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CRITERIA FOR THE SELECTION OF MATERIALS FOR WATER-COOLED REACTORS, WITH COMMENTS ON D_2O REACTORS

DL-51

by

W.B. LEWIS

Paper for presentation at the AIME Symposium
on Selection of Materials for Gas-cooled and Water-cooled Reactors
to be held in New York, October 31, 1962

Chalk River, Ontario

July 6, 1962

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ABSTRACT

When in combination intense radiation, high temperatures and high mechanical stresses act on different materials in contact, the familiar classification of materials into solids, liquids and gases is a handicap rather than a help. Water becomes a source of hydrogen that will pass into a metal and change its resistance to stress. Hydrogen diffuses rapidly through metals even at water temperatures. At the higher temperatures in nuclear fuel, oxygen and carbon diffuse rapidly. In some ceramic fuels the operating temperatures are so high that even heavy atoms will dwell less than a millisecond in any given lattice position. With so high a rate of the breaking of bonds, diffusion and interaction, the contribution of all substances present to the general ecology needs consideration. The stabilization of the mechanical properties of metals when bombarded by fast neutrons and exposed to wandering atoms, especially hydrogen, has to be studied. The behaviour of zirconium alloys and uranium dioxide in such environments has been extensively studied and useful design criteria have been set and explored.

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The requirements of durability under irradiation, corrosion resistance, strength at temperature and low neutron capture, leave only few materials that can be selected for duty in a water-cooled reactor core. The functional duties are few, namely those of fuel, coolant, thermal insulation, moderator and structure, in particular fuel sheathing, fuel support and coolant channeling.

Provided materials meet the performance criteria, the final choice will be a matter of economics. For all components that amount to many kilograms, it is now well-known that the capture of neutrons rather than the initial cost of the material usually determines the economic position, because for every neutron wasted a new fissile atom has to be supplied to the reactor core. To appreciate the force of this argument, note that the major cost of operating a large power reactor arises from the necessity to refuel. Moreover, refuelling can be postponed in most water-cooled reactors by the regeneration of fissile material in the core, and it takes just one neutron to produce one such new fissile atom.

Evaluation of the relative cost of two metals, for example stainless steel or a zirconium alloy, for any particular purpose is liable to be very complex. A crude approximation, however, may be obtained from the net fuelling cost (F) in \$/thermal megawatt day (= mill/kWh x 24 x efficiency) and the irradiation (I)of the material in question in n/kb (i.e. 10^{21} nvt or 10^{21} n/cm^2) in conjunction with the neutron capture cross-section, σ b/atom, and atomic weight, A. The irradiation cost per kg is then about 200 σFI/A or per litre, 200 $\rho\sigma$FI/A where ρ is the density in g/cm^3. (1) Values of these irradiation cost factors, 200 σ/A and 200 $\rho\sigma$/A, are given for some materials in Table I. (2)

For example, the cost of irradiating 1 kg of stainless steel for 2 n/kb in a reactor of 35% efficiency with a fuel cost of 1.5 mill/kWh is \$10.8 x (1.5 x 24 x 0.35) x 2 = \$272. In contrast the same irradiation of 1 kg of Zircaloy-2 costs \$14.5. For reactor components in a high flux such as 10^{14} n/cm^2/sec for 10 years the irradiation would be 30 n/kb and the irradiation costs therefore 15 times as high. Another example of interest is the economic assessment of fuel sheathing. Suppose stainless steel sheathing 0.5 mm (0.020 in.) thick is used with a mean heat flux of 100 W/cm^2 (317,000 Btu/hr/ft^2) in the same reactor. The mass of stainless steel per ekW of plant capacity is then

$$0.05 \text{ cm} \times \frac{1000}{35} \frac{\text{ekW/cm}^2}{\text{ekW/cm}^2} \times 7.92 \text{ g/cm}^3 = 11.3 \text{ g/ekW}$$

At 10^{14} n/cm^2/sec for 7000 hr in the year this costs

$\$272 \times \frac{2.52}{2} \times \frac{11.3}{1000} = \$3.87$/kW yr or 3870/7000 = 0.55 mill/kWh.

This is such a heavy penalty that designers are likely to reduce the thickness to 0.25 mm (0.010 in.) and the neutron flux to 5×10^{13} n/cm^2/sec, thereby reducing the irradiation cost by a factor of 4. Alternatively a zirconium alloy might be chosen. (4)

TABLE I

Material	Atomic Weight	Density (ρ) g/cm^3	σ(1) barn	Irradiation Cost Factor $/kg/FI 200 σ/A	$/litre/FI 200 $\rho\sigma$/A
Graphite	12	1.7	0.004	0.067	0.113
Sodium	23.0	0.97	0.50	4.35	4.22
Magnesium	24.3	1.74	0.06	0.49	0.86
Aluminum	27.0	2.7	0.23	1.70	4.6
Iron	55.8	7.86	2.6	9.3	73
Stainless Steel(2)	55.3	7.92	3.0	10.8	86
Nickel	58.7	8.9	4.6	15.7	140
Zirconium	91.2	6.50	0.23*	0.50	3.3
Zircaloy-2(3)	91.2	6.50	0.263*	0.577	3.75
Zr-2.5% Nb-0.5% Cu	"	"	"	"	"
Niobium	92.9	8.55	1.1	2.37	20.3
Tin	118.7	7.28	0.60	1.01	7.4
Lead	207.2	11.37	0.17	0.164	1.9
$\frac{1}{2}(H_2O)$	9	1.00	0.33	7.3	7.3
$\frac{1}{2}(D_2O)$	10	1.10	0.00060*	0.0120	0.0132
MgO	40.3	3.58	0.06	0.30	1.07
$\frac{1}{2}(Al_2O_3)$	51.0	3.7	0.23	0.90	3.3
SiO_2(fused)	60.1	2.2	0.13	0.43	0.95
ZrO_2	123.2	5.6	0.23	0.373	2.09

(1) Cross sections marked * are effective reactor values, others are standard σ(2200) values. See note 3 at end.

(2) Composition 68% Fe; 18% Cr; 10% Ni; 2% Mn; 0.8% Nb; 1% Si.

(3) Composition Zr; 1.5% Sn; 0.15% Fe; 0.1% Cr; 0.05% Ni.

It should perhaps be emphasized that the above evaluation uses a very crude approximation that serves only as a rough guide. The limited validity of the approximation I have discussed in Reference 1.

From the cost viewpoint it almost invariably works out that the best arrangement is one that calls for long irradiations. This in turn calls for long-term stability of the materials under the working conditions. The ductility of metals that allows the sharing or relief of excessive stress without rupture must be preserved in the presence of intense radiation and of the products of radiolysis and corrosion.

Long and satisfactory experience has been accumulated on the use of aluminum in water reactors in the most intense radiation, but its susceptibility to corrosion has proved a severe limitation. The temptation to use graphite to secure the lubrication of moving parts has to be rejected in aluminum systems. (5) Water of the high purity required for low radiolysis is a very poor lubricant. Metals in sliding contact in pure water will gall, and radiation increases the tendency. For wear resistance a hard surface of zirconium oxide where applicable is probably most free from difficulties (6) Many cobalt-containing alloys offer hard-wearing surfaces. (7) Cobalt, however, is one of the elements that may have to be avoided if the high residual radioactivity of Co-60 causes maintenance difficulties.

Mechanical moving parts, seals and sliding contacts, are kept out of the highest neutron flux regions or limited in load or motion where possible.

In the design of heavy water reactors there is considerable economic advantage in keeping the moderator cool. Three factors contribute to the advantage:

(1) the higher neutron yield from U-235 and Pu-239 in a colder neutron spectrum;

(2) the lower neutron migration area in the denser moderator;

(3) the availability of cool liquid of high heat capacity to limit hazards of hot coolant escape.

Many reactors using cool D_2O moderator have employed aluminum reactor vessels, among them the Canadian Nuclear Power Demonstration reactor, NPD. (8) The use of aluminum has been abandoned in the Canadian full-scale power reactor, CANDU. (9) Instead, zirconium alloys are used in the high neutron flux regions and austenitic stainless steel in low flux regions. There are many reasons for the change although ultimately the criterion is economic.

The calandria tubes through which the fuel channels pass are thin-walled tubes of 4" diameter. For the required strength they can be thinner in Zircaloy than in aluminum and thus reduce the neutron

wastage. The large power, 200 eMW, of the plant, makes it economic to provide an ample D_2O reflector surrounding the reactor core, and the neutron flux is sufficiently reduced at the boundary. Removing aluminum from the whole reactor system makes it possible to choose the incidental materials, such as lubricants and bearing surfaces, as well as additives to the D_2O to control pH and decomposition rate without the limitations set by aluminum corrosion.

In this review we are concerned with the physical phenomena that set the detailed specifications of materials in a reactor system. To keep the review short and objective I shall select a particular type or class of reactor to which CANDU belongs. Reactors of this class employ UO_2 as fuel, zirconium alloy as fuel cladding, pressurized water at or near its boiling point as coolant, zirconium alloy tubes carrying the coolant, joined first to 410 stainless steel extensions and then via carbon steel piping to Inconel or Monel heat exchanger tubes. The reactor vessel is of austenitic stainless steel carrying zirconium alloy tubes and containing cool heavy water moderator.

ZIRCONIUM ALLOYS

Zircaloy-2 was developed by the USAEC and Westinghouse for its corrosion resistance in high temperature water, (10) and more experience has been gained with this than with any other zirconium alloy. Interest is now growing in an alloy of zirconium containing 2.5% niobium + minor additions to control corrosion; one promising example contains 0.5% copper. This alloy is heat-treatable and has higher strength at 300°C than Zircaloy.(11)

Under irradiation annealed Zircaloy hardens, but heavily cold-worked Zircaloy becomes partially annealed. There is therefore advantage in specifying a limited amount of cold work in the initial metal. It has been shown that advantage can be taken of the increased initial strength, for although some modification of the properties takes place during irradiation, the strength is maintained. (12) Zirconium is an anisotropic metal and it has been found important to specify the texture, not only for the advantage of initial strength but also for strength when hydride has accumulated.

At one time it was thought advisable to set as the lifetime limit on Zircaloy components the time of reaching the hydrogen solubility limit at the operating temperature, namely about 100 ppm at 300°C. Experiments have shown, however, that ductility is well maintained at least up to 400 ppm, so for many applications the limit can be raised. (13)

Despite a great number of experiments it has not been possible to determine whether irradiation in water has a specific effect on hydrogen uptake. (14) Any effect is not large for Zircaloy-2, but the experiments suggest that at higher nickel content the hydrogen uptake is increased by radiation. (15) The corrosion and hydrogen uptake have been shown to depend on

(1) The nature of the oxide film on the surface,

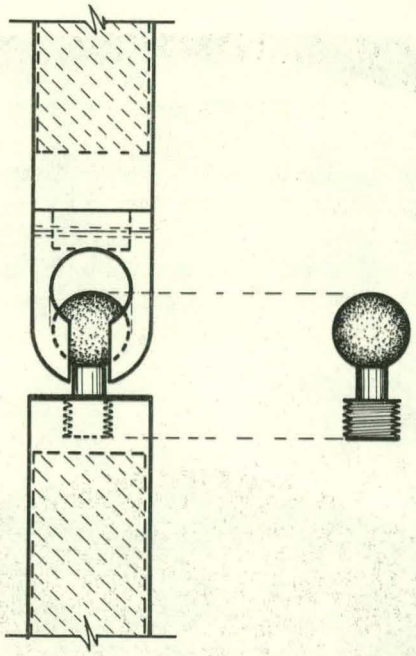
(a) both initially and after "transition," a stage after which the corrosion rate increases, and

(b) in the presence of fluoride even at a few ppm in the water. If the surface has only a very thin protective oxide layer, fluoride greatly increases the corrosion and hydrogen uptake. Only small effects have been established at low fluoride levels when the surface is protected by the type of oxide layer normally specified. (16) Note that polymerized fluorinated materials commonly used for gaskets may contribute unacceptable amounts of fluoride ion to hot water.

(2) The electro potential of the Zircaloy; if it is anodic (+ve) the hydrogen uptake is reduced.

There appears reason to expect a relaxation of the engineering specifications now used when a fuller understanding of the surface processes has been established.

Two examples may serve to illustrate weaknesses in practical designs. A very sound mechanical design shown in Fig. 1 was adopted for experimental work as a means of coupling individual fuel rods to form a train. The train can be separated



BALL-SOCKET CONNECTOR ASSEMBLY

FIG. 1.

easily but only by turning a rod out of line by more than 45° to free the ball. There should then be no danger of the train separating when it is confined to the irradiation channel, but the coupling has fractured. (17) It is true that the design would not be suitable for beryllium or carbon steel that become embrittled by irradiation so that an impact could break the stem below the ball. Most stainless steels can be embrittled by stress corrosion in the presence of chloride or oxygen; again the stem would break at the point of attack. In Zircaloy, however, the break was some distance from the point of attack.

What happened was that the weld securing the threaded portion of the stem was not a perfect seal. Water entered along the screw threads to the end that is close to the fuel and therefore very hot so that rapid corrosion occurred. The hydrogen taken up moved down the thermal gradient to the stem where it precipitated as hydride and embrittled the metal. This hydrogen migration down the thermal gradient and precipitation as hydride is a known and well established phenomenon in zirconium and has been discussed explicitly by Sawatzky and Vogt. (18) Thermal gradient precipitation in liquid solutions was studied more than a century ago by C. Ludwig in 1856 (19) but is usually known as the Soret effect after Ch. Soret who discovered it independently in 1879. (20)

The second example of a weakness in design was discovered while developing the garter spring spacer used to separate the

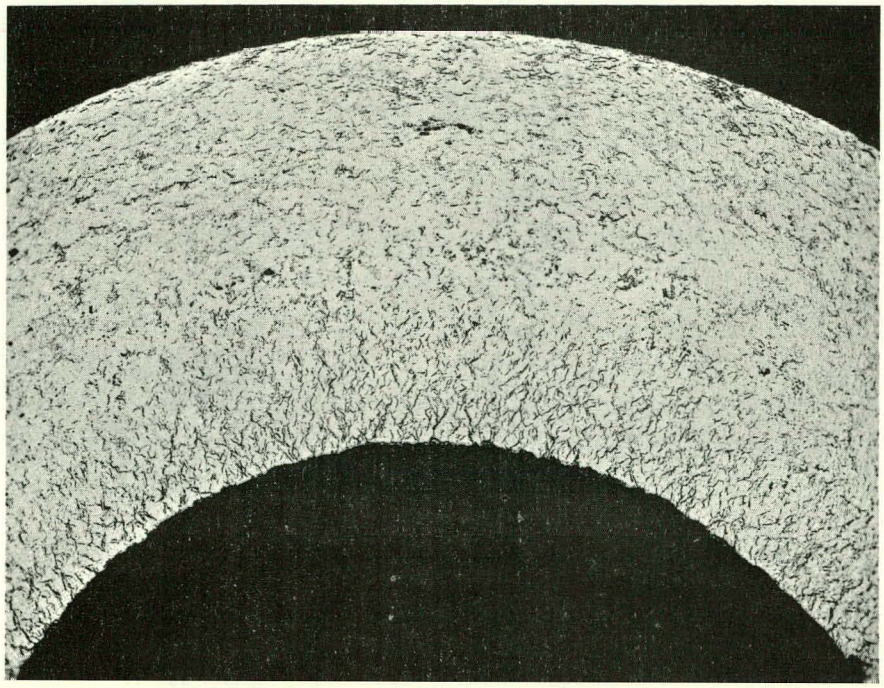
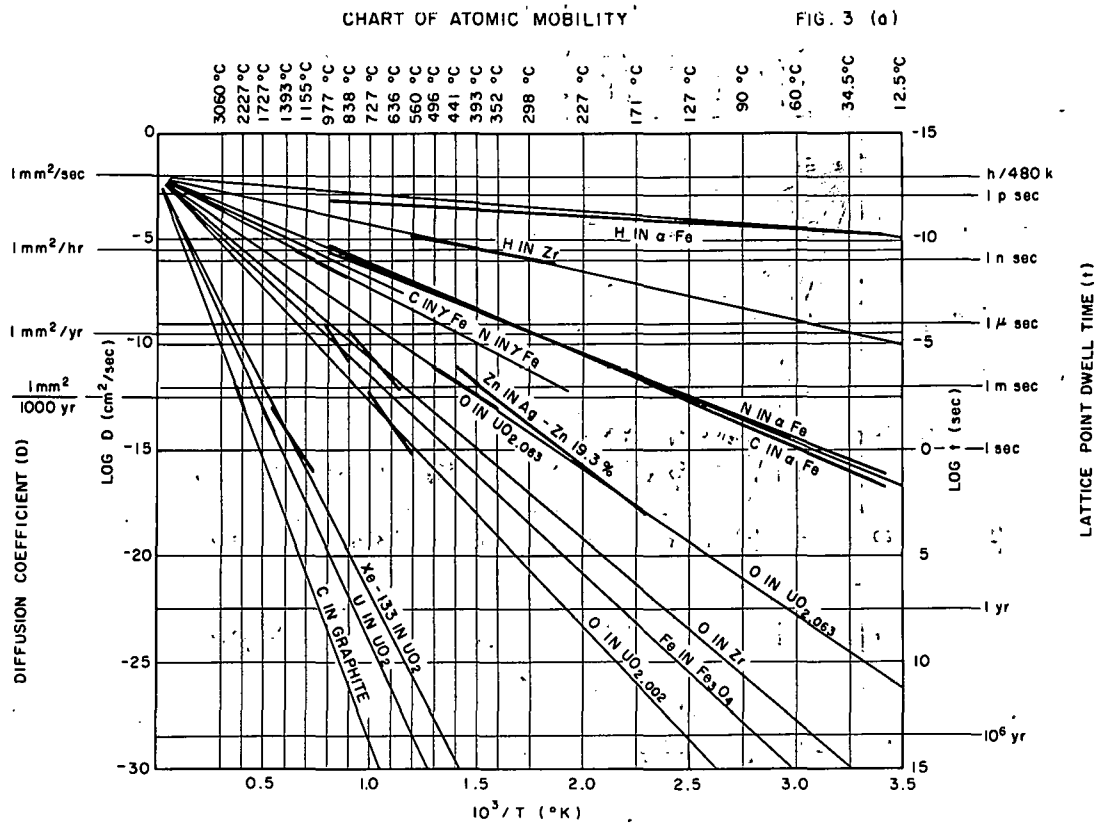


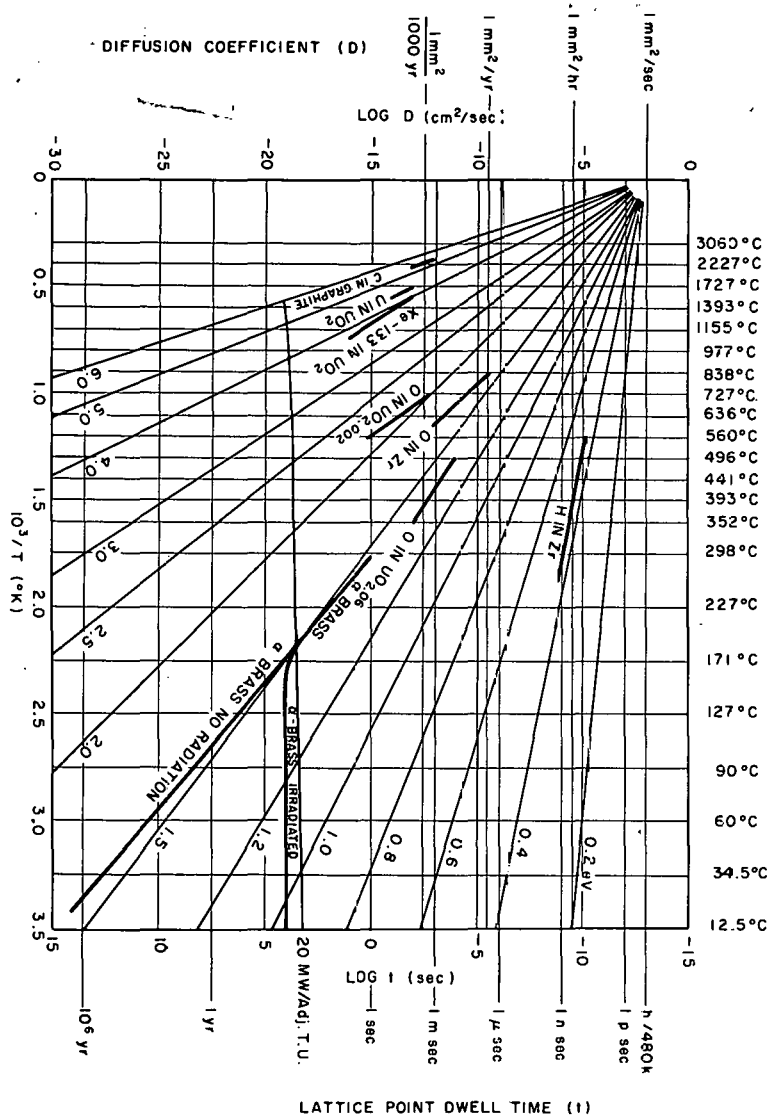
Fig. 2 - Section of Zircaloy-2 garter spring. (50X)

hot pressure tube carrying the coolant from the surrounding thin-walled calandria tube cooled by the moderator. The garter spring is simply a helically-coiled wire formed into a loop or garter that embraces and can roll along the outside of, the pressure tube. (21) The operation of coiling the Zircaloy wire introduces a compressive strain on the inside giving the metal a grain texture directed along the coil radius. After a long life hydride platelets would precipitate there also in a radial orientation as shown in the longitudinal section through the wire in Fig. 2. The garter spring carries a load that would tend to flatten the coil on two sides and eventually it would fail there by cracking at the hydride platelets. The difficulty would be avoided by forming the spring by cutting it as a helix from a tube, for the texture in drawn tubing is satisfactory. Heat-treatable Zr-2.5% Nb alloy offers other means of fabrication. Inconel garter springs are used in NPD but for economy it is planned to use a zirconium alloy in CANDU.

A study is in progress to assess the prospects of substantially reducing the rate at which hydrogen accumulates in zirconium alloys and thereby of extending the useful life of components in service to a hundred years, or effectively indefinitely. Before discussing this in particular it may be helpful to introduce an extensive map or chart of atomic mobility shown in Figs. 3(a), 3(b) and 3(c).



The units of the abscissa of Figure 3(b) should read "$10^3/T(^oK)$" instead of "$10/T(^oK)$."



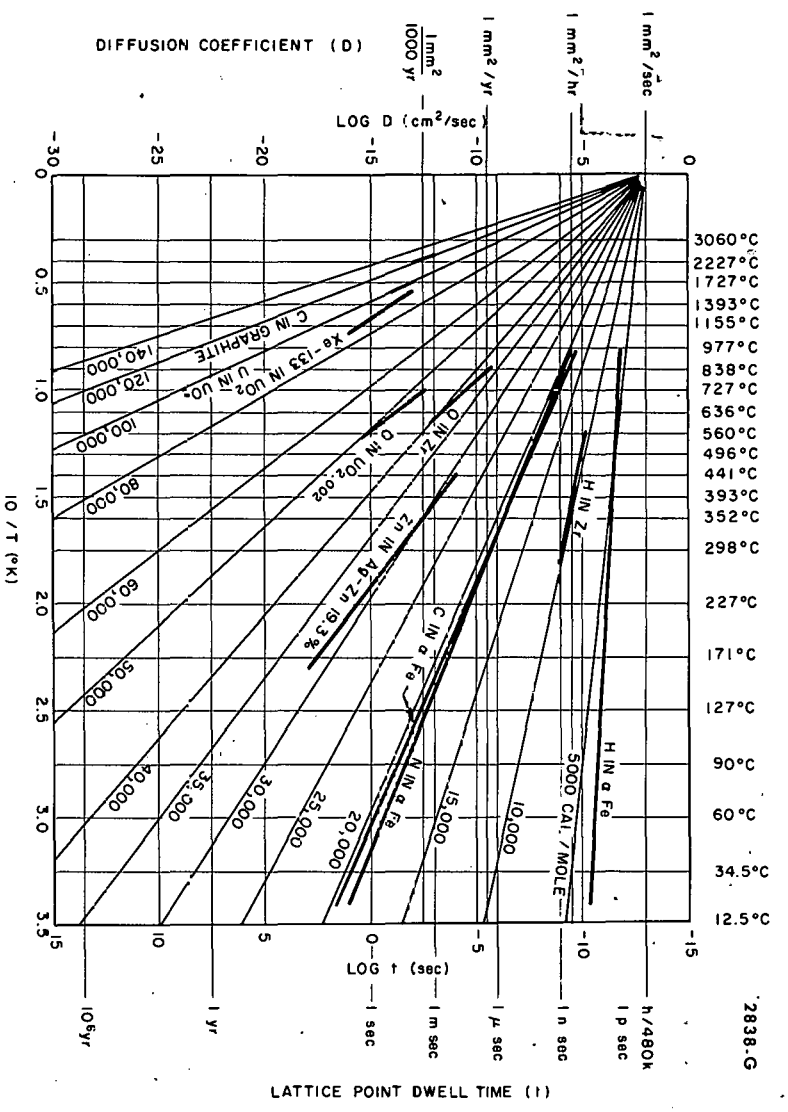


CHART OF ATOMIC MOBILITY

The charts, Figs. 3(a), (b) and (c) are simply the Arrhenius-type plot of diffusion coefficients with appropriate scales.

The scales on the right are derived by first noting that 10^{-15} cm^2 (i.e. $\approx$ (3 Å)^2) is approximately the area of a single cell of the atomic lattice. Dividing D by 10^{-15} cm^2 then gives the frequency with which an atom makes a jump from one lattice cell to the next or the number of jumps per second. The reciprocal of this jump frequency is termed the lattice point dwell time and represents the mean time spent by the diffusing atom before leaving any given position in the lattice. The range of dwell times extends from 10^{-13} sec to extremely long times so the chart is cut off at 30 million years.

For example, H at 300°C in zirconium has a dwell time of 1 nanosecond (10^{-9} sec). By the scale on the left this corresponds to a diffusion rate of about 0.3 mm^2/hr.

The reason for drawing the lines on the chart that radiate from the point $T = \infty$ and jump frequency = 10^{13}/sec, is to serve as guide lines to show up the departures of the experimental results from the idealized form $D = 0.01\ e^{-Q/kT}$ cm^2/sec. The frequency 10^{13}/sec (which is the same as 0.01 cm^2/sec on this chart) is chosen because it is the atomic vibration frequency for Fe. The atomic vibration frequency or Debye frequency, ν, is related to the Debye temperature Θ_D by $h\nu = k\Theta_D$. Since the actual Debye temperature for Fe is about 480°K and for most substances the Debye temperatures range from about 80°K for Pb to about 2000°K for diamond, the factor by which these differ from 480°K is never large enough to warrant complicating the chart. The use of the guide lines does not imply that the actual diffusion constants should conform to them but does imply that departures have some significance. In fact the literature is crowded with discussions of all the many factors that affect the mobility of atoms in solids. (22)

Suppose the concentration of hydrogen in Zircaloy-2 at 300°C has reached the solubility limit of 100 ppm by weight that corresponds to 1 atom of H per 110.5 Zr atoms. (18) (Note that the reason for retaining more significant figures than justified by the accuracy is to identify the quantities when they appear at different addresses, this particular number is $(A_H/A_{Zr}) \times 10^4 = 1.0081 \times 10^4/91.2$.) On any average plane within the metal the population density σ(atoms/cm^2) = n(rate of arrival/cm^2) x γt (dwell time) where γ is a geometrical factor of about 3 because 4 out of 6 adjacent positions lie

in the plane. Now the average density of zirconium atoms in a plane is $(\rho N/A)^{2/3} = 1.226 \times 10^{15}$ (for $\rho = 6.5$ g/cm^3) and $\sigma_H = 1.226 \times 10^{15}/110.5 = 1.11 \times 10^{13}$, and from the chart, $t = 0.9 \times 10^{-9}$, so $n = 1.22 \times 10^{22}/\gamma \doteq 4 \times 10^{21}$ atoms/cm^2 sec. At 300°C the same rate of arrival of hydrogen atoms at a surface in hydrogen gas occurs at a pressure of about 0.9 mm Hg.

Since the concentration was chosen at the solubility limit, this flux of hydrogen must be that between the surface of ZrH and the solution. It follows that the dwell time at the surface of the hydride is greater than in the solution by the ratio of concentrations, or about 100. This is a relatively small factor and conforms with the relatively low heat of formation of zirconium hydrides.

Conditions are different at a boundary between zirconium and hydrogen gas. By extrapolation of the equilibrium between hydrogen gas and zirconium at higher temperatures, it is inferred that at 300°C the equilibrium pressure would be about 10^{-7} mm Hg. (23) This means that hydrogen is very readily trapped by a zirconium surface. To explain the very low rate of hydriding observed in reactor practice it appears that the oxide layer at the zirconium surface is a very effective barrier against the ingress of hydrogen. Moreover, the high flux of hydrogen possible within the metal implies that the surface layer is also a strong barrier against egress.

By bonding two sheets of Zircaloy together with nickel and exposing the edge in high temperature water, it has been possible to promote a very rapid uptake of hydrogen. (24) By exposing such a system in an oxidizing medium it should be possible to drain off hydrogen. Experiments on these lines have given inconsistent results but two cases of a loss of hydrogen from Zircaloy have been reported when the medium was air containing nitrogen oxides. (25) If such a mechanism could be brought under control it might at least be possible to achieve an indefinitely long life for the pressure tube since it is exposed to a gas on the outside.

It may be noted that the density of hydrogen at the 300°C solubility limit of 100 ppm is about 3.9×10^{20} atoms/cm^3 or equivalent to 14 atmospheres at 0°C in a monatomic gas. Comparing this with the "equivalent pressure" of 0.9 mm found above, the dissolved hydrogen must be regarded as a very "sticky" gas despite its nanosecond dwell time.

RADIATION EFFECTS

To complete the overall picture it is of interest to evaluate the effects of radiation intensities at practical levels. The major components of radiation adjacent to, but not within, reactor fuel that contribute to energy deposition, are fast neutrons and γ-rays. From both fast neutrons and γ-rays ionization events and electronic excitation result in the course of the degradation of the energy to thermal levels. Almost all the energy at some stage of this degradation passes through the range of 5 to 0.2 eV that covers the activation energies necessary for breaking chemical bonds and displacing atoms. Given the rate of energy deposition from fast neutrons and γ-rays it is then possible to derive a crude estimate of the upper limit of the effect of the radiation on atomic movements. There are at least two basic reasons why the estimate cannot be exact: on the one hand for many atomic movements, since only an activation energy is involved and not necessarily any absorption of energy, a whole chain of events can result from one excitation; on the other hand the time required for atomic displacement can be much longer than electronic de-excitation times so that much of the excitation energy is ineffective. Nevertheless it seems of interest and significance to plot on the chart of Fig. 3 the crudely estimated upper limit of radiation effects. Fig. 3 is reproduced in three forms, (a), (b) and (c); Fig. 3(c) shows activation energies in units of eV/atom and appears most directly relevant. It is not easy to make estimates of radiation energy deposition from first principles, so empirical calorimetric results will be used. (26) The energy deposition in matter close to fuel operating at 20 thermal watts/g U (i.e. 20 MW/tonne U) is about 1 W/g (more exactly 0.6 W/g for heavy elements and 1.5 W/g for hydrogenous moderator), or 6.2×10^{18} eV/sec g or 10^{-5} A eV/sec atom of atomic weight A. Setting A = 50 gives for the mean time interval $10^5/A = 2 \times 10^3$ sec/eV at 20 MW/adjacent tonne U. Within the fuel the interval is 20 times less or 100 sec/eV at 20 MW/tonne U. These results are plotted as radiation dwell times against activation energy in Fig. 3(c).

It is possible to compare this rough estimate with the experimental results of Dienes and Damask (27) on the effect of radiation on atomic diffusion in α-brass, also shown in Fig. 3(c). Their results were obtained in the Brookhaven reactor at a flux of 5×10^{12} n/cm^2/sec of which about 25% was fast. This corresponds to a level of about 1 to 2 MW/adjacent tonne U, so their observed effect of radiation-enhanced diffusion in α-brass is perhaps five times as much as expected from the rough estimate. Probably the agreement should be considered satisfactory.

Some further examples of the use of the rough estimate and the chart of atomic mobility may be mentioned. Below 1500°C radiation in sufficient amount would be expected to leave some

damage in graphite. At this temperature, however, only certain special changes may remain because the diffusion results plotted refer to diffusion of C-14 tracer in crystals of natural graphite. Movement of interstitial atoms and atoms at grain boundaries takes place more readily.

Recalling that in the fuel itself the energy deposition rate is 20 times higher than in adjacent matter, it may be expected that the behaviour of Xe-133 in UO_2 fuel is markedly affected by irradiation at temperatures up to 1000°C. Experiment confirms this expectation.

There are additional indirect effects of radiation, particularly in materials of low thermal conductivity where local high temperatures and steep thermal gradients may be established.

URANIUM DIOXIDE

One of the many excellent properties of UO_2 as reactor fuel is that no significant swelling occurs if the cladding is ruptured and water comes into contact directly with the oxide. Cases have occurred in which a rupture of the cladding has developed with local hydriding and embrittlement. Such events have been rare and most have been attributable to a cause that can be guarded against in the specification; for example, the presence of CaF_2 in the oxide (28) or excess moisture in compacted oxide powder (29) without a means of escape except by attacking the inside of the hot cladding. As the rupture enlarged in all cases there has been a marked escape of radioactive fission products into the coolant and some escape of UO_2. One case at high temperature power reactor conditions was allowed to develop for eight weeks. (30) In all cases the subsequent decontamination of the coolant piping has been the most costly of the real, as distinct from the imagined, consequences. In many tens of experiments with deliberately defected fuel cladding, the system has appeared stable. The one irradiation carried on for more than a year showed no sign of change or of reaching a limit (31); the hydrogen content of the cladding was no greater than expected. The one or two cases where the rupture has extended without an obvious cause, have appeared quite exceptional, and it is not possible to exclude some built-in fault such as a fluoride pickle stain (32) that may have existed on the inside of the cladding. It seems probable that the present specifications for both oxide and cladding are more than adequate. (33)

There still appears to be no case on record that shows it to be necessary to leave any free space to accommodate escaping fission gases. Cases are known where adsorbed gases on compacted oxide fuel have been released and expanded as the fuel heated up and have led to distension and even explosive rupture of the cladding. (34) There are also a number of cases of flat plate

fuel elements taken to high irradiations of 25,000 to 100,000 MWD/T that have failed by swelling. (35) The later stages of this swelling seem attributable to the presence of a gas-filled gap between the fuel and the cladding that allows all the fuel to reach a very high temperature and expel the fission gases. This mechanism would not exist for the usual solid round rod of oxide with a thin metal clad in high pressure water without any special free space.

Failures have occurred by a mechanism termed "water-logging" to which the existence of free space contributes. When a reactor is shut down and the fuel cools, water may enter such a free space through the rupture, that may be only a pinhole. On return to power the water may vaporize so rapidly that the cladding is split.

Many cases of irradiated fuel in normally sealed cladding have been reported in which the fission gas released has been 20% or even more of that produced, and it has been calculated that without added free volume a pressure sufficient to distend the cladding would have been produced. In all known cases, however, there was a free space present and the resultant pressure was insufficient to distend the cladding.

The direct evidence that free space may not be necessary comes from the experiment that showed natural xenon at a density corresponding to 200 psi at room temperature to be absorbed by UO_2 of density 10.1 g/cm^3 at a rate proportional to the rate of fission and greater than the rate of gas production in fission. (36) A subsidiary experiment has shown that the trapped gas is firmly bound and only about 1 per cent is released below 1450°C. (37)

Economic reasons for wishing to eliminate free space are quite strong, because free space reduces the competitive neutron capture by the fuel. Also, if free space is left at the end of a long rod that is not also the end of the reactor core, the resulting peaking of the neutron flux may require a reduction in the mean power rating of the fuel. It is then better to leave any free space by dishing or grooving the ends of individual UO_2 pellets forming the rod.

The case against free space is not complete; it may still be necessary to specify some degree and distribution of closed pores within the fuel, or there may be advantages of operating fuel at higher temperatures at which the gases are expelled.

FURTHER DIFFUSION PHENOMENA

Despite the much higher diffusion rates for oxygen in excess of the stoichiometric UO_2 composition shown in Fig. 3(a) there will still be a drift of oxygen down the thermal gradient until balanced by the concentration gradient. This Soret effect is most marked at high temperatures, both because the diffusion rates are higher and because the differential with concentration is smaller. When UO_2 fuel cools, the concentration gradient is liable to be frozen in, but regions that were over 2000°C when hot may have time on cooling to separate into two phases, UO_2 and U metal, and this has been observed in a number of laboratories. One interesting result observed by Rothwell at Harwell is that at 1450°C where this redistribution is believed to take place, a burst of fission gas previously trapped in the oxide is released.(38)

It is clear from Fig. 3 that the rate of diffusion of xenon in UO_2 is much less than that of oxygen. It is also less than that of many fission products. The actual movement of xenon, especially at temperatures below 1500°C, may be due to the movement of these other atoms. In dense sintered UO_2 there are usually a large number of small closed pores of 0.1 to 1 μm diameter that will trap migrating xenon atoms that reach them. In the absence of radiation these pores will be very secure traps and could fill to a very high gas pressure. Under irradiation, however, it has been demonstrated that xenon is absorbed back into the oxide at a significant rate. (36) Such pores must, however, still be regarded as deep traps requiring more than 5 eV to dislodge most of the contained atoms, but probably much less to trap those already adsorbed on the pore walls. (In fact more than 500 eV kinetic energy is required to drive a free xenon atom into the surface. (39)) In the interpretation of diffusion measurements, the possibility must be examined that only a fraction of the contained population is taking part in the observed migration.

CONCLUSION

In the selection of materials for use in reactor cores, the effects of temperature- and radiation-induced rearrangements such as of hydrogen in Zircaloy, oxygen in Zircaloy, oxygen and fission products in UO_2 fuel, the radiolysis of water and surface interactions, must be studied as normal phenomena to be incorporated in engineering design considerations. In the present limited state of knowledge overdesign is common and ignorance factors are too often euphemistically described as safety factors. In part the ignorance is attributable to the appearance of complexity in the established knowledge. The rough approximations introduced in this review are offered as aids to comprehension and not as substitutes for detailed studies.

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   The capture cross section per kg of material of atomic mass A is $1000\ N\sigma \times 10^{-24}/A\ cm^2$ where N is the number of atoms per g atom. In an irradiation I n/kb the number of neutrons captured is then $N\sigma I/A$ and the mass of neutrons captured is $\sigma I/A$. Now 1 g neutrons + 238 g U → 239 g Pu. If Pu in the reactor is assigned the same value as U-235 that yields 0.8 MWd/g, the value is \$0.8F by definition of F. The corresponding value of neutrons is $\$0.8 \times 239F = \$191.2\ F \approx \$200F$ and the cost of irradiating the kg of material is $\$200\ \sigma IF/A$.

   One weak point of this derivation is that Pu and U-235 are not equivalent in neutron yield that is proportional to $\sigma(\eta-1)$, the higher cross-section of Pu is valuable at the end of the irradiation but can be a costly embarrassment at earlier stages in some fuelling systems. Another weak point is that the limiting irradiation is more than proportional to the fissile material content in practical cases of bi-directional slug fuelling. These advantages of producing plutonium are offset by the necessity to remove it with the spent fuel.

   The evaluation should be accepted as a crude approximation.

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	mW/g		mW/g
Graphite	68.5	Magnesium	60.3
Polyethylene	188	Aluminum	60.3
Polystyrene	139	Beryllium	73.3

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