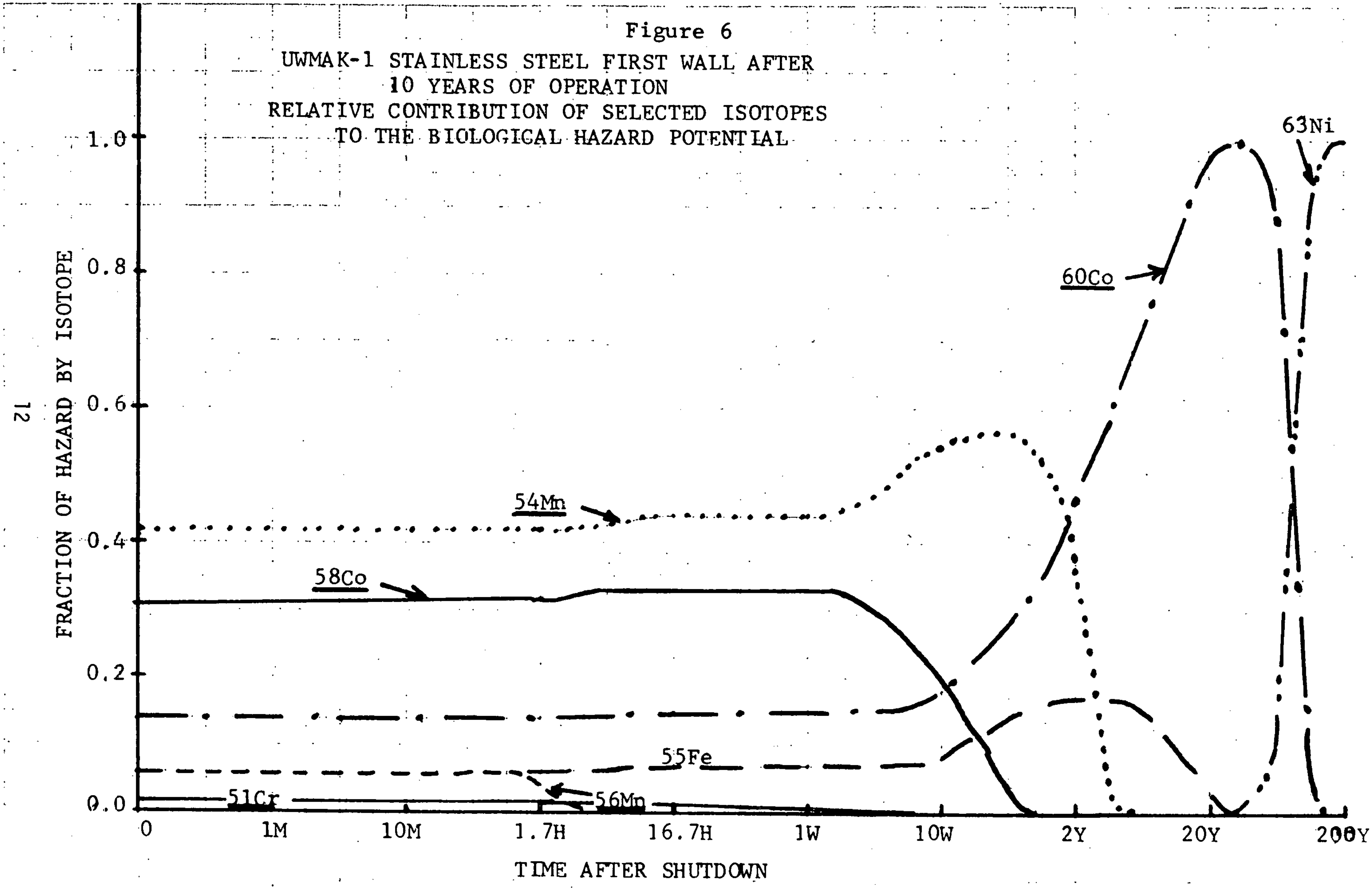


Figure 6

UWMAK-1 STAINLESS STEEL FIRST WALL AFTER 10 YEARS OF OPERATION
RELATIVE CONTRIBUTION OF SELECTED ISOTOPES TO THE BIOLOGICAL HAZARD POTENTIAL

In summary, there are many possible applications for the use of materials with altered isotopic compositions in both fission and fusion reactors. The potential benefits of these isotopically tailored components need to be more quantitatively assessed. Concurrently, the isotopic separations techniques for producing such isotopically altered material need to be explored and cost estimates made, similar to the cited cost study made for the production of nitrogen-15. Because of recently developed advanced isotopic separations techniques and the economies of large-scale production, the prospects for obtaining the required isotopically tailored materials at reasonable cost are encouraging.

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