1. NUCLEAR REACTOR HEAT SOURCES FOR FUTURE POWER GENERATION

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The prospects for the continued increase of nuclear energy sources for the generation of electric power has received much attention. This discussion reviews available data pertaining to this question. It then explores the characteristics of the fast-breeder reactor which seems to offer the greatest potential for the power industry.

Most of the experience at Lewis Research Center has been with reactors for space propulsion or space power systems. These reactors are designed to operate at much higher temperatures and for shorter times than those of commercial power systems, and thus the problems are quite different. Therefore, for this discussion, information was gathered (see bibliography) from the power industry, the AEC, and their contractors and consolidated in light of experience at Lewis. Discussion of this information will serve to establish a basis for the material presented at this conference.

Three general areas of fast-breeder reactors are discussed: (1) the difference between breeder reactors and the power reactors now in use, (2) some of the potential materials problems associated with breeder reactors with major emphasis on the fuel-element materials, and (3) the relative merits of gas and liquid-metal coolants for reactors.

FUTURE POWER REQUIREMENTS AND THE BREEDER REACTOR

The Edison Institute's estimate of the electric power capability for this country up to the year 2000 is shown in figure 1-1. The upper curve indicates that during the next 30 years the electric power demands are expected to increase nearly tenfold, doubling every 10 to 15 years. The lower curve shows the estimated nuclear power capability for this same period and indicates that by the year 2000, 50 to 60 percent of the total electric generating capability will come from nuclear power.

If this prediction of nuclear growth in the power industry is assumed to be correct, a very large increase will occur in the amount of uranium required. Figure 1-2 shows the annual uranium requirements during this period if reactors of the type being used today continue to be built to keep pace with the increasing power demand. The refined uranium ore from which reactor fuel is eventually made is U_3O_8. Within 20 years, the annual uranium ore requirement will have risen to over 100 000 tons per year, nearly 10 times the 1967 production.

For the present type reactors, only a small fraction of natural uranium is actually consumed or burned. Only seven-tenths of 1 percent of the natural uranium is uranium 235 (U^{235}); yet this isotope is the one which produces essentially all the power in present-day reactors. Only about 10 percent of the reactor power comes from high-energy fissioning of U^{238}; most of the uranium, the U^{238} isotope, is not used. A large stockpile of this relatively nonfissionable U^{238} will accumulate to satisfy the U^{235} requirement of these "burner" reactors.

One way in which this growing stockpile of U^{238} can be utilized is by converting it in a reactor to the more fissionable fuel, plutonium 239 (Pu^{239}). This process is shown schematically in figure 1-3 which indicates that U^{238} is classified as a fertile material. When one of these fertile nuclei absorbs a neutron from the reactor, it becomes radioactive U^{239}. This isotope has a half-life of 23 minutes, emits a beta particle, and decays to radioactive neptunium 239. This isotope has a half-life of 2.3 days, gives off a beta particle, and decays to Pu^{239} which is a relatively stable fissionable isotope that can be used as fuel in a reactor.

The total process by which the fertile U^{238} is converted to a fissionable fuel in a reactor is called breeding. The breeding ratio is defined as the ratio of the fissionable material produced to the fissionable material consumed in the reactor. When this ratio is less than 1, as it is in the present-day reactors, there is a net consumption of fuel. However, for reactors in which this ratio is greater than 1, there is actually more fuel produced than consumed so that, at the end of a fuel cycle, a net surplus of fuel will exist. Such a reactor is therefore a true breeder.

The doubling time is defined as the time it takes to double the total fissionable inventory within the reactor and within the fuel reprocessing cycle. For example, a typical breeder reactor could have a core loading of 3000 kilograms of plutonium with another 1000 kilograms being processed out-of-core; the doubling time is therefore the time to produce 4000 kilograms of additional plutonium.

The fact that the breeder reactor can produce plutonium from U^{238} means that the uranium ore can be used more efficiently than with present-day reactors. It also means that the net cost of fuel for the breeder reactor will be less than that for burner reactors since the breeder is, in fact, producing rather than consuming fuel. Table 1-I shows a comparison of the fuel costs for a fossil plant, a nuclear burner, and a nuclear breeder. The fuel costs are expressed as fractions of the total cost of generating power. These relative costs are approximate since they vary with geographic location, type of fuel used, and many other factors. The primary point illustrated is that fuel costs in a breeder reactor are 2 to 3 times less than the fuel costs in nuclear burners or in fossil-fueled plants.

This reduced net fuel cost for a breeder reactor could have an important effect on the overall design philosophy of the breeder system. Since fuel costs are relatively low, thermal efficiency, that is, the efficiency with which fuel is converted to electric power, is less important in a breeder reactor than it is in a nuclear burner or fossil-fueled plant. Therefore, it is possible that, in the design of the breeder reactor, instead of emphasizing thermal efficiency, greater emphasis could be placed on plant cost and system reliability even if a reduction in overall system efficiency were required.

Table 1-II shows a comparison of the breeding characteristics of a present-day burner reactor and a representative breeder reactor. The primary fuel of the burner is U^{235}, while the breeder uses Pu^{239}. Both kinds of reactors have a substantial quantity of the fertile U^{238} present. In the burner reactor, U^{238} is present because the fuel used is only partly enriched in the U^{235} isotope. In the breeder reactor, a large quantity of U^{238} is added to provide a large number of fertile nuclei.

An important difference between the burner and breeder is the average neutron velocity in the two reactors. At present, burner reactors operate most efficiently when neutrons are slowed down to thermal neutron velocities; however, because of the poor nuclear properties of plutonium at thermal neutron velocities, neutrons in a breeder reactor must be prevented from slowing down and kept at high velocity. Reactors which operate on high velocity neutrons are called fast reactors.

Typical breeding ratios for these two types of reactors are 0.6 for the burner and 1.43 for the breeder. The doubling time for such a breeder can be from 8 to 12 years. With this doubling time, the breeder reactors could readily keep pace with the predicted electrical growth of the country.

BURNER AND BREEDER REACTOR CHARACTERISTICS

Today's burner reactors are divided into several classes. Two of the most well known of these are the boiling-water reactor and the pressurized-water reactor. Either type could be used to typify today's reactor systems.

A schematic drawing of a pressurized-water-reactor system is shown in figure 1-4. This plant is composed of a two-loop system: a reactor loop in which nuclear fuel produces thermal energy, and a power-generation loop in which thermal energy is converted to electrical energy. The energy conversion loop is discussed in detail in the papers on Brayton cycle and Rankine cycle systems. In the reactor loop, pressurized-water coolant enters the reactor at about 550° F, is heated to about 600° F, produces 500° F steam in the steam generator, and is pumped back into the reactor. The net efficiency of the overall system is about 30 percent.

The breeder reactor system and its operating goals, as presently proposed by the nuclear power industry, are shown in figure 1-5. Schematically, this system resembles the pressurized-water system except for an extra loop consisting of a heat exchanger and a pump. Besides the added components, there are some other significant differences. The reactor loop is sodium cooled instead of water cooled. The reason for the change is that water is too good a moderator. Maintaining the high neutron velocities required for an effective breeder makes replacing the water coolant with sodium desirable. Other coolants such as helium and steam are also being considered as fast-breeder coolant candidates, but most of today's emphasis is on liquid-metal cooled systems. In the reactor loop, the sodium enters the reactor at 900° F, is heated to about 1150° F, passes through the heat exchanger where it heats the sodium in the intermediate loop, and is again pumped into the reactor.

The sodium in the intermediate loop enters the heat exchanger, is heated to 1075° F by the sodium in the reactor loop, enters the steam generator where it produces 1000° F steam, and is again pumped back into the heat exchanger. The sodium-cooled intermediate loop was inserted for safety. The reactor loop, which is highly radioactive, can be located within the containment chamber. However, because of the possibility of sodium-water fires, it is desirable to keep the water loop out of the containment chamber. There is still the possibility of a sodium-water fire in the steam generator, but this could be more easily extinguished since it would be located outside the containment chamber.

It should be noted that the outlet coolant temperature in the breeder reactor is 500° F higher than in the pressurized-water reactor in order to produce 1000° F steam for the turbine. The net efficiency in the breeder reactor is 40 percent compared with about 30 percent in the pressurized-water reactor. The high coolant temperature has no effect on the breeding capability of the reactor. As far as reactor performance and nuclear characteristics are concerned, the reactor would be just as effective a breeder at 600° F as it is at 1150° F.

The specific power at which the reactor is operated is among the operating conditions of the breeder which are important to consider because of their influence on performance. Figure 1-6 shows the effect of power density on the breeding characteristics of a typical reactor. The breeding ratio is plotted against the doubling time in years. The parameter shown in the figure is the specific power of the reactor core for 100, 300, and 500 kilowatts (thermal) per liter of core. The shaded band represents the previously discussed 10- to 15-year period during which the electrical power requirements of the country will double. In order for breeder reactors to keep pace with this growth, the doubling time of breeder reactors should be equal to or less than this 10- to 15-year period. For a given breeding ratio, it is necessary to operate the reactor at as high a specific power level as possible to minimize the doubling time. For a breeding ratio of 1.43, the specific power of the core must be about 300 kilowatts (thermal) per liter in order to lie in the shaded band. This specific power represents a large increase over present-day burner reactors which operate at power densities of 60 to 90 kilowatts per liter.

This necessity for high power density has some important effects on the operating requirements of materials for fast breeders. The fuel burnup required for a breeder fuel pin would be approximately 10 atom percent of the fuel, whereas present reactors have burnups of the order of 2 to 4 percent. Such high burnups are required in breeder reactors because of the direct relation to high power density and to the need for keeping the refueling interval to about a year. If the fuel burnup were limited to 2 to 4 percent as in the present reactor, the higher specific power associated with the breeder would mean that some of the fuel pins would need to be changed every month or two. This refueling procedure would result in a large percentage of down time for such a system and would reduce the load factor of the plant considerably.

Another important result of the higher specific power of the breeder is the level of neutron exposure which the cladding of the fuel pin must withstand. In the present systems, the cladding receives a total fast flux neutron exposure of about 10^{22} neutrons per square centimeter. This exposure would be increased an order of magnitude to 10^{23} neutrons per square centimeter in the breeder reactor. As shown later, this increase in neutron exposure seriously affects the performance of the cladding material.

Development of the proposed liquid-metal-cooled fast-breeder reactor appears to be much more difficult than was the development of the present-day burner reactors because first, the specific power at which the breeder fuel element must operate is increased; second, the operating temperature is increased; and third, the coolant is to be changed from water to sodium.

MATERIALS PROBLEMS IN BREEDER REACTORS

An examination of the potential materials problem areas can be made by considering the fuel element and its basic component, the fuel pin. A typical breeder reactor fuel pin is shown in figure 1-7. It is made of stainless steel, is 1/4 inch in diameter and 8 feet long, and has a wall thickness of about 15 mils. Oxide fuel pellets are placed in the tube, which is then capped at both ends. The figure shows that there are two void regions in the pin: one, an annular gap between the fuel and clad; the second, an axial void region above the fuel. The annular gap allows for thermal expansion and fuel swelling; the axial void is used to store fission gases which escape from the fuel.

A breeder reactor contains about 5 times as much fissionable fuel per unit volume of core as a burner reactor. The breeder is therefore very sensitive to small movements of the fuel pins. The possibility of a power excursion, which could result in severe damage, is avoided by preventing the fuel pins from shifting during operation. This is accomplished by bundling a hundred or more fuel pins tightly together into a fuel-element assembly, as shown in figure 1-8. The fuel pins can be spaced by a wire wound spirally around the pin, as shown in the figure, or by some type of ferrule located between the pins. These spacing devices increase pressure losses and can cause "hot spots" which would result in local increases in fuel-element temperature. These factors make design of the fuel elements a major problem.

Numerous fuel-element assemblies are clustered together to form the reactor core. Figure 1-9 is a simplified schematic drawing of the breeder reactor. The reactor consists of a central-core region containing a mixture of U^{238} and the Pu^{239} fissionable fuel; this central section is surrounded by a blanket region containing U^{238}. Breeding of new Pu^{239} fuel takes place in the U^{238} both in the core and in the surrounding blanket. The heat generated in the fuel elements is transferred to the sodium which is pumped through the core.

The number of fuel pins required to dissipate the heat from a reactor is given in figure 1-10. The required number of fuel pins is plotted against reactor diameter for a 1000-megawatt-electric reactor. Curves are shown for three pin diameters, 1/4, 3/8, and 1/2 inch. There are about 200 000 1/4-inch fuel pins in the breeder reactor - about 4 or 5 times the number used in the present burner reactor. A considerable portion of the increased number comes from using the small-diameter fuel pins proposed for the breeder reactor. The justification for the small-diameter pin is discussed later.

For the breeder reactor, the large number of fuel pins, the high burnup, the high neutron exposures, and the high temperature require high performance materials. The fuel-pin materials that are in the most advanced stage of development are oxide fuel and stainless-steel cladding. The oxide fuel consists of a mixture of uranium oxide and plutonium oxide. The stainless steels being considered are the 300 series, austenitic type, particularly 304 and 316 stainless steel.

However, some alternate fuels and cladding materials also are being considered. The alternate fuels are a mixture of uranium and plutonium carbides and a mixture of uranium and plutonium nitrides. Uranium-plutonium metal fuels also are a possibility but are less promising than the ceramic fuels and are not discussed further. The alternate cladding materials (nickel alloys, vanadium alloys, and the refractory metals, including niobium alloys and molybdenum alloys) offer higher strength than stainless steels if higher strength is required.

Fuels

Three properties of a fuel are most important in determining its suitability for use in a breeder reactor: thermal conductivity, swelling under irradiation, and compatibility with cladding materials.

The thermal conductivity affects the maximum fuel temperature in the fuel pin for given operating conditions of heat flux and coolant temperatures. Figure 1-11 is a plot of maximum fuel temperature against thermal conductivity for typical breeder reactors being considered. The curves are calculated values for 1/4-, 3/8-, and 1/2-inch-diameter pins.

The maximum allowable fuel temperature is set at about 4000° F to prevent the fuel from melting. With the indicated thermal conductivity of oxide fuel, a 1/4-inch-diameter fuel pin is the maximum allowable size.

Both carbide and nitride fuels have considerably higher thermal conductivities, as indicated in figure 1-11. Therefore, pins containing these fuels could be as large as 1/2 inch in diameter without exceeding the fuel temperature limit of 4000° F. Earlier, mention was made of 200 000 1/4-inch oxide fuel pins required in large breeder reactors. Use of 1/2-inch carbide or nitride fuel pins could reduce this number by a factor of 4 and thereby greatly reduce the fuel fabrication costs. If preferred, the higher thermal conductivity of the carbide and nitride fuels could be used to reduce the fuel temperature. Keeping the diameter of the fuel pins at 1/4 inch, for example, would limit the maximum temperature of a carbide or nitride fuel pin to less than 2500° F.

Fuel swelling under irradiation is a particularly severe problem with breeder-reactor fuel pins because of the high fuel burnups required. That nuclear fuels swell under irradiation because of the generation of fission products is well known. This fuel swelling must be allowed for in fuel-pin designs to prevent external dimensional changes in the fuel pin which could restrict cooling flow and could lead to pin failure.

There are many factors that influence the rate of fuel swelling. These factors are not fully understood for breeder-reactor fuels because only a few irradiation tests have been completed on the mixed uranium-plutonium ceramic fuels. Therefore, existing swelling data on uranium-bearing fuels (particularly UO_2) must be heavily relied on in predicting fuel swelling for the mixed U-Pu fuels.

The seriousness of the swelling problem is illustrated in figure 1-12. Predicted swelling of fully dense uranium dioxide in the temperature range of interest is plotted as a function of fuel burnup under conditions where no fission gas can be released from the fuel. As the figure shows, breeder-reactor fuels which are subjected to about 10 percent burnup may swell as much as 17 percent in volume. The swelling would be much less in burner reactors - about 5 percent in volume because of the lower burnup of about 3 percent.

At the reactor operating temperatures, some of the fission products that cause this swelling are solid and others are gaseous. But the gaseous products (such as krypton and xenon) cause most of the problem. Thus, release of these fission gases from the fuels could greatly reduce the amount of swelling. At the same time, high stresses in the cladding of the fuel pin could be prevented by providing enough void space in the pin so that the released fission gases can be collected without building up high pressures.

Most of the fission gases are released from oxide fuel; very little is released from carbide fuel. Measurements have been made of fission gases released from uranium dioxide and uranium carbide test specimens that were irradiated to about 5 percent burnup at surface temperatures of 1100° F. The uranium dioxide released nearly 90 percent of its fission gases, but the carbide released only about 5 percent. This large difference is related to the difference in thermal conductivities of the fuels. The poorer thermal conductivity of the oxide resulted in much higher central temperatures and a larger temperature gradient in the oxide specimens than in the carbide specimens of the same diameter. These temperature conditions increased the mobility of the fission gases in the oxide fuel and allowed them to be released into the void space.

The interplay amoung fuel conductivity, fission gas release, and fuel swelling introduces a high degree of complexity into fuel behavior and selection. A fuel with low thermal conductivity will tend to release a high percentage of fission gas and will be limited to small-diameter pins. Larger diameter pins can be used with high conductivity fuels to reach comparable temperatures, but temperature gradients will be lower and fission gas release may not be as great. This would perhaps still leave a swelling problem.

Differences in fission gas release and swelling behavior between the low- and high-conductivity fuels will affect the fuel-pin design. They will require that somewhat different design approaches be used for oxide fuels than for carbide or nitride fuels. Because the carbide or nitride fuels will retain more of their fission gases, they will swell more than oxide fuels. Therefore, some void space must be provided within the fuel pin to allow for the swelling without straining the cladding. Void space is also required in oxide fuel pins to allow release and collection of the fission gases at low pressures. However, the locations of the required void spaces differ, as shown in figure 1-13. Schematic illustrations are shown of the cross sections of parts of pins containing oxide fuels and carbide or nitride fuels. The dots in the fuel area represent porosity initially fabricated into the fuel and the fission gas bubbles.

In the case of the oxide, the high temperature and the large temperature gradient cause porosity in the fuel to migrate to the hottest zone at the center and form a central void which usually extends the full length of the pin. The fission gases also migrate to the hottest region and into this central void. Once there, the fission gases can move axially into collection chambers provided at the ends of the fuel pin. These void spaces at the ends of the pins also provide space for some axial swelling of the fuel which occurs because of the solid fission products and residual unreleased fission gas. This design approach was used recently for a test pin containing U-Pu oxide fuel that was irradiated to about 12 percent burnup. About 83 percent of the fission gases were released to the collection chamber and the oxide fuel swelled about 7 percent axially, but the most important observation was that the external diameter of the fuel pin did not change.

For carbide or nitride fuels, which do not release as much of their fission gases, an annular void space between the fuel and cladding appears to be necessary, as shown in figure 1-13. This space should allow swelling of the fuel without imposing high stresses on the cladding. However, this annular void (or gap) introduces a problem in heat conduction across the gap. This problem may be remedied by using either high-pressure helium or liquid sodium to increase the thermal conduction across the gap. This design approach currently is being tested in several irradiation specimens containing mixed U-Pu carbide fuels. Only a few specimens have been examined so far, and these were irradiated to fuel burnups of less than 5 percent. However, the results appear encouraging in that no appreciable increase in pin diameter was found.

The third area of concern with respect to fuel selection is its compatibility with pin cladding; that is, the fuel and the cladding should not react excessively with each other. Compatibility data available in the literature have been evaluated and the results show that there is good compatibility of the oxide, carbide, and nitride fuels with the various cladding materials considered. However, this is so only if the stoichiometry of the fuel is precisely controlled, because changes in fuel composition can lead to fuel-clad reactions. The stoichiometric composition of UO_2, for example, is exactly two atoms of oxygen for each atom of uranium.

Stoichiometric effects seem to be more important for carbide and nitride fuels than for the oxide fuel. For example, figure 1-14 indicates the effect of fuel stoichiometry on compatibility with cladding for carbide fuels of uranium and plutonium.

The depth of attack of the fuel on the cladding is shown to vary with the carbon content of the fuel. The stoichiometric mixture is that at the minimum. Lowering the carbon content leads to the formation of free uranium and free plutonium which can alloy with the cladding. Raising the carbon content can lead to carburization of the cladding. Therefore, the composition of carbide fuel must be carefully controlled to maintain near-stoichiometric composition to minimize the depth of attack.

In summary of the fuels discussion, oxide, carbide, and nitride all have merit as breeder-reactor fuels. The oxide fuel is the prime candidate because of the experience with uranium dioxide and because the compatibility of oxide with clad materials is not sensitive to small changes in fuel stoichiometry. Carbide and nitride have better thermal conductivity and hence offer the potential of using larger and thus fewer fuel pins. However, the choice of fuel will ultimately be made on the swelling characteristics and the ability to accommodate the swelling by adequate fuel-pin design. This type of information is being obtained in extensive fuel irradiation tests throughout the country.

Cladding

The fuels, however, are only one part of the fuel element, the other aspect is the cladding material. As mentioned earlier, austenitic stainless steels are currently considered the prime candidates for use as fuel-pin claddings. However, alloys of nickel, vanadium, niobium, and molybdenum are also being studied for this use.

In selecting a cladding material for fuel pins, many factors must be considered, but three are most important.

(1) The first of these is material strength, particularly its long-term creep strength at the maximum operating temperatures. The cladding must be able to withstand the high internal stresses generated by the swelling fuel along with any fission gases released by the fuel. These stresses must be borne by the cladding without any appreciable strain.

(2) The effects of the cladding material on the operating costs of the reactor must be considered. Not only the cost of the material itself, but also its effect on the breeding ratio and doubling time of the reactor must be considered.

(3) The third major factor concerns the effect of irradiation on cladding materials. In fast-breeder reactors, cladding materials will be subjected to very high neutron exposures because of the high fuel burnup levels and the long operating times. Thus, neutron exposures of more than 10^{23} neutrons per square centi-

meter can be expected. Under these high neutron exposures, the claddings must remain ductile, must maintain good strength, and also must maintain dimensional stability.

Figure 1-15 compares the long-term creep strengths of alloys typical of each class of material. The stress to produce 1 percent strain in 10 000 hours is plotted as a function of temperature for type 316 stainless steel and for selected alloys of nickel, niobium, vanadium, and molybdenum. At 1200° F, stainless steel has a strength of about 10 000 psi, which is probably adequate for use as fuel-pin cladding at this temperature. However, hot spots in the fuel pins could push the cladding temperatures to 1400° F or higher. And, at these temperatures, 316 stainless steel has very little strength. Thus, it would be desirable to lower the operating temperatures of the fuel pins if stainless-steel claddings are used. Alternatively, the higher strength alloys shown in figure 1-15 could be used at higher operating temperatures.

Using these higher strength alloys can increase the cost of the power produced in two ways; the initial cost of the material and its fabrication, and the breeding ratio. Both these factors are shown in table 1-III. The estimate of the relative costs of tubing for typical alloys of each group is listed. For this comparison, the current cost of stainless-steel tubing was used as a base line, and the future costs of the less common alloys were predicted by assuming a normal advance in technology and experience with these alloys. These estimates show that nickel alloys, such as the Hastelloys, should be comparable in price with stainless steel. However, the other alloys are much more expensive primarily because of their scarcer supply and the more difficult fabrication processes involved.

Consideration must be given to the effects of these materials on the reactor breeding ratio, which can be greatly affected by changes in the cladding. The highest breeding ratios can be attained with either stainless steel or vanadium alloys, the next highest with nickel alloys. But the use of the other alloys involves a penalty in breeding ratio. The magnitude of this penalty is important in terms of increased doubling time for the fuel cycle. Because of the slightly lower breeding ratio for nickel alloys, the fuel doubling time increases from 12 to 15 years. And the very low breeding ratios for the refractory alloys of niobium and molybdenum will cause the fuel doubling times to increase, by a factor of 3, to about 35 years.

It is apparent that stainless steels are the most attractive cladding materials from the cost viewpoint provided that the fuel element can be limited to low enough temperatures. If higher strengths are required, fuel cycle costs will limit the practical alternatives to alloys of nickel or vanadium.

The effects of irradiation on these clad materials must also be considered. Results of in-pile testing are beginning to show that there is a major problem associated with the irradiation of stainless steels, for they become seriously embrittled under neutron bombardment. This is illustrated in figure 1-16 where ductility (expressed as percent uniform elongation) is plotted against test temperature for 304 stainless steel. The upper curve shows the typical behavior of unirradiated 300 series stainless steel; the ductility decreases at temperatures above 1000° F but it remains at tolerable levels at temperatures up to 1500° F. However, material which was irradiated at 1000° F to a neutron exposure of 1.72×10^{22} neutrons per square centimeter has lower ductility even at low temperatures and drops to intolerably low levels at higher temperatures. Furthermore, the embrittling effects increase with higher neutron exposures, and exposures nearly 10 times greater than those seen in this test are expected in fast-breeder reactors.

In addition to this problem of severe embrittlement of the cladding, irradiation to higher neutron exposures can also cause swelling of stainless steel. This problem is indicated in figure 1-17 which shows volumetric swelling measured in 316 stainless steel as a function of neutron exposure in the temperature range of 680° to 1080° F. This range is below the 1200° F reference temperature, but data are not available for 1200° F. There was very little growth at neutron exposures up to about 10^{22} neutrons per square centimeter. But in the extra decade of exposures that are required for materials in fast-breeder reactors, stainless steel actually increased nearly 10 percent in volume. This amount of swelling can cause serious problems in fuel-element claddings. Not only is the strength decreased, but gross dimensional changes can occur amounting to a 3-inch increase in the length of an 8-foot fuel pin and a diameter change of as much as 8 mils for a 1/4-inch-diameter fuel pin. However, there is still hope that by modifying the stainless steel the swelling problem can be eliminated. Some basis for this hope is shown by the triangular data points in figure 1-17. These particular points were obtained on 20-percent cold-worked stainless steel and show much lower swelling than annealed stainless steel for which the curve is shown. Although the data are limited, behavior of this type does indicate that there may be ways of reducing the irradiation-induced swelling of stainless steel.

Thus, it is apparent that there are some serious problems associated with the use of stainless steels as fuel-pin claddings in fast-breeder reactors. Many studies to overcome these problems are currently underway in various laboratories, particularly by the AEC and its contractors. As a result, researchers now have a fairly good knowledge of what causes embrittlement and swelling. The embrittlement and swelling at lower temperatures appear to be the result of clustering of irradiation-induced vacancies in the metal lattice. This microscopic vacancy-clustering eventually leads to macroscopic voids and the resultant swelling. This might explain the smaller swelling of cold-worked stainless steel. The microstructure of cold-worked stainless may slow the clustering of vacancies. The problem is compounded by the formation of helium bubbles at higher temperatures. The helium originates from the irradiation of some impurities in the stainless steel but the helium atoms produced do not collect to form bubbles until higher temperatures are reached.

Now that the causes of swelling and embrittlement under irradiation are being better defined, there is hope that methods of overcoming these effects can be found for stainless steels. However, because of these irradiation effects, more consideration is being given to alternate nickel and vanadium alloys as cladding materials. Unfortunately, the irradiation problems found in stainless steels are also being found with nickel alloys. It has been suggested that the similar behavior of both materials is a result of their having face-centered-cubic crystal structures. The vanadium alloys, on the other hand, have a body-centered-cubic crystal structure, and the current theory indicates that this structure should be more resistant to irradiation embrittlement. To date, only a few irradiation tests have been run on vanadium alloys, but the limited data do support the theory. It is observed that vanadium alloys withstand irradiation much better than stainless steels, so that work on vanadium alloys is being accelerated.

In this discussion on cladding materials, austenitic stainless steels are considered the prime candidates because of their low cost and the wide-spread experience with them. However, the strength of these alloys will limit their operating temperature, and they may severely embrittle and swell under irradiation. Thus, either an improved stainless steel must be developed or an alternate material, such as a vanadium alloy, must be used.

Liquid-Metal Corrosion

Another potentially serious materials problem is associated with the use of liquid sodium, which can be very corrosive if it is not used carefully. This corrosion can be detrimental to the fuel-element claddings, the reactor structural members, and coolant loop components, including piping, heat exchangers, and pumps. The presence of impurities like oxygen or carbon or the presence of two or more different metals in the same system can have large effects on corrosion rates.

Ideally, sodium corrosion should not be much of a problem in a single metal system at the planned reactor operating temperatures of about 1200° F. Figure 1-18 shows estimates of the upper temperature limits at which various alloys might be compatible with sodium. These estimates are based on laboratory-scale tests in which operating conditions are very closely controlled. This figure indicates that sodium corrosion should not be a problem with stainless steel at temperatures below about 1500° F.

However, these limits are very difficult to attain in practical large-scale systems because there are many factors which increase the corrosive effects of sodium. The prime factor is the amount of oxygen impurity in both the sodium and the container materials. The effect of oxygen on the corrosion rate of sodium on stainless steel as a function of temperature is shown in figure 1-19. The three curves indicate probable corrosion rates for sodium containing about 10, 50, and 1000 ppm oxygen. The 10- and 50-ppm curves are based on data obtained in various types of tests. The 1000 ppm curve is estimated. It is apparent from these curves that increasing oxygen content greatly decreases the useful temperature for the system. For example, a corrosion rate of about 1 mil per year is considered the maximum tolerable level for fuel-pin claddings. At this rate, oxygen levels of less than 10 ppm are needed to prevent severe corrosion of fuel-pin hot spots which may be at temperatures of about 1400° F.

This low level of oxygen puts heavy demands on the reactor and system designers, builders, and operators. Therefore,

(1) Reactor designers must insist on procurement of materials with the lowest oxygen content practically attainable.

(2) Extra cautious handling techniques must be used during construction to prevent contamination of either the sodium or the container materials. For example, welding procedures must not introduce impurities like oxygen or carbon.

(3) Gradual oxygen contamination of the sodium during the long reactor operating times will require continual cleanup of the sodium during operation. This requirement leads to complications in the coolant loops because small bypass loops must be added for continual oxygen analysis and purification. Developing components for purification and analysis becomes a major problem. These components must reliably maintain the oxygen content in the sodium below 10 ppm to provide good corrosion resistance.

As mentioned earlier, the presence of dissimilar metals in contact with sodium can aggravate corrosion problems. Dissimilar metals are usually used in coolant loops because of strength, cost, or other factors. However, use of dissimilar metals can lead to transfer of certain alloying elements from one material to another through the sodium. This mass transfer can lead to compatibility problems or to changes in the properties of the materials.

The microstructure of a molybdenum specimen, heated in a sodium-filled nickel container for 100 hours at about 1800° F, is shown in figure 1-20. As a result of this test, a nickel-molybdenum (Ni-Mo) alloy layer about 2 mils thick was formed on the surface of the molybdenum. The Vickers hardness number test indentations indicate that the Ni-Mo layer is much harder than the molybdenum substrate. This hard surface layer could decrease the ductility of the molybdenum and could render it useless for further operation under high strain conditions.

The net effect of using dissimilar metals in sodium loops is to lower the maximum temperature at which they can be used. Figure 1-21 shows the revised estimates of the compatibility limits of various metals in high-purity sodium. The lower, darker portions of the bars indicate the range in which we think the metals should be usable in dissimilar metal systems, for example, in a system where stainless steel is used with a ferritic steel or where a nickel alloy is used with stainless steel. These estimates indicate that stainless steels or nickel alloys used at temperatures of about 1200° F in fast-breeder reactors would be operating in a region near their compatibility limits if other metals are present. Then a potentially dangerous situation would arise that would not allow for any hot spots in reactors nor changes in other effects which could also increase the corrosion rates.

Therefore, sodium-cooled systems that use stainless steel or nickel-alloy components should be limited to operating temperature of 1000° F or lower to ensure hot-spot temperatures less than 1200° F. Since sodium corrosion may limit the operating temperatures of breeder reactors, the use of a less corrosive coolant should also be considered.

BREEDER REACTOR COOLANTS

The choice of coolant for fast-breeder reactors is an area which has received much attention. Gas, steam, and liquid metal have all been considered as possibilities, although much effort is presently being directed toward the use of liquid metals. Some factors which must be evaluated in selecting a coolant are heat-transfer and pumping-power characteristics, the effect on the breeding ratio, and the safety aspects of a loss-of-cooling accident. Several other factors should also be considered.

Although steam, gas, and liquid metal are being considered as coolants, the potential breeding ratio with steam is lower than that of the other coolants. It is somewhere in the area of 1.1 to 1.2 so that the doubling time for a steam-cooled breeder reactor would be well over 25 years. Therefore, this discussion is limited to the two most promising coolants for fast-breeder reactors, sodium and helium. Sodium, like most liquid metals, has excellent heat-transfer properties, but potentially serious corrosive characteristics. Therefore, helium must be considered as a candidate reactor coolant even though it is a poorer heat-transfer medium than sodium. Comparable heat transfer can be obtained with helium by using high pressure and high velocity. High velocity results in large pressure losses and increases the pumping power requirements of the system.

This point can be illustrated by comparison of two identical reactors operating at the same power level; one reactor is cooled with sodium and the other with helium. Figure 1-22 shows how the surface temperature of the fuel pins will vary in these two reactors if the same inlet and exit coolant temperatures are maintained. While such a comparison may not represent the optimum conditions for either coolant, it does serve to demonstrate the effects. In this figure, the surface temperature is plotted along the length of the fuel pins with the flow of coolant from the inlet at the left to the outlet at the right. The lower curve is for the system where sodium was used as the coolant; the upper curve is for the helium-cooled system.

The maximum cladding temperature of the helium-cooled system is about 200° F higher than the sodium-cooled reactor. In addition, the relative pumping power across the reactor core is twice as great for the helium-cooled system. However, since the pumping power requirement for a typical sodium-cooled reactor loop is less than 1 percent of the power output of the plant, even a factor of 2 on pumping requirement may be an acceptable value.

There are several ways of improving the heat-transfer characteristics of a gas-cooled reactor. One is to roughen the surface of the fuel pin in the region of high clad temperature. This will improve the heat transfer and reduce the surface temperature of the fuel pin. Surface roughening is only done to the portion of the pin where the temperature is high because roughening the fuel pin increases the pressure drop and pumping power requirements. In the example shown here (fig. 1-22), the relative pumping power across the core has increased by about 50 percent as the result of roughening only the hot portion of the fuel element. However, the maximum surface temperature has been reduced to less than 100° F over that of the sodium-cooled system.

Achieving even this moderately good heat transfer required the use of relatively high-pressure helium (1250 psi). This will introduce some additional design problems for the helium-cooled system, but these must be weighed against the problems involved in using a potentially highly corrosive liquid metal.

Another problem is that of the hot spots associated with the fuel pin spacer, which was mentioned earlier. Because of the power heat-transfer characteristics of helium, this problem would be more severe than in the sodium-cooled reactor.

It can be concluded that the use of helium instead of sodium probably will result in an increase in the surface temperature of the fuel pin. In addition, the pumping power required for a helium system will be higher than for the sodium system. Also, helium must operate at a pressure which is considerably higher than that required for sodium. The magnitude of these problems does not, however, rule out the use of helium as a possible fast-reactor coolant.

The use of helium as a fast-breeder coolant enhances the breeding characteristics of the reactor, as shown in table 1-IV. Because helium is much more transparent to high-velocity neutrons than to sodium, there are less-parasitic captures in the helium. For every 1000 neutrons in a sodium-cooled reactor, 30 are absorbed by the coolant. In a helium-cooled system, less than 1 out of every 1000 neutrons would be absorbed in the coolant. This fact, coupled with other neutronic effects of helium relative to sodium, increases the breeding ratio from 1.43 for sodium to about 1.55 for a comparable helium system.

This increase in breeding ratio could be used to offset the poorer heat-transfer-pumping characteristics of the helium-cooled system. For example, the helium-cooled reactor could operate at a power density 20 percent lower or about 240 kilowatts per liter compared with 300 kilowatts per liter in the sodium-cooled reactor and still have the same doubling time.

The reason that the safety aspects of a loss-of-coolant flow must be considered in comparing coolants can be explained with the use of figure 1-23 showing a normal reactor power decay following shutdown. For a considerable period of time after initiation of shutdown, the actual power generated in the reactor is appreciable. After a few hours, for example, about one-tenth of 1 percent of the original power is still being produced. Because the thermal power required for a 1000-megawatt-electric system is 2500 megawatts, about 2.5 megawatts of thermal power still are being generated 2 hours after shutdown.

In the sodium-cooled reactor shown in figure 1-9, even the rupture of a coolant pipe would still leave the reactor submersed in coolant, and heat could be dissipated by conduction and natural convection of the coolant.

In a high-pressure gas-cooled system, however, a break in a line would result in loss of coolant in a very short time and there would be no effective means of removing the heat from the reactor core. One could consider using a reserve gas coolant which would be pumped from an external storage container through the core for as long as necessary. The amount of gas required for such a procedure makes this method of core protection unattractive. A more likely approach would be to spray the core with water. However, to assure that the introduction of such an effective nuclear moderator into the core did not result in a nuclear excursion, the water would have to contain a soluble neutron absorber.

The safety aspect of a loss-of-coolant flow seems to be a major problem with the use of a gas coolant. A system would have to be designed which either precluded the possibility of such an accident or was protected in the event of such an occurrence.

Several other coolant considerations can be mentioned. The use of an inert gas elimenates the fire hazards associated with the use of sodium. Second, helium does not become radioactive as sodium does. The high level of radioactivity of sodium may require as much as a 10-day waiting period after shutdown before access to the containment area can be made. There is more operating experience with sodium than there is with gas for cooling fast reactors. However, if experience with all types of reactors, heat-transfer loops, and other systems were included, a far greater amount is available on gas systems than on liquid-metal systems.

Helium could be an attractive alternate for a fast-reactor coolant and offers some real advantages. If helium were used, it could be used in a one-loop system transferring heat directly from the reactor to the steam system. This would eliminate the intermediate loop required by a sodium system because the isolation requirement for the intermediate loop in the sodium system is eliminated. However, the single helium loop could become contaminated by fission products leaking from ruptured fuel pins which would pose a radiation hazard around components of the steam system. Finally, the consequences of a loss-of-helium-flow accident pose the problem of how to ensure removal of afterheat from the core to prevent excessive overtemperatures in the fuel elements.

If the hazard of the loss-of-coolant-flow accident in the gas system is really an insurmountable problem, helium could be used in another way. It could be substituted for sodium as the coolant in the intermediate loop (fig. 1-24). Then, the possibility of any reactions between the sodium and the water would be eliminated because they would be physically separated by the inert loop. Also, many of the problems which result from leaks in the sodium steam generator would be solved. Of course, the use of a gas coolant instead of a liquid metal would probably increase the size of the heat exchanger and steam generator and, as in the reactor, would increase the pumping power requirement of the system. However, the use of helium in the intermediate loop does offer some potential advantages.

CONCLUDING REMARKS

This discussion has shown that many of the operating conditions of fast-breeder reactors are much more severe than those for current burner reactors. Therefore, major advances must be made before fast-breeder reactors become a commercial reality. For breeder reactors to be economically competitive, they must

(1) operate at much higher power densities, 300 kilowatts per liter instead of 60 to 90; (2) require higher levels of fuel burnup, 10 percent rather than 2 to 4 percent; and (3) withstand higher neutron exposure, 10^{23} instead of 10^{22} neutrons per square centimeter. These factors make fuel-element development for breeder reactors a more difficult task than for burner reactors.

Because of the large fuel swelling associated with the high levels of fuel burnup, the best choice of fuel material is not clear. More data on fuel swelling of oxide, carbide, and nitride fuels is needed before the decision can be made.

The additional neutron exposure encountered in breeder reactors causes problems of swelling and loss of ductility in austenitic stainless steel. Thus, more radiation-resistant stainless steel must be developed or a more expensive cladding material such as vanadium alloys must be considered.

The use of sodium as the coolant presents the possibility of serious corrosion problems at reactor operating temperatures of 1100° to 1200° F. Because these corrosion problems would be greatly eased at lower temperatures, a drop in reactor operating temperature should be seriously considered for breeder-reactor power systems. The lower temperatures would also ease some of the fuel and cladding problems. Of course, lower temperatures will require some penalty in power-conversion efficiency. But since operating costs of nuclear power systems are not as sensitive as fossil-fueled plants to small changes in efficiency, a drop in system operating temperature seems to be economically feasible in order to increase the reliability of the fuel elements.

Last, the use of helium as the coolant is an attractive alternate to sodium for both the reactor and the intermediate loop. Helium would overcome the problems of sodium corrosion and sodium-water reaction problems; however, it also introduces a potential safety problem in the event of a loss-of-coolant accident. If methods of overcoming this safety problem can be developed, helium should be more seriously considered as a coolant.

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TABLE 1-I. - COMPARISON OF FUEL COSTS

Type of powerplant	$\frac{\text{Fuel costs}}{\text{Total generation costs}}$
Fossile	0.48
Nuclear burner	.37
Nuclear breeder	[a].16

[a]Estimated for 1985.

TABLE 1-II. - COMPARISON OF BREEDING CHARACTERISTICS

	Burner	Breeder
Primary fuel	U^{235}	Pu^{239}
Fertile material	U^{238}	U^{238}
Average neutron velocity	Thermal	High
Typical breeding ratio	0.6	1.43
Doubling time, yr	--------	8 to 12

TABLE 1-III. - FACTORS AFFECTING CHOICE OF CLADDING MATERIAL

Material	Relative tubing cost	Breeding ratio
Stainless steel	1	1.43
Nickel alloy	1 to 2	1.35
Vanadium alloy	5 to 10	1.43
Niobium alloy	15 to 20	1.14
Molybdenum alloy	20 to 25	1.14

TABLE 1-IV. - EFFECT OF COOLANT ON BREEDING RATIO

Coolant	Coolant captures per 1000 neutrons	Breeding ratio
Sodium	30	1.43
Helium	1	1.55

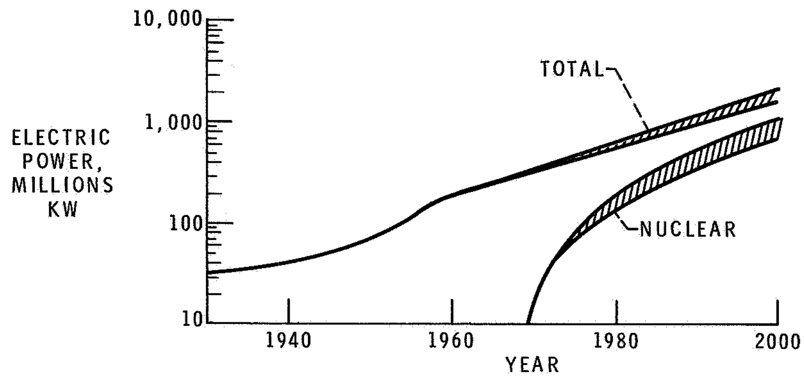

Figure 1-1. - Estimated electric power trends.

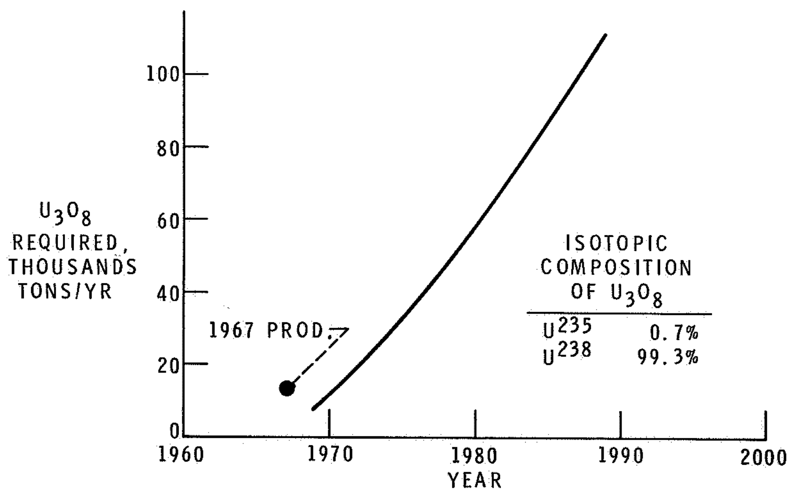

Figure 1-2. - Annual U_3O_8 requirements using present reactors.

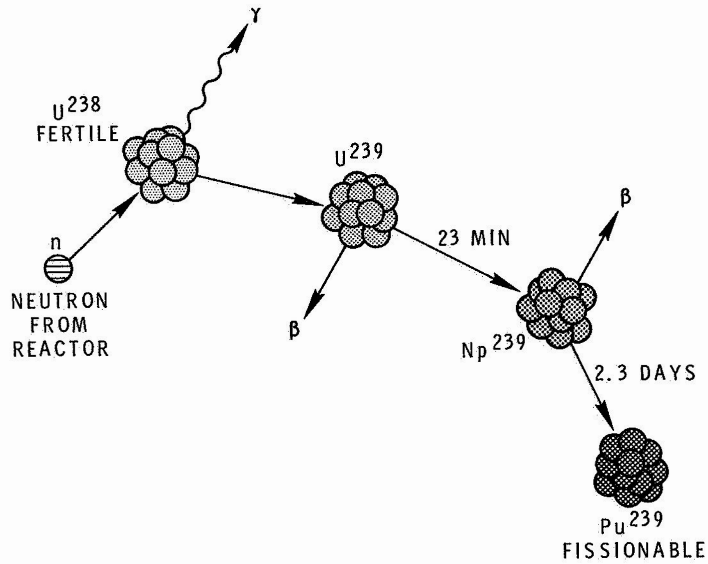

Figure 1-3. - Plutonium 239 production in a reactor.

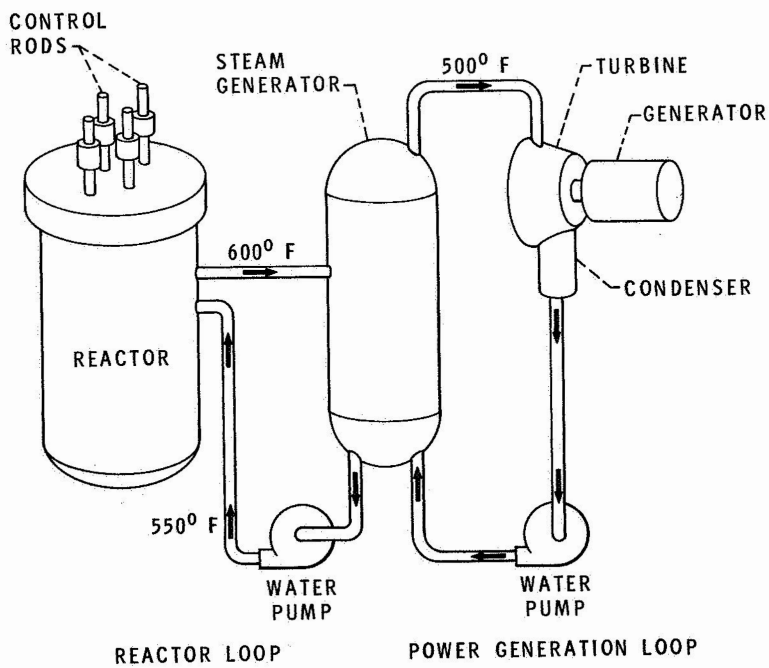

Figure 1-4. - Pressurized-water burner-reactor system.

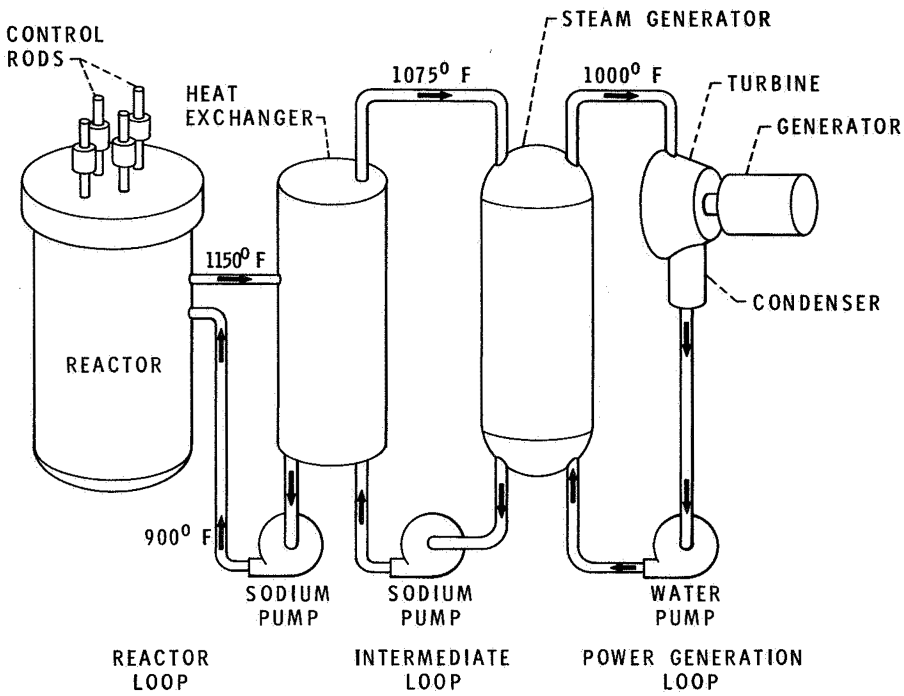

Figure 1-5. - Liquid-metal breeder-reactor system.

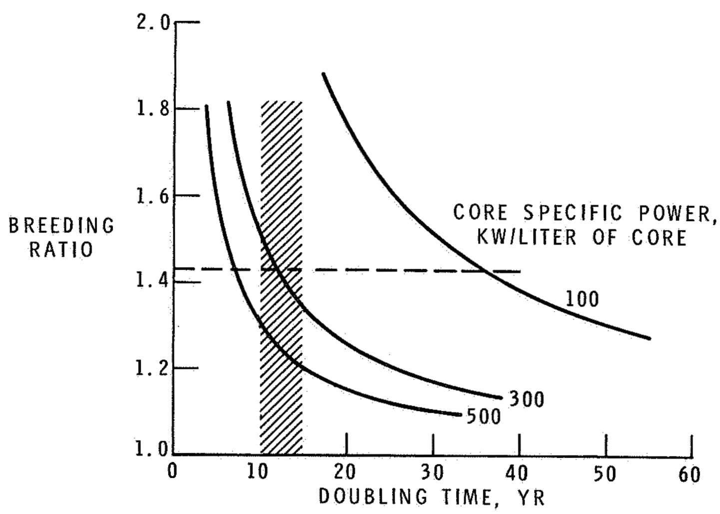

Figure 1-6. - Effect of power density on breeding characteristics of a typical reactor.

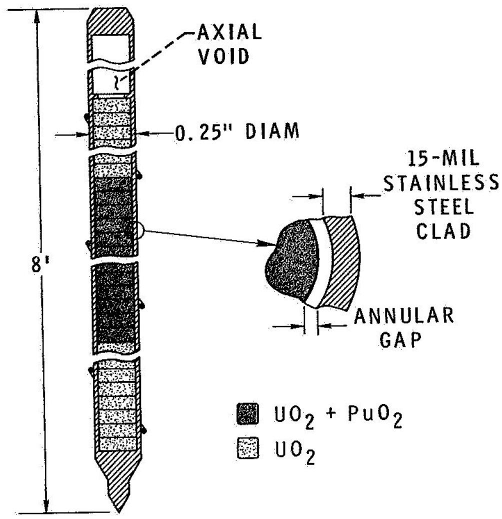

Figure 1-7. - Typical breeder-reactor fuel pin.

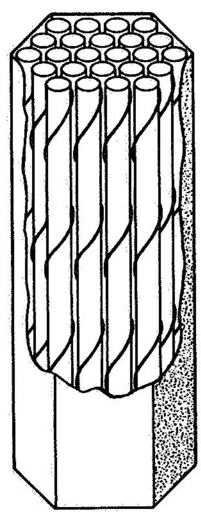

Figure 1-8. - Fuel-element assembly.

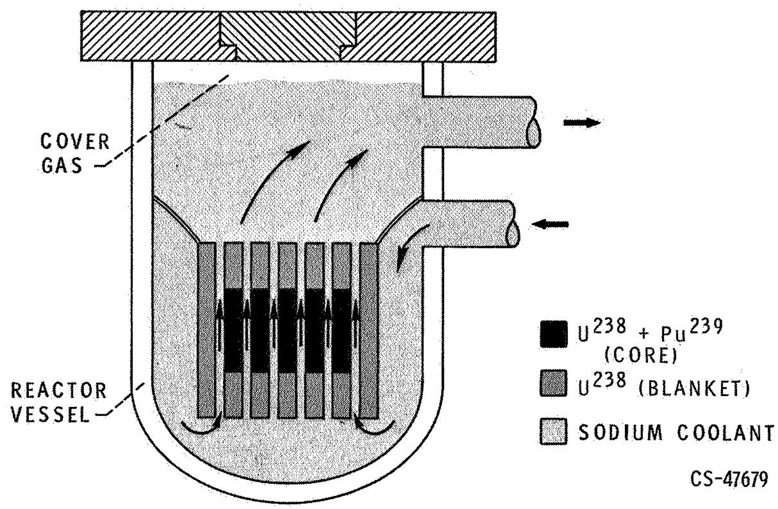

Figure 1-9. - Breeder reactor.

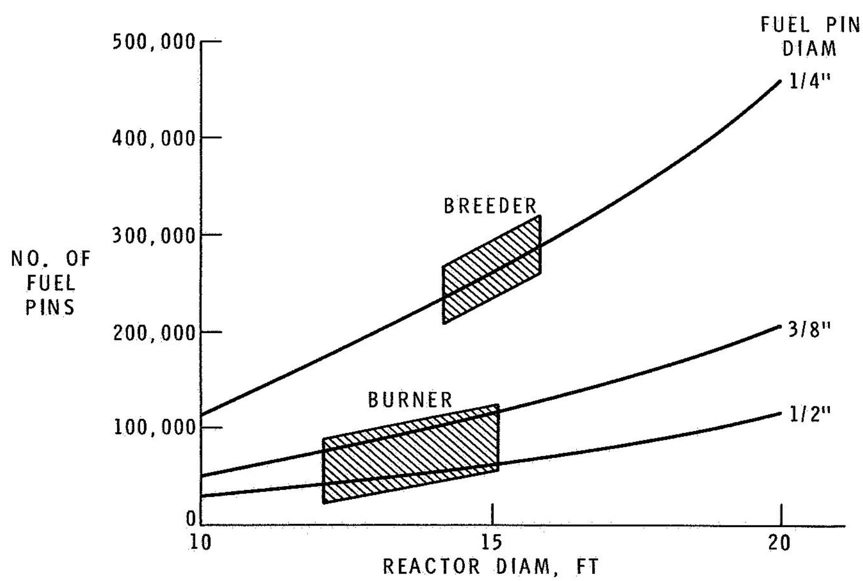

Figure 1-10. - Number of fuel pins required for 1000-megawatt-electric reactors.

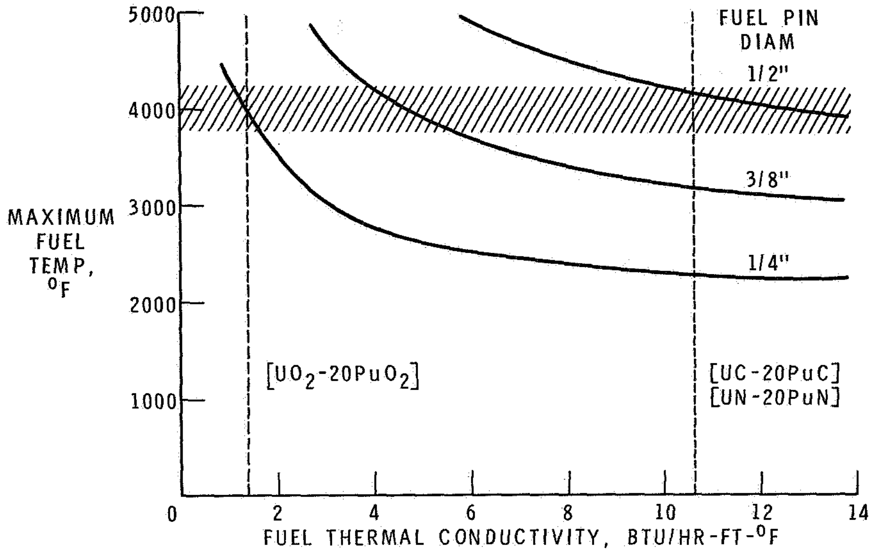

Figure 1-11. - Thermal conductivity and allowable fuel-pin diameter at 300 kilowatts per liter.

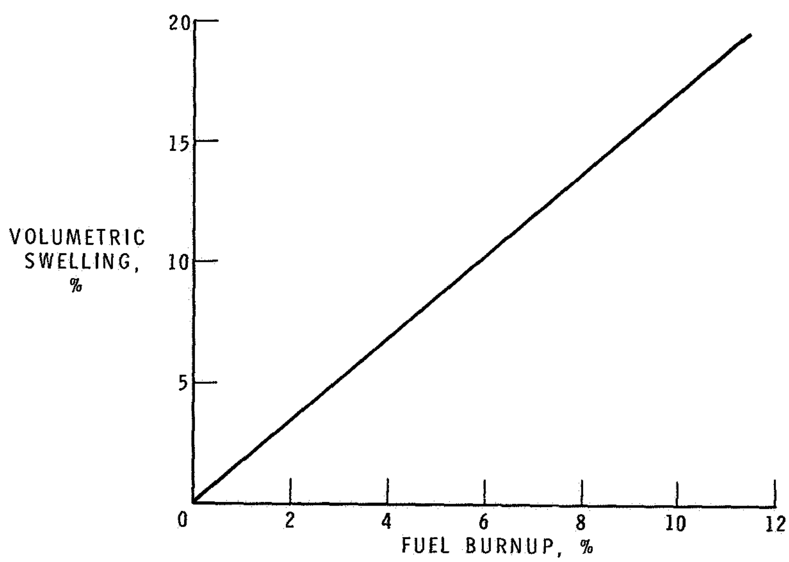

Figure 1-12. - Irradiation swelling of dense oxide fuels with no gas release.

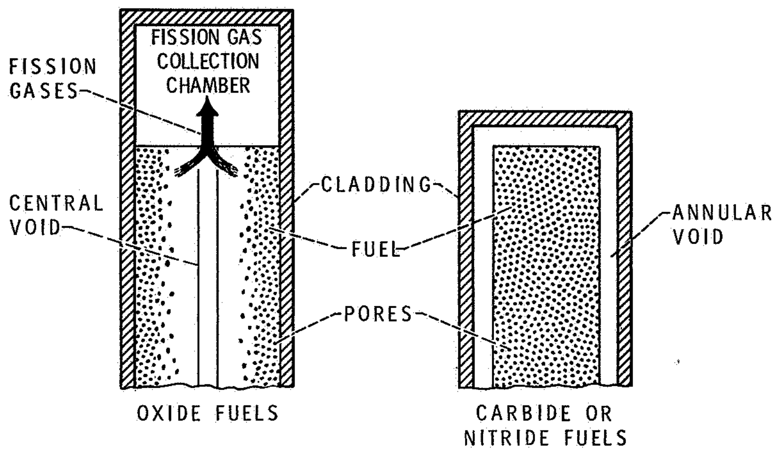

Figure 1-13. - Fuel-pin configurations.

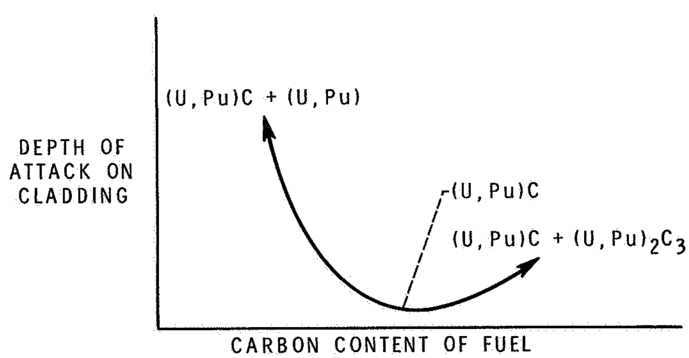

Figure 1-14. - Effect of fuel stoichiometry on compatibility with cladding.

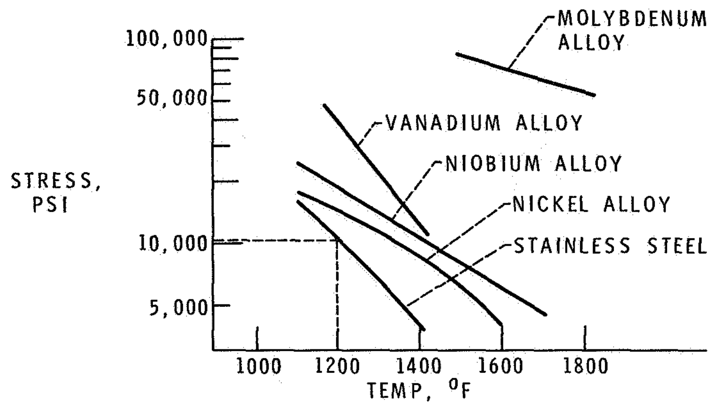

Figure 1-15. - Creep strength of cladding alloys. Stress to produce 1 percent strain in 10,000 hours.

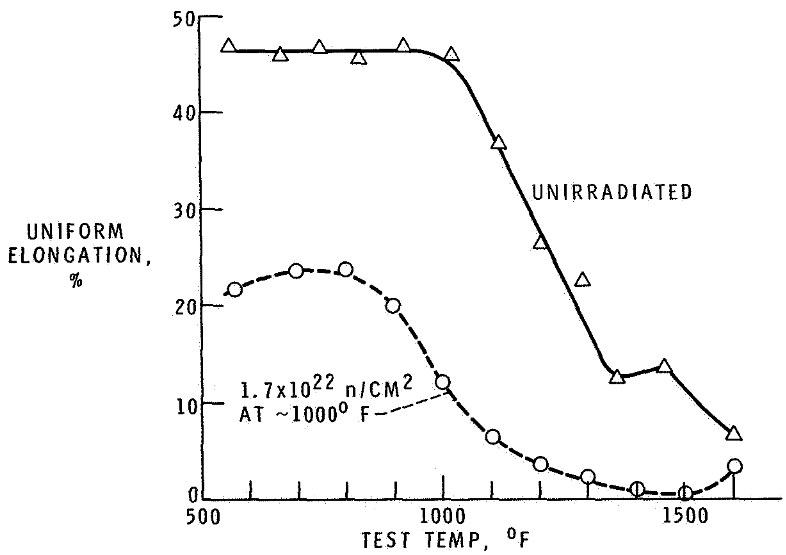

Figure 1-16. - Effect of irradiation on ductility of stainless steel.

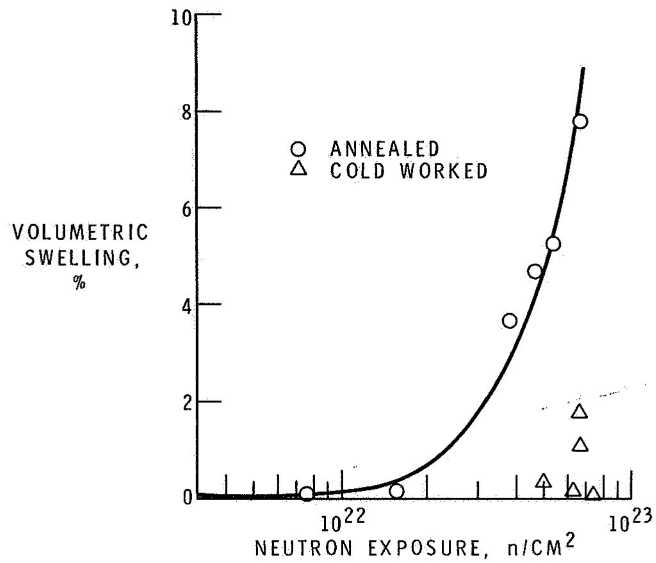

Figure 1-17. - Irradiation swelling of 316 stainless steel. Irradiation temperature, 680° to 1080° F.

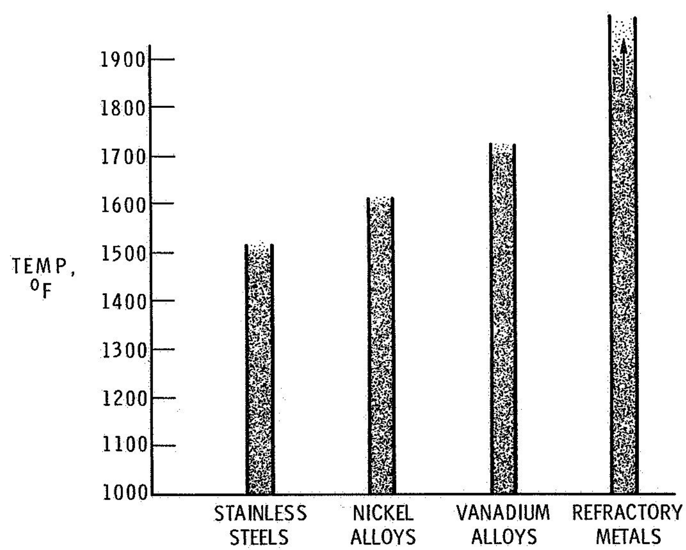

Figure 1-18. - Estimated compatibility limits of metals with high-purity sodium.

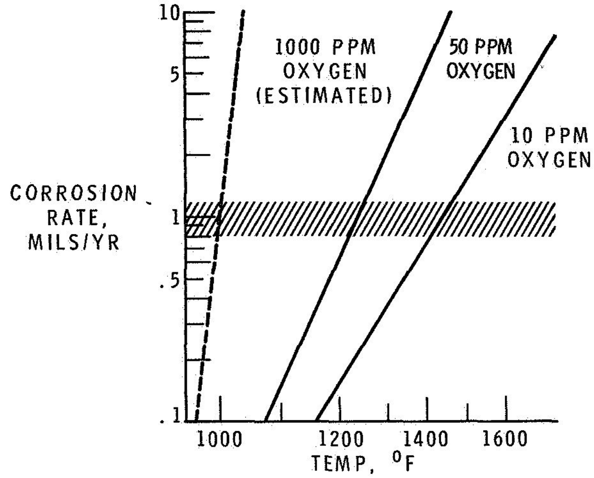

Figure 1-19. - Effect of oxygen on corrosion rates of stainless steel in sodium.

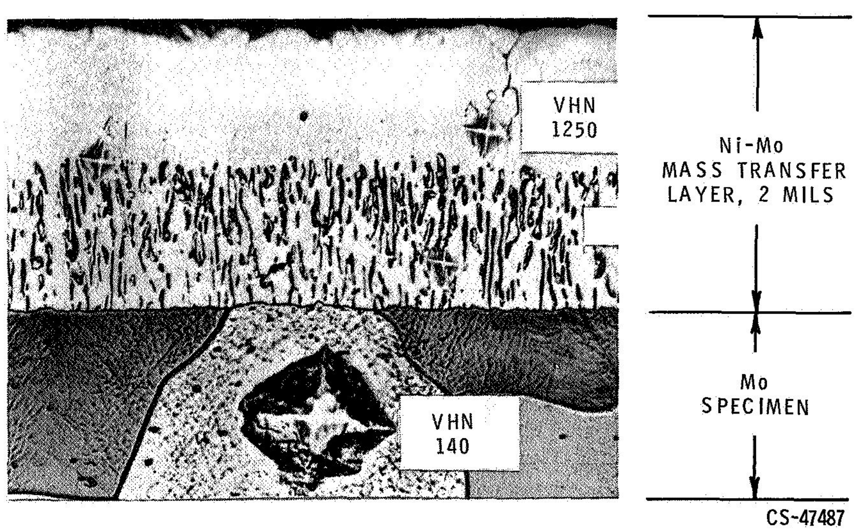

Figure 1-20. - Mass transfer of nickel to molybdenum for 100 hours in sodium at 1830° F.

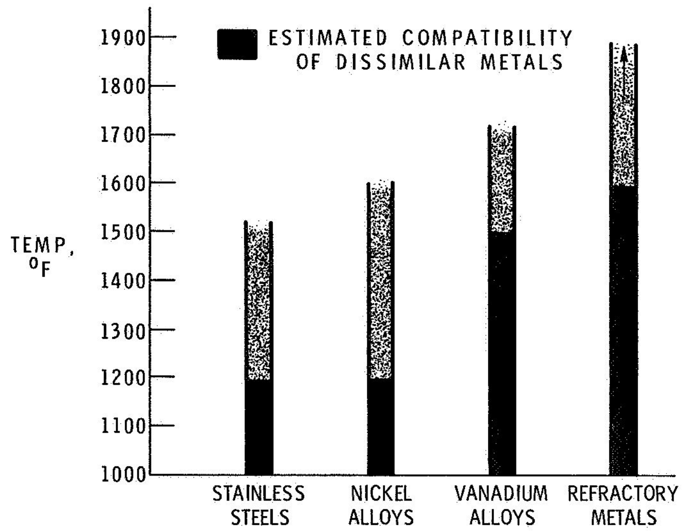

Figure 1-21. - Effect of dissimilar metals on compatibility with high-purity sodium.

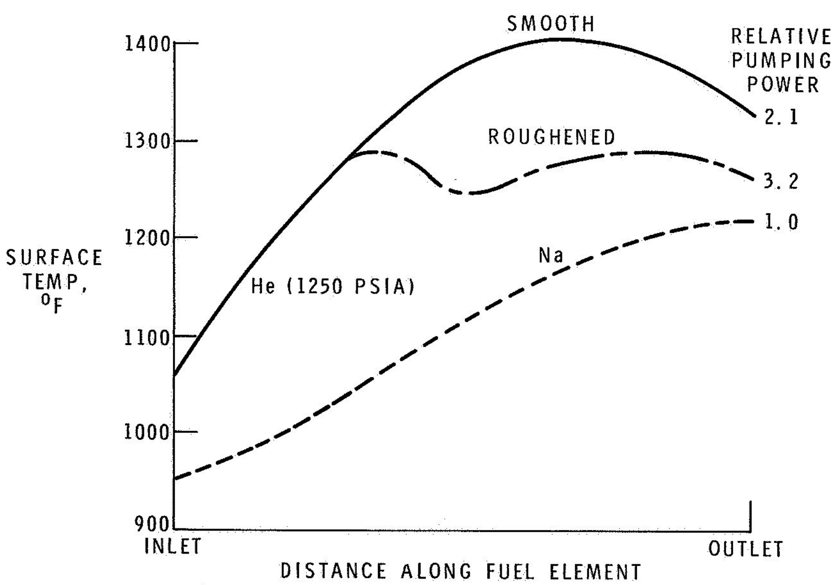

Figure 1-22. - Fuel-pin surface temperature distribution. Outlet temperature, 1200° F; coolant temperature rise, 250° F.

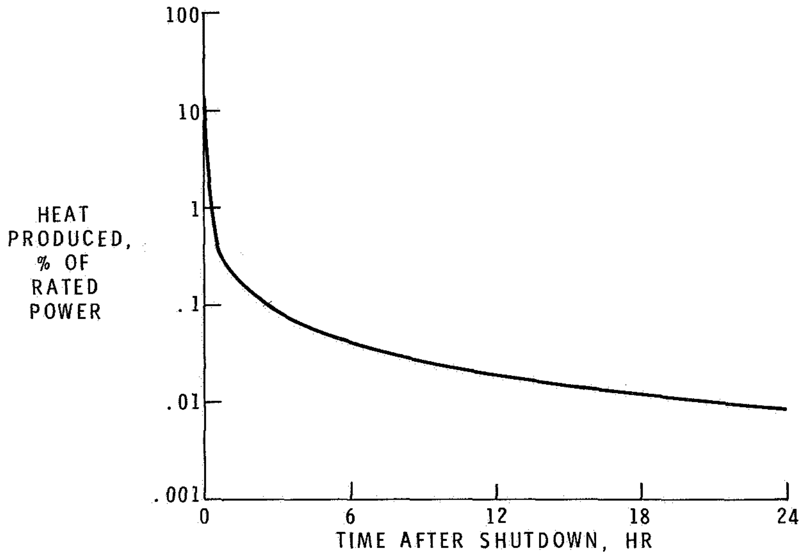

Figure 1-23. - Reactor power decay after shutdown.

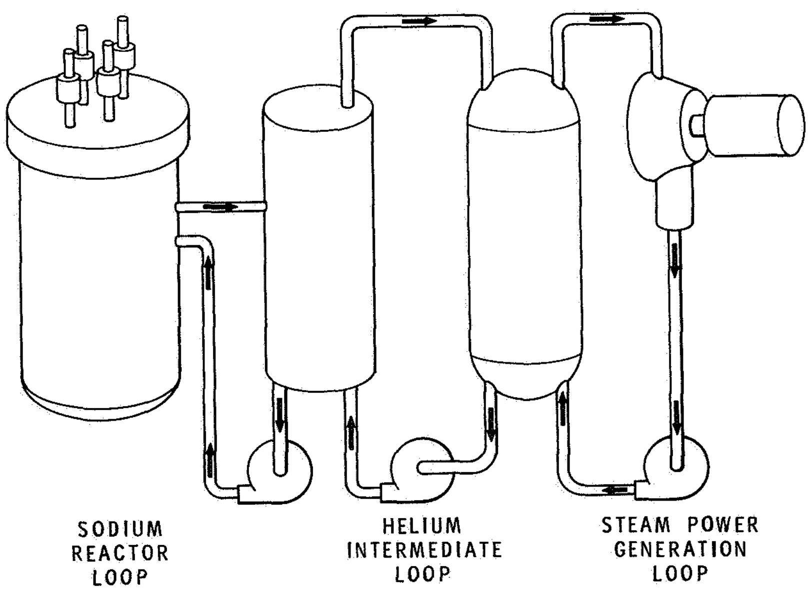

Figure 1-24. - Breeder reactor system.