CHAPTER 3

Diversity of Fuel Forms

A feature of nuclear fuel which oil fuels cannot offer is the diversity of fuel forms. Although uranium is mined as a metallic ore, it can be used in a variety of different physical and chemical forms . . . solid, liquid, or gaseous. Even in the solid form, which categorizes the present state of nuclear ship fuel art, there are a wide variety of fuel shape possibilities. This diversity opens up opportunities for new reactor systems in the future. Actually, the diversity of fuel forms has been forced upon the nuclear fuel designer as a means of overcoming the capricious nature of uranium metal, and as a means of maximizing its heat extraction efficiency. In a sense, this is analogous to the atomizing process of conventional oils. With nuclear fuels, however, we must train our minds into the framework of fissionable atoms, and pursue their fate in the reactor core. In particular, we want to burn as many of these atoms as possible . . . with minimum self-damage to fuel elements.

3-1 Properties of Uranium

The uranium content in uranium-bearing ores throughout the world averages around 1%; less in the United States. At the mines, therefore, effort is made to upgrade the uranium content before shipment to process plants. At the process plants, the ore concentrates are converted into "green salt" (uranium tetrafluoride: UF_4) which then is the feed material for making U-metal. When purified and poured into ingot molds, uranium can be purchased as "reactor grade" metal at the Government-controlled price of $18 per pound. From ingots, the U-metal is fabricated into useful fuel shapes.

Freshly prepared U-metal is dense (approximately 1200 pounds per cubic foot), and white-to-grayish in color with a distinct luster. It oxidizes readily in air; it readily absorbs impurities from process equipment; and, when in finely divided form, it is pyrophoric (ignites spontaneously). It is radioactive, but only slightly so . . . with a decay half-life of several billion years.* It emits a high energy alpha particle, which is an inhala-

* The radioactive half-life of a substance is the time required for its radioactivity to decay to one-half of its original amount.

tion hazard. Aside from its radioactivity, it is also toxic. For all of these reasons, fabrication of the metal is generally done in an inert atmosphere or under a vacuum.

The most intriguing property of U-metal is its crystalline behavior between ambient temperature and its melting point of 2070°F. Curiously, its crystalline structure (i.e., the arrangement of atoms into metallic crystals) prevails in three different phases, designated as alpha, beta, and gamma (nothing to do with radioactivity). Alpha uranium (below 1220°F) has three unequal crystalline axes, each with a different coefficient of linear expansion. In the beta phase (between 1220°F and 1420°F), one of the axes disappears entirely, leaving two unequal lengths . . . also with different expansion coefficients. In the gamma phase (1420°F to melting), the crystals return to three axes—this time, of equal lengths, and of equal expansion coefficients. A summary of these anisotropic characteristics of uranium is given in Table 3-1.*

Table 3-1. The Anisotropic Properties of Uranium

Phase	Crystal Dimensions 10^{-8} cm	Crystal Volume 10^{-24} cm^3	No. Atoms per Crystal	Expansion Coefficient x 10^{-6}/°F
Alpha (to 1220°F)	a_o = 2.920 b_o = 5.834 c_o = 5.064 (orthorhombic)	86.27	4	a_o = 20.4 b_o = -5.2 c_o = 19.1
Beta (to 1420°F)	a_o = 10.759 c_o = 5.656 (tetragonal)	654.7	30	a_o = 12.8 c_o = 2.5
Gamma (to 2070°F)	a_o = 3.493 (cubic)	43.76	2	a_o = 10.0

The intrigue comes when we try to use U-metal as a fuel in a reactor. We can't predict with precision the upper limits of the crystalline distortions: much depends on the purification and fabrication history; on the grain size and heat treatment; on the volumetric number of atoms fissioned . . . and on the residues accumulated. In extreme cases, a two-inch sample (¼-inch diameter) has grown to 12 inches!† This phenomenon is called the "growth problem" of uranium, and is brought about by the volumetric changes during its phase transitions. Rarely do the transition characteristics retrace themselves upon heating then cooling (see Fig. 3-1).

* Anisotropic means different properties in different directions.

† Ref: *Nuclear Fuels*, Gurinsky and Dienes, Van Nostrand, 1956, p. 213.

Fortunately, the very property of anisotropy makes uranium an unusually versatile fuel material. It has a high chemical affinity for many elements. As a result, many additives (i.e., alloys and compounds) combine readily with uranium in the solid, liquid, and gaseous states. These additives, when properly chosen, can significantly improve the properties of uranium. Note that we say "improve." We cannot negate entirely the capricious nature of the U-metal.

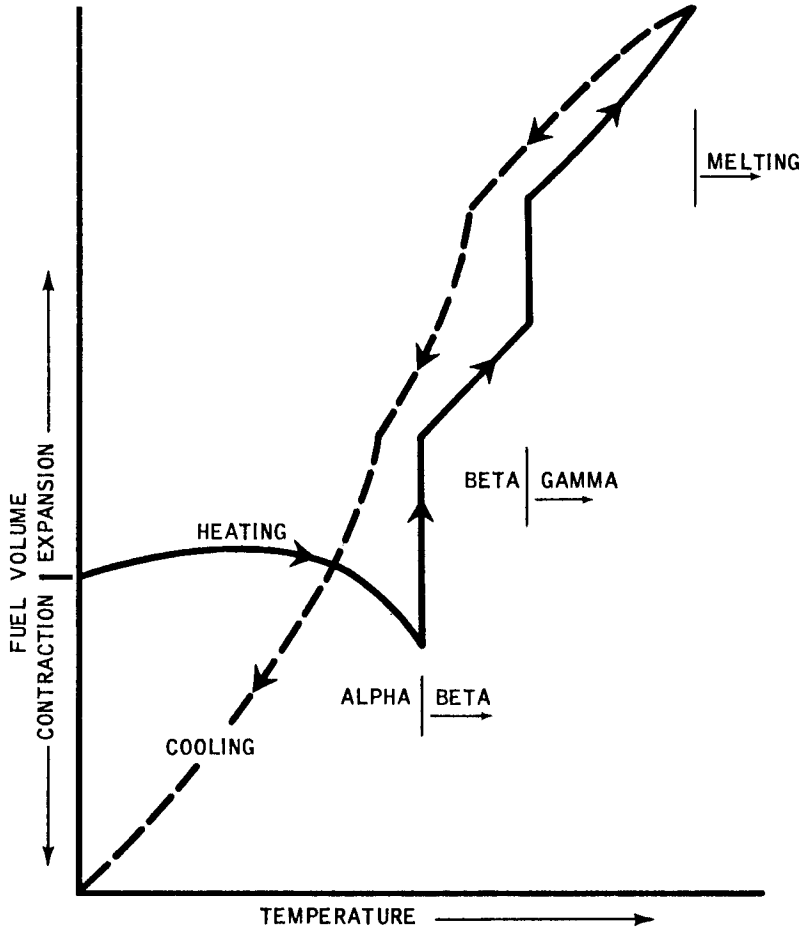

Fig. 3-1 A Typical "Phase Transition" History of U-Metal

3-2 The Enrichment Process

Regardless of the number of additives to U-metal, we still have natural uranium with its natural enrichment of 0.72% U-235. As was indicated earlier, we want to better this natural state. To do so, we hold back the non-fissionable U-238 by successive stages of "isotope separation."*

* U-235 and U-238 are called "isotopes"; that is, they have the same physical and chemical properties but their nuclear properties are quite different.

When green salt is reacted with fluorine gas, we get uranium hexafluoride: UF_6. This substance is solid at ordinary temperatures but at about 135°F (atmospheric pressure) it sublimes into a gas. This UF_6 gas is the feed material for isotope enrichment through Government gaseous diffusion plants.

The principle of isotope enrichment is this: in the gaseous state, molecules will separate themselves through a porous barrier in proportion to their molecular weights. The lighter molecules, being faster, will get through before the heavier ones (provided a pressure gradient exists across the barrier). For each barrier transversed, the theoretical enrichment factor is

[Eq. 3-1]

$$\text{E.F.} = \sqrt{\frac{\text{M (heavy)}}{\text{M (light)}}} - 1$$

In the case of UF_6 gas, the M-heavy is $238 + (6 \times 19) = 352$; the M-light is $235 + (6 \times 19) = 349$.* On this basis, the enrichment factor of U-235, after passing through a barrier, is 0.0043. Thus, theoretically, after the first barrier, the enrichment factor would be 0.0072 + 0.0043 or 0.0115 (1.15%). This single-stage enrichment is not achieved in practice, and further increases in enrichment require many, many stages of porous barriers.

Table 3-2. Cost of Enriched Fuel in UF_6 Form

Enrichment Fraction	$/kg U	$/g U-235	$/lb U-235
Nat.	40.50	5.62	2,640
0.01	75.75	7.58	3,440
0.015	145.50	9.70	4,400
0.02	220.00	11.00	5,000
0.04	535.50	13.39	6,430
0.06	862.50	14.38	6,550
0.08	1,195.00	14.94	6,970
0.10	1,529.00	15.29	6,920
0.25	4,078.00	16.31	7,400
0.50	8,379.00	16.76	7,600
0.75	12,721.00	16.96	7,690
0.90	15,361.00	17.07	7,750

f. o. b. Oak Ridge, Tenn.

Ref: Nucleonics, December, 1956 (Vol. 14, No. 12), p. R2.

Unfortunately, the number of barrier stages required to achieve a specified degree of enrichment is not a straight-forward linear relationship. Not only is the process inherently inefficient, but much depends

* Fluorine has a molecular weight of 19; there are 6 atoms of fluorine associated with each atom of U, hence 6×19 is the proportionate weight of fluorine in a molecule of UF_6.

on the enrichment take-off, the waste bleed-off, the interstage cycling, and the pressure differences across the barriers. The consequence is that it takes about 1000 stages (in round numbers) to go from natural uranium to 10% enrichment; another 500 to go from 10 to 50% enrichment; another 500 to go from 50 to 90% enrichment. It takes approximately an additional 1000 stages to go from 90 to 100% enrichment. So, 90% enrichment is about the practical limit for nuclear fuels.

The number of enrichment stages directly influences the cost of enriched fuels. Typical cost figures are given in Table 3-2. Note that the cost is cited in two ways: dollars per kilogram uranium (which is U-235 plus U-238) and dollars per gram U-235. The use of two costs is the usual practice in the nuclear industry. Corresponding dollars per pound of U-235 are shown in Table 3-2 for convenience.

3-3 Thorium and Plutonium

Two other nuclear fuel materials could possibly be used in ship reactors, namely: U-233 produced from Th-232 (thorium), and Pu-239 (plutonium) produced from U-238. Like U-235, both U-233 and Pu-239 are thermally fissionable. These last two isotopes are not found in nature and hence the enrichment process cannot be used to obtain them. Instead, they are produced in a reactor by the consumption of neutrons. Their nuclear reactions of production are:

$$^{232}\mathrm{Th} + \mathrm{n} \longrightarrow {}^{233}\mathrm{Th} \xrightarrow{23.3\ \mathrm{min}} {}^{233*}\mathrm{Pa} \xrightarrow{27.4\ \mathrm{days}} {}^{233}\mathrm{U}$$

$$^{238}\mathrm{U} + \mathrm{n} \longrightarrow {}^{239}\mathrm{U} \xrightarrow{23.5\ \mathrm{min}} {}^{239*}\mathrm{Np} \xrightarrow{2.3\ \mathrm{days}} {}^{239}\mathrm{Pu}$$

Thorium ores are about three times as plentiful throughout the world as uranium ores and, hence, hold promise for the future. When produced in commercial quantities, natural thorium could be used as a fertile material in a reactor for converting to U-233. In marine reactors, the fertile thorium could be arranged in the form of thermal shields (recall Sec. 1-6) between the regular fuel elements and the inside walls of the reactor vessel. Thorium has a melting point in excess of 3000°F. Its corrosivity, pyrophoricity, and fabricability compare with uranium. Yet, thorium does not have the anisotropy of uranium; it acts as a "well-behaved" metal. To extract U-233 from irradiated thorium, chemical separation (not enrichment) techniques are used.

Since U-235 cannot be bought in completely pure form, there is always some U-238 present. To some extent, this is a bonus factor, as the U-238 is a fertile material for producing Pu-239. Depending on the degree of enrichment used, some of the U-238 will convert to Pu-239 directly in the fuel elements. This is called "internal conversion." Internally converted Pu-239 is fissionable along with U-235.

* Pa is protactinium; Np is neptunium.

The internal conversion of U-238 to Pu-239 is an inefficient way of using vital neutrons. In the first place, only 70% of the neutrons thermally captured by Pu-239 actually cause fission . . . compared to 85% fission from U-235. Secondly, the number of net neutrons produced from Pu-239 is about 10% less than the number from U-235. And, thirdly, the U-238 conversion doesn't stop at Pu-239 but goes on to Pu-240, Pu-241, and Pu-242. All of these higher Pu isotopes capture neutrons but only Pu-241 is fissionable. Hence, the internal conversion of U-238 to the Pu chain generally is not by design, but since U-238 is present in the fuel anyhow, every advantage is taken of it.

Pu-239 is more efficiently produced in special reactors ashore, where, subsequently, it could be available as the primary fissionable fuel for nuclear ships. The physical properties of Pu differ considerably from those of U. Instead of the three crystalline phases of U, Pu has six; instead of a prominent growth problem, Pu suffers a prominent *contraction* problem. Pu melts at 1185°F as against 2070°F for U. However, Pu-239 is more readily fissionable by fast neutrons than is U-235, and it is more feasible to achieve 100% Pu-239. For these last two reasons, particularly, Pu-239 is more valuable as a military explosive than as a nuclear ship fuel.

Of the three fissionable fuels available for reactors, more is known and more has been done about the technology of uranium—and its U-235—than about thorium or plutonium. Consequently, the discussion of fuel forms from this point on pertains only to uranium . . . at any degree of enrichment desired.

3-4 Metallic Base Fuels

Uranium metal can be shaped and joined by all conventional metal-forming techniques (i.e, rolling, forging, extruding, machining, welding, brazing). Because of this fabricability, U-metal is one of the most practical materials for forming nuclear fuel elements. When "sandwiched" between its jacket or cladding material, metallic uranium can be made virtually into any shape desired.* For example, it can be made into parallel flat, curved, or corrugated plates; into concentric or finned thin-walled tubes (see Fig. 3-2). This versatility in shape is particularly advantageous for maximizing surface areas for the removal of the fission heat. Furthermore, clad U-metal fuels are particularly useful where the coolant serves also as the moderator. The cladding material may be aluminum, zirconium, stainless steel, or other good heat conductive materials.

A necessary feature of the uranium-clad design is the proper bonding of the U-metal to the jacket material. A special bonding material is required for several reasons, namely:

(1) to provide structural support for the uranium metal (uranium loses strength rapidly at elevated temperatures);

* Note that "jacket" and "cladding" are used synonymously.

(2) to provide good heat transfer from the metal fuel to the metal jacket;
(3) to retard corrosion, either of the fuel by its jacket, or of the jacket by the fuel;
(4) to retard interatomic diffusion of the cladding material into the uranium, which would jeopardize the integrity of the cladding; and
(5) to provide a "cushion" or resilience against the thermal distortion, volumetric growth, and radiation damage of the U-metal.

The bonding material may be a solid (e.g., Al-Si coating), a liquid (e.g., NaK: a low melting-point metal), or a gas (e.g., helium).* In some cases

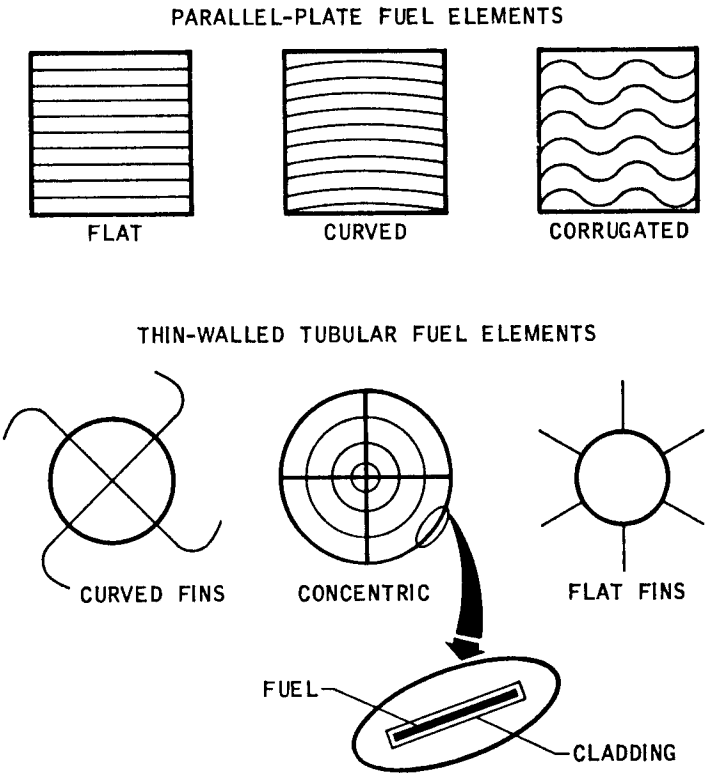

Fig. 3-2 Typical Shapes for U-Metal Fuel Elements

a separate bonding material is not required. Instead, an extra thickness of cladding is used and the fuel metal is drawn or pressed to fit the fuel-cladding interface. (See Fig. 3-3 for a generalized arrangement of bonding.)

* Al-Si is aluminum-silicon; NaK is sodium-potassium.

One of the most troublesome features of cladded fuel metal is self-damage. This damage is caused by the high-energy fission fragments and neutrons. When a fission event occurs within the fuel, the flying-apart fragments tear loose some of the neutral bonds of the fuel metal crystals. In addition, the destructive fragments may knock-on unfissioned fuel atoms from one crystal to another; or the fragments may electronically excite the fuel atoms so that the fuel atoms vibrate out of their crystal lattices. Furthermore, some of the fission residue atoms may unite chemically with neutral fuel atoms and thereby change radically the properties

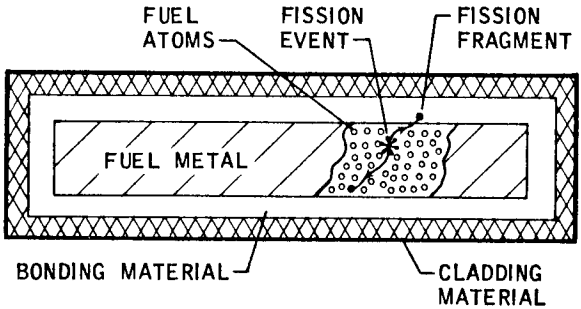

Fig. 3-3 Functional Arrangement of Fuel Element Bonding

of the fuel metal. A gaseous fission residue may lodge within the interstitial spacing of fuel crystals and "burst" it from the inside. Neutrons racing back and forth do similar damage to the fuel material. The use of alloying materials helps only partially to overcome the severity of the total damage. As a consequence, seldom can a U-metal fuel element exceed a fuel atom burnup of 1%. Beyond this amount of burnup, the fuel material is so badly radiation-damaged that it has to be replaced.

There is another limitation, too. Because of the marked anisotropy (Sec. 3-1) of the U-metal at its alpha-to-beta phase transition, the maximum central fuel temperature must be design-limited to 1200°F. This is a very low "driving force" when compared to the 3500°F combustion gas temperature in an oil-fired boiler. As a consequence, the maximum heat atainable per gram of fissionable fuel (called "specific power") is only about 500 watts per gram of U-235.* This is a far cry from the theoretical possibilities of Sec. 1-3.

3-5 Ceramic Base Fuels

Most of the disadvantages of metallic base fuels are overcome by the use of nonmetallic fuels in ceramic form. These fuels are compounds (not alloys) of uranium with such nonmetallic materials as oxygen, silicon, carbon, nitrogen, etc. (see Table 3-3). The atoms of these ceramics com-

* 1 watt = 3.413 Btu/hr.

bine readily with the atoms of uranium to form extremely tight and well-organized crystal lattices. A number of uranium-ceramic compounds have been investigated and their significant features are as follows:*

(1) superior dimensional stability;
(2) excellent resistance to corrosion;
(3) very high melting points (up to 5000°F);
(4) low thermal neutron absorption in the non-fuel material;
(5) good resistance to radiation damage; and
(6) reasonable density of contained fuel.

Table 3-3. Selected Properties of Ceramic Fuel Compounds

Compound	Uranium Wt. %	Uranium* g/cm³	Vol. Change %	Melting Pt. °F	Nuclear** Absorption b/atom
UAl_2	81.52	6.64	-30.9	2850	0.230
USi_2	80.91	7.48	-22.1	2900	0.13
UFe_2	68.0	8.98	- 6.5	2200	2.53
UO_2	88.1	9.60	0.0	5150	0.0002
UC_2	90.8	10.61	+10.4	4500	0.0032
UC	95.19	12.97	+35.4	4000	0.0032
UN	94.44	13.52	+40.5	4750	1.88

* Corresponding to density of uranium metal 18.9 g/cm³.
** For second element of compound only (not of the U).
Ref: "Power Reactor Technology", General Nuclear Engineering Corp., Vol. 1, No. 23, Feb., 1958, pp. 28-29.

Because of the advantages above, ceramic base fuels are able to attain fuel element center temperatures of from 2000°F to as high as 5000°F. The attainable specific powers are on the order of 10,000 watts per gram of U-235.

Particularly impressive among ceramic fuels is UO_2 (uranium dioxide). Largely because of its superior dimensional stability, low neutron absorption by oxygen, and chemical inertness in water, much study and developmental technology have been devoted to it. For all practical purposes, UO_2 represents the "backbone" of nuclear ship fuels.

Uranium dioxide typically is fabricated in pellet form: a right cylinder approximately 0.35 inches diameter by 0.50 inches length. In extreme cases, samples have been irradiated to as high as 5% burnup . . . with a dimensional change of only 0.01 inches.† This is an average volumetric change of only 2.5%, which is a phenomenally small amount for such a severe test.

The pellets are generally formed by powder metallurgy techniques involving compaction and sintering (heat treatment to enhance packing density of the fuel particles). The packing density attainable is about

* Ref: "Ceramic Fuel Materials for Nuclear Reactors," *Journal of Metals*, May, 1956, pp. 660 ff.

† Ref: "Power Reactor Technology," *General Nuclear Engineering Corp.*, Dec., 1957 (Vol. 1, No. 1), p. 26.

95% of theoretical density; this minimizes the porosity of the ceramic fuel. The reason for pellet-type construction is that fuel pellets are amenable to mass production methods, with the consequent cost advantages thereof. Exclusive of the cost of fuel material itself, UO_2 pellets can be fabricated for about 25 cents each. But there are many hundreds of thousands of fuel pellets per reactor core.

The undesirable features of ceramic fuels (compared with metallic fuels) are poor thermal conductivity and extreme brittleness. Though dimensionally stable, ceramic fuels crack quite readily when subjected to radiation or to thermal stresses. The consequence is that the fuel must be structurally contained and suitably bonded.

Ceramic fuel jacketing materials are generally the same as for metallic fuels. The cladding thickness is approximately 0.025 inches. Though ceramic materials themselves may be formed to any shape desired, brittleness under irradiation generally limits the fuel elements to simple structural shapes, such as tubes. Hence, the pellet-in-tube construction of Fig. 1-1.

The choice of a UO_2 bonding material is usually helium, though thermal conductivity-wise it is not so good as lead (see Table 3-4). Lead,

Table 3-4. Temperature Drops in UO_2 Bonding Materials

Material	Temp. Drop* °F
Lead	2
Hydrogen	278
Helium (He)	288
Fission Gases (Xe + Kr)	5400
50% He + 50% (Xe + Kr)	900
Steam	1490

Note: Minimum temperature drop is desired.
*In a bonding gap of 0.001 inches
Ref: "Power Reactor Technology", General Nuclear Engineering Corp., Vol. 1, No. 3, June, 1958, p. 31.

however, has an appreciable affinity for the capture of thermal neutrons whereas helium does not. One disadvantage of helium is that when the fission gases xenon and krypton combine with it, the bonding conductivity worsens almost to the point of becoming an insulator. Hydrogen could be used but it reacts with UO_2 to form steam, which also acts as an insulator (see Table 3-4 again).

From the above, it is evident that the interdiffusion of fission gases with the bonding medium is one of the principal limitations of ceramic fuels. Studies have shown that the release of these fission gases is related to the porosity of the ceramic fuel: the less the porosity, the less the fission gas release. Hence, the desire for compaction densities which approach theoretical density, that is, zero porosity. But even if there were

no porosity whatever in new fuel, cracks would develop once the fuel were in a reactor. This follows from the extreme brittleness of ceramic fuels. The net consequence is that, sooner or later, fission gases escape through the cracks to mix with the bonding material, thereby seriously limiting the fuel heat flow.

Temperature-wise, ceramic fuels represent the upper limit that we need to go, insofar as the fuel material itself is concerned. Particularly with UO^2, there exists in the fuel portion of a fuel element enough temperature driving force (5000° F) for all conceivable types of nuclear ship fuels. But all ceramic fuels are plagued with low thermal conductivity brittleness, and poor bondability.

3-6 Solid Dispersion Fuels

To overcome the inadequacies of ceramic fuels, solid dispersion fuels are used. The "dispersion" feature is a ceramic "fissile" material (fissile means fissionable) dispersed in a metallic or ceramic non-fissile matrix. In other words, the dispersion matrix is a material chosen for its desirable properties of thermal conductivity, structural integrity, resistance to corrosion, and bondability to the cladding. By proper choice of materials, we can achieve a near-homogeneous solid mixture of uranium-rich fissile fuel dispersed as "islands" throughout the non-fissile matrix material (see Fig. 3-4).

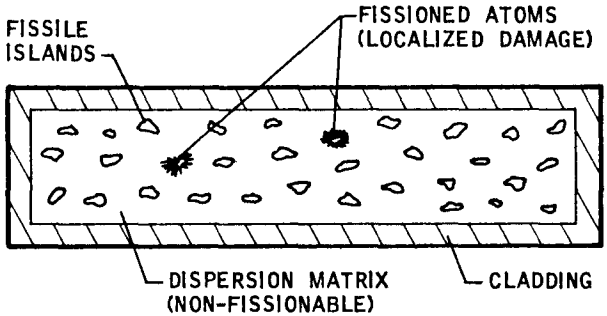

Fig. 3-4 Concept of Fissile "Islands" Dispersed in a Solid Matrix

Solid dispersion fuels offer a number of practical advantages, principal among which are:*

(1) localization of fission damage to the immediate vicinity of each fissile island (the average range of fission fragments is approximately 0.00025 inches), thus leaving a fission-product-free region of matrix material beyond the zone of damage; and

(2) large choice of fuel-matrix combinations without metallurgical interaction, thus furthering the attainment of physical, chemical, and mechanical properties which otherwise could not be attained by metallic or ceramic fuels alone.

* Ref: *Nuclear Fuels,* Gurinsky and Dienes, Van Nostrand, 1956, p. 279.

In concept, at least, the dispersion of fissile fuel into a non-fissile matrix represents the best all-round compromise for solid fuel element design.

In the case of metallic matrices, the matrix material could be metallurgically bonded directly to the fuel element cladding without any other intervening medium. Like metallic base fuels themselves, ceramic fuels dispersed in metals could be shaped to any fuel element geometry desired. The metallic matrix is particularly useful in reactors in which the coolant and moderator are one and the same. Here, the metallic matrix serves as a diluent to uniformly distribute the heat from the spike-like sources of fission (like the metal plate over the burner of a gas cooking stove). Typical metallic matrix materials are aluminum, molybdenum, niobium, and thorium. These matrices, however, are more suitable to low-temperature reactor systems where it is desired to avoid boiling of the coolant and moderator.

In the case of ceramic matrices, typical useful materials are beryllium oxide, silicon carbide, aluminum oxide, and zirconium hydride. These are noteworthy high-temperature materials . . . and they are also excellent neutron moderating materials. Thus, for those reactor systems using a coolant separate from the moderator, ceramic matrices provide the neutron moderation mixed directly with the fuel. This feature is particularly advantageous for high-temperature reactor systems which have not yet come into being for nuclear ships.

Depending on the particular ceramic matrix chosen, previously mentioned cladding materials may or may not be used. Instead, glazed coatings might be used on the fuel elements, in much the same sense as glazed fire-brick in the furnace of an oil-fired boiler.

The principal drawback to solid dispersion fuels is the distortion of the fissile islands. All fabrication techniques involve mechanical and thermal working to bring the final fuel material to the desired size, shape, and density. This squeezes, twists, and elongates the fissile particles so that they are no longer at their optimum shape and dispersion for high fuel burnups. These alterations in the fissile particle structure and in the dispersion uniformity set an upper limit to the potentially realizable advantages from solid dispersion fuels.

3-7 Liquid Metal Fuels

The distortions of the fissile particle shape and the alterations in its dispersion uniformity, inherent in solid dispersion fuels, can be overcome by liquid metal fuels. Here, the fuel consists of a molten metal carrier (e.g., bismuth) in which the fissile material is dispersed either as a solid or as an intermetallic compound. The resulting fuel system is a fluid or slurry which permits the fissile particles to adjust to their own liking, both before and after fission events. The molten metal carrier performs all the functions of a solid metallic matrix previously described.

The most successful liquid carrier metals are bismuth and the eutectics of bismuth-lead and bismuth-lead-tin. All three of these metals have

reasonably low thermal neutron capture characteristics and sufficiently low melting points. Although the solubility of uranium in these metals is not large (e.g., in Bi the U solubility ranges from about 0.5% by weight at 900°F to about 3% at 1200°F), it is sufficient to obtain a feasible reactor system. Thus, fuel preparation simplicity is achieved, and it is possible to operate with high temperatures at low pressures.*

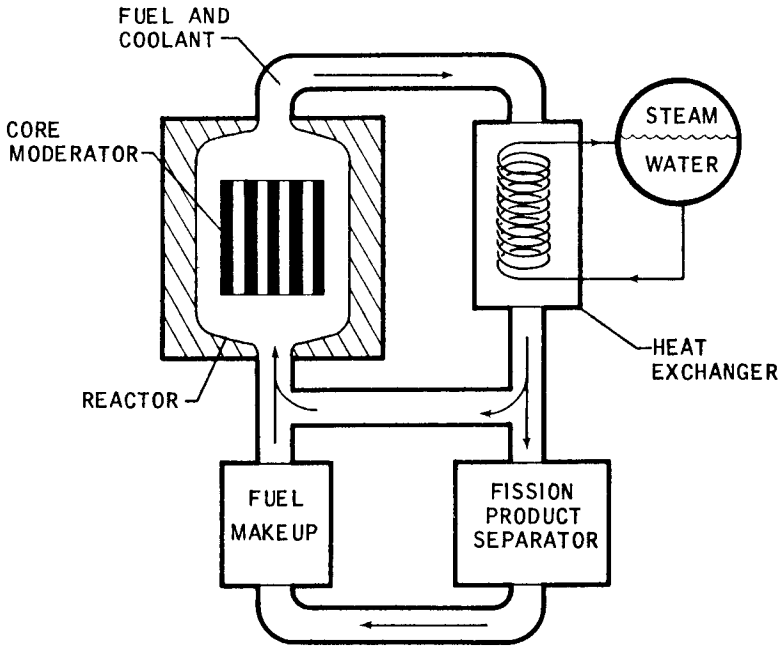

Fig. 3-5 Generalized Concept of a Fluid Fuel Reactor Loop

A distinguishing feature of liquid metal fuels is that the fluid fuel carrier is also the reactor coolant! Both fuel and carrier can be circulated in a continuous loop between the reactor core and its heat exchanger outside. The fuel-coolant is pumped into tubes in the reactor which are interarranged among a moderating material. Circulating the fluid fuel permits external processing of the fuel without reactor shutdown. In other words, fission gases and fission residues can be removed, and new fuel can be added, on a continuing basis (see Fig. 3-5). This, indeed, is a unique feature of nuclear fuel design.

The major problems with liquid metal fuels are dynamic corrosion and mass transfer. The corrosion of molten carrier metals is aggravated by the fuel material. In addition, container materials (i.e., piping, tanks, etc.) readily dissolve in the molten metal on the hot side of the circulating

* Ref: "Liquid Metal Fuel Reactor Systems for Power," *Nucleonics*, July, 1954, pp. 11 ff.

loop, and are deposited on the cold side. The buildup of the corrosion-transfer products quickly plugs up flow channels and elbows on the cold side of the loop, and opens up hundreds of leaks on the hot side. Experiments with inhibitor metals have been made, but no completely satisfactory container materials have resulted. The consequence is that a profusion of liquid metal leaks inevitably develops. These leaks become simultaneously a fire hazard and radiological concern.

While liquid metal fuels probably may never be seriously considered for nuclear ships, such fuels, nevertheless, introduce us to one principle of interest. This principle is the removal of fission gases and residues, and the replacement of fuel, without reactor shutdown. If this concept could be adapted to other fuel designs, fuel burnup would be substantially improved.

3-8 Aqueous Fuels

Aqueous fuels carry the principle of circulating fuels one step further . . . to include the neutron moderating material. Instead of a molten metal fuel carrier, ordinary water is used. This water is both the carrier of the fuel and the moderator of neutrons. The fuel is in the form of a uranyl salt (i.e., uranyl sulphate, nitrate, or phosphate) which is dissolved in the water.* Thus, an aqueous fuel is a three-in-one combination of fuel, coolant, and moderator.† As a result, the physical size of the reactor core can be surprisingly small . . . on the order of a 2-foot diameter sphere, or so.

Like liquid metal fuels, aqueous fuels also circulate between the reactor core and the heat exchanger outside. Accordingly, all of the advantages of liquid metal fuels are retained. In addition, because the water also is the moderator, an inherent self-regulation feature exists. If the moderator gets too hot, it overexpands, and the fission cycle tends to shut itself down. Thus, only a few control rods are required.

The fact that there is no heat-transfer barrier (i.e., no bonding and cladding), and therefore no extraneous neutron absorption materials in the core, means greatly improved nuclear and thermal efficiency. In many ways, aqueous fuels have much in their favor. Reactors using aqueous fuels can be started up quickly and can be shut down quickly. Literally, the fuel can be dumped in, or dumped out . . . with safety.

There are several limitations to aqueous fuels, however. The foremost limitation is the low solubility of the uranyl fuel. Uranyl sulphate (UO_2SO_4), for example, is one of the more soluble fuel salts. At best, its solubility is low, being on the order of a few % U at temperatures up to 650°F. Beyond this temperature, the fuel solubility falls off considerably. This necessitates the use of highly enriched fuels. Furthermore, as the fuel circulates through the heat exchanger outside of the

* Ref: "Aqueous Fuel Systems," *The Reactor Handbook—Vol. II, Engineering,* USAEC-3646, pp. 507 ff.

† Reactors using aqueous fuels are called "homogeneous reactors."

core, it gives up its heat, drops in temperature . . . and drops also in fuel solubility. Consequently, fuel precipitants form undesirably.

A still further limitation is the formation of radiolytic gases. These gases are the decomposition products of the water (i.e., hydrogen and oxygen) when in the reactor core. Radiolytic gases are formed at greater rates than fission gases due to the combined damage to the aqueous fuel from fission fragments, neutrons, and gammas. The formation of free hydrogen and oxygen necessitates special off-gassing, gas recombination, and feedwater make-up equipment.

Corrosion, though not so severe a problem as with liquid metal fuels, is supplemented by a new problem: erosion. Because aqueous fuels are considerably less dense than liquid metal fuels, to extract the same amount of heat from each, aqueous fuels must be pumped at a faster rate. The result: fast-moving fuel particles erode the containment piping, control valves, and pump parts.

From a fuel inventory and external radiation point of view, neither of the two circulating fuels offers advantages. Both require more initial fuel than their non-circulating counterparts, by the amount of fuel outside of the core. Because of fission gases and residues outside of the core, considerable external shielding is required. This is partly offset by the lesser shielding required for the small-sized core.

3-9 Gaseous Fuels

The use of gaseous fuels has not been seriously considered for nuclear reactors largely because of the low density of the fuel gases involved. Though technically feasible, the resulting reactor would be too large. However, as reactor pressurizing and corrosion-protection techniques improve, the use of gaseous fuels may warrant future consideration.

There are two types of gaseous fuels of interest. One involves the suspension of U-particles (e.g., UO_2 powder) in an inert carrier gas (e.g., helium, nitrogen, carbon dioxide);* the other, a uranium compound which is gaseous in its own right, namely UF_6 (uranium hexafluoride).

To utilize these gaseous fuels, let us visualize a cladded element consisting of a porous ceramic moderator with a central hole through it. At one end of the element there is a fuel feed; at the other end, a porous barrier or pebble bed to release the gaseous fission products (see Fig. 3-6). By this arrangement, it is possible to attain the advantages of solid dispersion fuels, plus the fission product removal advantages of fluid fuels. An entire reactor core could be built up from the Fig. 3-6 type of ceramic elements.

The gaseous fuel, however, would not be circulated in the normal sense of a reactor coolant. Instead, the fuel would be fed in at one end of the core and allowed to distribute itself in the ceramic moderators.

* Ref: "Fluidized and Liquid Fuel Reactors with UO_2," *Nucleonics*, Sept., 1954, p. 16.

Being gaseous, the fuel can be pressurized and its flow can be regulated by pneumatic devices which are sensitive to temperature changes in the reactor core. A separate coolant would be passed external to the ceramic moderator elements to remove the fission heat.

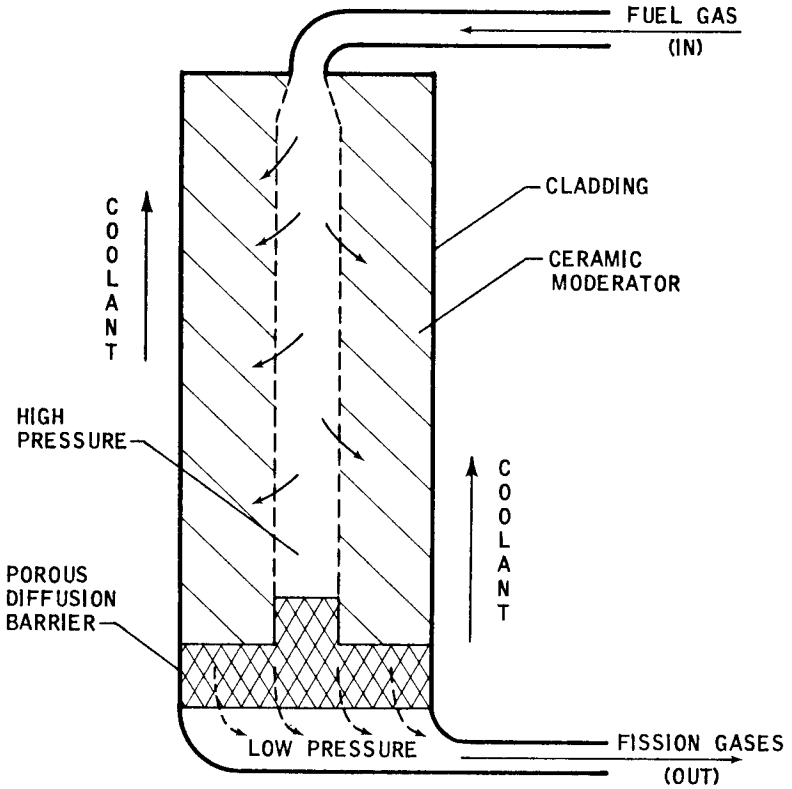

Fig. 3-6 Possible Fuel Element with Gaseous Fuel Feed

The fission gases, being lighter than the fuel gases, would diffuse through the porous barriers at the lower end of the moderator elements (recall the enrichment process in Sec. 3-2). Here, the fission gases can be collected and removed from the core region. The removal of these fission gases during reactor operation would enhance the efficiency of heat extraction from the reactor core (recall Table 3-4 again), and would considerably extend fuel life. It should be stressed, however, that these features represent—for the time being—only "attractive possibilities" for nuclear ships.

SUMMARY

We see that there are three types of parent fissionable fuels possibly suitable for nuclear ships, namely: U-233, U-235, and Pu-239. Of these, only U-235 occurs in nature and requires enrichment from its non-fissile partner, U-238. Enrichment is accomplished by gaseous diffusion techniques, but beyond 90% U-235 enrichment is hardly feasible.

In its basic unfabricated form, metallic uranium has certain capricious characteristics which warrant attention. These are its crystalline phase properties, called "anisotropy." In the alpha phase (to 1220°F), the crystals have three unequal axes; in the beta phase (1220° to 1420°F), the crystals shift to two unequal axes; in the gamma phase (to melting at 2070°F), the crystals revert to three axes . . . but this time of equal lengths. On one hand, this anisotropy is much undesired because of the dimensional instability (growth and damage) that results, but, on the other, it is the very feature that makes uranium such a versatile fuel.

Uranium forms a wide variety of metallic alloys and ceramic compounds, and it can be formed into any fuel shape desired. A particularly noteworthy compound of unusual dimensional stability is UO_2: uranium dioxide. This ceramic fuel provides us with a high-temperature driving force (up to 5000°F) considerably in excess of present-day marine reactor needs.

UO_2 itself, however, has low thermal conductivity and it is brittle. In addition, fission gases find their way through the ceramic pores and cracks to mix with the bonding material and thereby obstruct the flow of heat from the fuel to the coolant. These drawbacks have necessitated the use of matrix materials in which the UO_2 is dispersed as fissile particle "islands." But in the final fabrication into solid fuel elements, unfortunately, mechanical and thermal working techniques squeeze, twist, and elongate the fissile particles. The result is that their optimum shape and dispersion uniformity are altered, and this limits the utility of solid dispersion fuels.

Fluid fuels—liquid metal, aqueous, and gaseous—permit the fissile islands to distribute themselves on their own. The fluid carrier material may be also the reactor coolant, moderator, or both . . . or neither. Fluid fuels can be pumped outside of the reactor core where fission gases and residues can be withdrawn, and new make-up fuel added. Gaseous fuels, conceivably, could be pressurized into a reactor core consisting of ceramic moderator elements. The fission gases could diffuse out through porous barriers similar to those used at the UF_6 enrichment plants. All fluid fuels, however, introduce severe corrosion problems and require added shielding for the fuel equipment external to the core. Fluid fuels have not yet been seriously considered for nuclear ships.

From all of this we can conclude that there is a wide variety of fuel forms available for nuclear ship use. Inasmuch as it is the nuclear fuel that represents the technical shift from conventional marine practice, the type of nuclear fuel used sets the stage for the reactor core and reactor plant design that follows. But, as we shall see, selecting the type of fuel is one thing, and calculating its amount is another.