**SODIUM GRAPHITE REACTOR
MATERIALS SURVEY**

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ABSTRACT

The materials problems associated with the present sodium graphite reactor system have generally been approached by using existing knowledge and data to meet the proposed operating conditions. This discussion reviews the general reactor concept and the specific materials used for the major reactor components:

- 1) shielding materials
- 2) core materials
- 3) sodium cooling system materials

In each case, the materials problems and the materials used to minimize or eliminate these problems are described. Economical nuclear power is currently dependent on the flow of improved materials for high temperature use in high radiation fields. Rapid progress is being made in this respect.

I. INTRODUCTION

The Sodium Graphite Reactor (SGR) system represents one of the major approaches of the AEC nuclear power demonstration program to develop economic nuclear power. It is one of the more advanced types of thermal reactors. Within the engineering scope of this project, there have been many unusual, interesting, and perplexing materials problems. The approach to these problems has been to use existing engineering data, with a minimum of extrapolation. This discussion will describe the general problems associated with sodium graphite reactors. In some instances, the specific problems encountered in the Sodium Reactor Experiment (SRE) are used as illustrations. The procedure here has been to solve the immediate problem to the point of operability and also carry on a development program for longer range optimum performance. It is understood that a sodium graphite reactor can be designed in many ways; and, with a specific design, there may be alternative materials, depending on that design. The AEC material development program is generating data at such a rapid rate that certain uses are almost obsolete, by the time construction and testing are complete.

The method to be used here to survey the SGR materials is quite straightforward. First, the general reactor system will be described. Then, this system will be broken down into the major components of:

- 1) shielding materials, consisting of thermal and biological shields;
- 2) core materials, consisting of the sodium coolant, the moderator and reflector, the fuel element, and control and safety elements;
- 3) the sodium cooling system materials, consisting of the main tank, pumps, valves, traps, and heat exchangers.

The physical environment and operating conditions are then reviewed in each case, together with the materials chosen for these conditions.

In a heterogeneous reactor, such as the SGR, the fuel elements are arranged in a lattice design, and the moderator and coolant are kept separate. Both the fuel and the moderator are protected by a metal can. The function of the moderator can is to prevent interaction between sodium and graphite. The fuel element can also prevent minor reactions between the fuel and sodium, but its primary

functions are to retain fission products and act as a stress-bearing member. In all thermal reactors, one of the primary concerns of the materials engineer is the neutron capture cross section of each of the components. Each problem must be judged on its individual merit. The end result is usually a compromise between the nuclear engineer, trying to minimize neutron absorption, and the materials engineer, recommending the optimum material for the operating conditions. Some circumstances require a relaxing of the operating conditions for mutual compatibility. One should recognize that considerably more emphasis is placed on the materials problems in nuclear reactors, due to the new and unusual circumstances. In general, the materials choice depends on:

- 1) quality of the original source material
- 2) method of fabrication
- 3) subsequent special treatments
- 4) inspection
- 5) operating environment
- 6) maintenance
- 7) in specific cases, reprocessing of special materials.

A brief description of the SRE will be used to illustrate the basic design and relationship of the components. The SRE is designed for the production of 20 Mwt. The reactor is cooled by sodium, which circulates in a primary system and becomes radioactive. This primary sodium transfers its heat to a secondary, nonradioactive sodium system through intermediate heat exchangers. The general arrangement is shown in Figures 1 and 2. The reactor is located below grade, with the upper surface of its top shield at floor level in the reactor room. Motors for the mechanical sodium pumps and for the control rod drives are located above floor level for easy maintenance. A 75-ton capacity bridge crane, capable of supporting a lead-shielded cask, is used, with the cask, to insert and remove fuel elements from the core. The entire core is contained in a stainless steel vessel. The coolant system is at approximately atmospheric pressure, due almost entirely to helium gas which blankets the sodium. The sodium itself has a vapor pressure of about 0.1 mm at SRE conditions. No building containment vessel is used. The graphite moderator is supported on

Figure 1. General SRE Arrangement

Figure 2. SRE Reactor

a stainless steel grid plate near the bottom of the core tank. The zirconium-clad graphite moderator is in the form of hexagonal prisms. The graphite reflector is at the top and bottom of the moderator and is an integral part of the canned assembly. The graphite assemblies contain an axial zirconium tube, in which the fuel elements are suspended. The sodium enters at the bottom plenum, at 500°F, and passes up through the axial tubes and between the moderator cells, and exits at 960°F. The main core tank is surrounded by a steel thermal shield and an outer tank to contain the sodium, in case of a leak. An outer steel cavity liner serves as a support for the concrete foundation. This foundation extends up to the biological shield at floor level. As a closure for the reactor vessel, there is a shield supported by the cavity liner, and also a rotatable shield. Large-diameter bellows provide a gas seal for the core tank atmosphere. Separate bellows and diaphragms seal the galleries and permit the thermal expansion of the various piping which penetrates this region. Table I shows the principal operation conditions.

TABLE I
SRE Operating Conditions

Sodium inlet temperature	500°F
Sodium outlet temperature	960°F
Average thermal flux	2.5×10^{13}
Max. fuel temperature	1200°F
Fuel	slightly enriched U
Sodium flow	~15 ft/sec
Pressure drop, total	~15 psi

II. SHIELDING MATERIALS

There are two principal types of functional shields used in sodium graphite reactors. They are the thermal shield and the biological shield. The specific design will determine, to some degree, the types of materials and conditions under which they must operate.

The thermal shield hangs inside the main core tank. Its purpose is to absorb gamma rays and some neutrons from the core, thereby smoothing out the thermal gradients and reducing the stresses in the stainless steel main core tank. The design provides for a loose-fitting assembly of concentric cylinders in contact with sodium. The thermal shield can be fabricated of low carbon steel, due to the low stress conditions and adequate corrosion resistance.

The biological shield consists of high density concrete. Magnetite ore is used in the aggregate to raise the density to about 230 lb/ft^3 . This is not a maximum density concrete, but represents a compromise between cost and operational requirements. Higher densities have been obtained in reactor concrete shields, by using steel punchings. The magnetite ore was chosen over the steel punchings for SRE use because of the ease of aggregate mixing, more uniform density and homogeneity, and cost. Since cost is a major consideration in large quantity bulk items, such as shielding, the local ground rules for choice of aggregate type may change with reactor location.

The general reactor design is a major factor in the choice of shielding materials. For instance, in the gallery areas of the SRE, the shielding requirements were satisfied with ordinary concrete in massive, movable sections. In areas where penetrations of the shielding are required, such as fuel-element and rod-system ports that go through the top shield, it is important to use high density shielding. The thickness of the top shield affects many other structural decisions. By reducing the top shielding thickness to a minimum, the total length of a fuel element shield plug and assembly is minimized. This, in turn, reduces the lead-handling-cask height, the building height, cleaning cell length, and hot cell shielding. In addition, the length of the steel sleeves, which are used as liners for the shield penetration ports, is also decreased. These modifications all reduce cost.

It is worthwhile to mention briefly a possible alternate dense aggregate for concrete shields, namely, ferrophosphorous. This is available commercially as a crude mixture of FeP and Fe_2P . For comparison, the three types are shown in Table II for the SRE top shield conditions.

TABLE II
SHIELDING MATERIAL COMPARISON

Ordinary concrete required	9 ft
Magnetite concrete required	6 ft
Ferrophosphorus required	4 ft

After a complete survey, which included an analysis of shipping costs, the ferrophosphorus was found to be too expensive for the particular conditions of the SRE.

The radiation levels directly determine the amount of shielding and the configuration required. In areas routinely occupied by personnel, the design radiation level is 10% of tolerance. In areas not routinely occupied, tolerance level is observed. Tolerance, in this case, is 300 mr/week or 5 r/yr.

Other examples of shielding used are the lead shielding of the fuel handling cask and the steel shot used in special forms around pumps and minor supporting structures. Lead is used for the handling cask principally because of its low bulk and ease of assembly. Steel shot adapts itself to complexities of design.

III. CORE MATERIALS

The core materials represent the most challenging materials problems in a reactor. The known effects of environment upon these materials are severe; probably many more will become apparent as work progresses. Fifteen years ago, the materials that today are considered fairly conventional were only curiosities. The chemical processes and high quality production standards achieved for these core materials represent truly great advances.

In the SGR concept, the principal material problems encountered in the core were:

- 1) corrosion of zirconium by sodium containing oxygen and other impurities,
- 2) radiation stability of fuel materials,
- 3) the reaction between sodium and graphite

The last problem was resolved by design, rather than by choice of materials, when the moderator cells were enclosed in cans. The two former problems are still large, but not insurmountable. In describing the core problems, the major topics have been divided among the coolant, fuel, moderator, and control and safety materials.

A. SODIUM COOLANT

Most of the materials problems with sodium result from the impurities in the sodium and the materials with which it comes in contact. Sodium has excellent properties for a heat transfer medium. It has:

- 1) low density, and thus low pumping costs
- 2) relatively low material cost
- 3) relatively low neutron absorption cross section
- 4) high thermal conductivity
- 5) relatively high boiling point
- 6) low melting point*

*A basic review of the sodium problems is very well presented in a paper by McIntosh and his associates in the Journal of the Institute of Metals - 1956.¹

These factors, and general conditions to be described, provide a background for the subsequent more detailed discussions on sodium corrosion and sodium-zirconium reactions.

There are three main types of reaction involved between sodium and the other reactor materials. First, a reaction takes place between the liquid and the container. This may result in either selective or general corrosion. The results of this type of interaction include alteration of the container material properties, and mass transfer from one part of the circuit to other parts of the circuit. In the second type of reaction, an insoluble reaction product is formed. The continued effect depends on whether this is adherent or nonadherent to the container material. Adherence to the container can affect the heat transfer properties, both by film growth and by interference with coolant flow. Continued film growth, particularly when the film is nonadherent, can lead to mechanical failure, as a result of erosive or corrosive attack by the coolant. In the third group of reactions, the principal effects are due to impurities, either entrained or in solution in the dynamic liquid metal. Oxygen may be introduced during initial loading or by the reduction of oxide films in other parts of the circuit. Engineering factors, such as the surface finish of pipes, the condition of welds, and the presence of residual stresses, are no less important than the metallurgical nature of the solution formed.

In the case of the SGR, oxygen is one of the most important impurities in sodium. The solubility of sodium oxide in sodium is presented in Figure 3.

Figure 3. Solubility of Na_2O in Sodium

This data is valuable in at least two ways:

- 1) to indicate a method for removal of oxygen,
- 2) to provide an explanation of the temperature dependence of interactions in the oxygen-sodium-zirconium system.

The specifications for reactor-grade sodium will illustrate the requirements and need for a high quality product. Due to the large amount of sodium present in the core, the chemical specifications are based on impurities that contribute to the cross section, long life activity, and general corrosion. Table III shows the chemical specifications.

TABLE III
SODIUM SPECIFICATIONS
(Allowable Limits of Impurities)

Equivalent Effect on Cross Section		Equivalent Effect on Long Life Activity		Other Impurities	
Element	ppm	Element	ppm	Element	ppm
B	20	Co	2	Al	20
Li	150	Cs	6	C	30
Cd	5	Fe	300	H	100
Gd	0.5	Rb	100	O	70
Sa	1	Se	0.5	Mg	20
Eu	3			Ni	10
Hg	40			Si	10
				Ti	30
					20

The cross section requirement is based on an equivalent thermal neutron absorption of 20 ppm of boron of any of the elements mentioned. The long life activity values are based on the need for minimizing the activity after the normal sodium decay, in order to permit accessibility for maintenance. In all instances, careful handling procedures throughout the sodium processing, handling, loading, and unloading operations are necessary.

B. MODERATOR AND REFLECTOR

Present designs of sodium graphite reactors use a canned moderator configuration. This technique is principally based on the need for isolation of the sodium and the graphite. Tests have shown that contact between these materials results in absorption of sodium into the pores of the graphite, which causes two phenomena to occur. One is a loss of nuclear reactivity, which may lead to shutting down the reactor. The other is a swelling of the graphite (about 1%), which could cause stresses on structural members. The macroscopic cross section (Σ) for reactor-grade graphite is in the order of $4 \times 10^{-4} \text{ cm}^{-1}$ and, when the graphite is permeated by sodium, $3 \times 10^{-3} \text{ cm}^{-1}$. The swelling observed in nearly pure sodium is greatly increased when potassium is present, as is the case with the liquid metal (NaK) bond used in the SRE fuel elements.

There is a unit size for each graphite canned cell which is determined by optimizing the quantity of canning material, the ease and cost of manufacture, and the accessibility for replacement. Since the can material is in the direct path of neutrons, the cross section should be as low as structural, corrosion, and other conditions permit. With these limitations, zirconium, zirconium alloys, and stainless steels are the logical choices for immediate use. More exotic materials which appear promising include beryllium and niobium. However, large scale fabrication, welding, and cost problems are, at present, barriers to the use of these materials. On the basis of the relative importance of neutron absorption cross section, it is apparent that zirconium meets the immediate requirements for the moderator can, and that stainless steel can be used for the reflector canning material. A sodium temperature limitation of about 960°F has been found to exist for unalloyed zirconium. This consideration is based on structural strength and grain growth in the zirconium when in contact with sodium containing what is presently the minimum amount of oxygen attainable. Zirconium alloys, such as Zircaloy II and III, are receiving consideration as a future substitute. Development work is in the early stages, and decisions on their use will depend on experimental results. A typical moderator can assembly is shown in Figure 4.

Figure 4. Moderator Can Assembly

There are two additional major areas for material problem examination in this section of the reactor: the moderator and reflector material itself, and the canning materials.

The moderator and reflector material is reactor-grade graphite. The requirement of sheathing the graphite makes it necessary to investigate the gas evolution problem of the graphite under operating conditions. Commercial artificial graphite exhibits substantial sorption properties for all but the chemically inert gases. Oxygen, nitrogen, hydrogen, and carbon oxides are among the gases most easily absorbed. All of these gases are also readily absorbed by the zirconium cladding at reactor temperatures. This results in some deterioration in the physical properties of the zirconium. The effect on the zirconium is much more evident on the low-temperature properties than the high-temperature properties. The types and quantities of gases absorbed and released by the graphite by thermal stimulation are directly related to the

method of graphite manufacture and the chemical quality of the product. This problem, which is inherently quite complex, is further complicated by radiation. The net volume of gas released from a given type of graphite, on thermal annealing at 1800°C, has been arbitrarily taken to represent the amount of gas available as a result of operating at 600°C in a sodium graphite reactor. A number of commercial sources of graphite supplied samples for evaluation. The graphite finally selected represented an adjustment in some of the standard industrial graphite fabricating techniques to minimize the absorption of gases. The assembly of the graphite canned units involved outgassing operations and letting down to a final atmosphere of helium. To avoid possible effects like embrittlement of can materials and excessive gas pressure, a vent tube is provided in each cell which permits the excess gas to escape to the helium atmosphere blanket above the top sodium surface. There are modifications of graphite, now in the development stage, which may be applicable in the future. These efforts include higher density graphite, impervious graphite, and coatings on graphite. Each improvement will require evaluation in terms of improved reactor operating conditions and lower cost. The cost problem is not limited to the graphite alone, but includes the consideration of canning material costs and assembly costs.

Another important materials problem with graphite is radiation damage. The changes in properties of graphite under irradiation are directly related to temperature. These changes involve thermal conductivity, stored energy, lattice parameters, and physical dimensions. Irradiation at low temperature results in a large decrease in thermal conductivity and substantial dimensional increases. Experiments have indicated that postirradiation thermal annealing at 2000°C is accompanied by the return of all properties to substantially their initial values. The relative roles of postirradiation annealing and spontaneous annealing during irradiation have been examined through controlled temperature irradiations. The changes in thermal conductivity, lattice parameter, and physical dimensions transverse to the axis of extrusion of the graphite, are shown in Figures 5, 6, and 7. These data indicate that changes produced by irradiation at elevated temperature are much less severe than those produced at 30°C. Extrapolation of these data to typical SGR exposure, at approximately

Figure 5. Change in Lattice Parameter Due to Irradiation of Graphite

Figure 6. Change in Dimensions Due to Irradiation of Graphite

Figure 7. Change in Thermal Resistivity Due to Irradiation of Graphite

600°C, will not indicate significant deterioration of the graphite properties. The changes that are expected can be taken care of by approximate design tolerances.

In the use of stainless steel and zirconium canning materials for the graphite moderator, there are chemical and thermal conditions affecting the can which must be considered when making a choice of material. Externally, there is the corrosive action of sodium. The internal conditions are the compatibility with the graphite, and the gases released from it on heating. An additional condition which requires examination results from the assumption of a sodium leak. This would place sodium, graphite, and metal in contact at high temperature. With the stainless steel, the principal problems are:

- 1) carburization of the stainless steel
- 2) mass transfer of highly radioactive constituents from the core region to remote regions of the coolant circuit that are supposed to be directly maintainable

The ordinary problems of fabrication, strength, thermal conductivity, etc., do not apply, other than to govern the choice of materials whose properties are adequate.

A series of capsule tests was conducted, in which Type 347 stainless steel, graphite, and sodium were in contact at various temperatures. The stainless steel was then examined for carburization. Table IV shows the results.

TABLE IV
CARBURIZATION OF TYPE 347 STAINLESS STEEL

Temperature (°C)	Effect
450	No effect
600	Moderate carburization
675	Heavy carburization
750 to 900	Heavy to massive carburization

Similar tests were made under irradiation with substantially the same results. The data shows that stainless steel will carburize at temperatures of about 600°C in the presence of sodium and graphite. Heretofore, most metallurgists have assumed that the carburization reaction required oxygen. The capsule test method appears to produce results without the action of oxygen or, at most, with extremely minute quantities of oxygen present in the sodium as an impurity. A trace quantity of oxygen may be sufficient to act as a carrier. Very recently, the British have observed compound formation between sodium and carbon; but data are too meager to be significant. Under other special conditions, particularly in the ferritic steels, a reverse mechanism of decarburization has been observed.

The mass transfer problem is principally concerned with the temperature-dependent solubility in sodium of certain constituents. Although the exact mechanism is not fully understood, the transfer of radioactivity apparently occurs, even though high solubility may not be present. It is assumed, in the general reactor design, that certain major components in the coolant system will be available for maintenance. If small quantities of highly radioactive materials concentrate at these components, it would render the maintenance extremely

hazardous and expensive. Small-scale dynamic loop tests were conducted, in our laboratories and others, to explore this phenomenon of mass transfer. The results indicated that the process is sensitive to the oxygen content of the sodium, with the transfer rate varying directly with the oxygen content. Additional evidence of the oxygen dependence is offered by the fact that the additions to sodium which most effectively inhibit the activity transfer are those which tie up oxygen as insoluble oxides. Further tests were made to inhibit these reactions. Of the materials used, barium was the most effective. The problem of mass transfer does not now appear to be major in the SGR systems, providing the oxygen content of the sodium is maintained at a low level. The problems with the zirconium can material are somewhat different.

Zirconium is much more susceptible than stainless steel to attack by both the oxygen and other impurities in the sodium and the gases evolved from the graphite outgassing. The problems with zirconium are principally three-fold:

- 1) limited temperature
- 2) solubility of oxygen, nitrogen, and carbon
- 3) fabrication

The use of zirconium as a canning material does not involve substantial stresses; consequently, the low strength of the material at elevated temperature offers no serious disadvantage, although this does limit the sodium to a maximum of about 1000°F. This problem is also interrelated to other problems to be described. Zirconium alloys containing aluminum, molybdenum, and niobium are being developed for high temperature use, but currently are at the laboratory stage.

Another limiting problem with zirconium is the grain growth at elevated temperatures. This is particularly important in thin sections, such as the 35-mil-thick moderator can. Grain growth has been shown to occur at approximately 1000°F and higher. Two mechanisms of grain coarsening are found to occur in zirconium at elevated temperatures:

- 1) uniform grain growth
- 2) germinative grain growth

Large grains produce a structure that is more susceptible to cracking, due to creep, fatigue, and embrittlement from impurities, than are the fine-grained structures. This phenomenon was discovered during the can fabrication process and received intensive study. The large grains resulted in abnormally short fatigue life. This property is particularly important in the moderator can head, which may flex, due to changes in sodium temperature from the inside process tube to the outside sheath of the can. The fatigue problem is not singularly associated with grain growth, but is also affected by the oxygen and hydrogen impurities. It is believed that the grain growth problem may be reduced when zirconium is replaced with zirconium alloys.

The fabrication of zirconium also had its share of difficulties, careful control being necessary at each phase of processing. Excessive temperatures or contact with contaminating mediums, such as salt baths or gaseous atmospheres, required rigid control. These important technical details are further complicated by the relatively high cost of zirconium. This makes a high processing yield imperative. Experience at Atomics International emphasizes that double arc-melting under vacuum is required to produce soft ingots and thus relatively high process yields. Annealing in vacuum is an important aspect of the ingot-to-sheet fabrication. The product of ingot rolling is 35-mil sheet, which is then blanked into panels 6-in. wide. All panels are ultrasonically inspected for defects, formed on a power brake (see Figure 8), and welded. The latter operation is also quite crucial. A special automatic welding machine was modified to provide three important conditions:

- 1) an inert gas backup beneath the weld deposit
- 2) a trailing shield to provide inert gas while cooling
- 3) chill bars.

Almost two miles of butt welding without filler was required. In cases where heavier stock was used on the can head, a filler wire of zirconium was used under the same conditions. One hundred percent inspection was made by dye penetrant, X-ray, and helium leak detector. A complete set of can components is shown in Figure 9.

Figure 8. Blanked and Formed Zirconium Panels

Figure 9. Zirconium Can Components

Finally, let us review the susceptibility of zirconium to external reactions by oxygen and possibly hydrogen in the sodium, and the carbon and gaseous products from the graphite in the interior of the can. There has been no evidence of solubility of zirconium in sodium, even at higher temperatures than those anticipated under SGR conditions. It should be noted that it is virtually impossible to obtain completely oxygen-free sodium on a commercial basis. Furthermore, it would be impractical to assume that minor amounts of oxygen would not find access to the sodium, either through minor leaks or planned reactor removals and insertions of components, such as fuel elements. Corrosion experiments on zirconium in sodium have shown that the rate of weight gain in zirconium is nearly independent of the oxygen content of the sodium, in the range above 0.005 wt % oxygen. From the data obtained to date, the corrosion rate is determined by the diffusion of oxygen through the initial oxide layer. The oxidation rate curves appear to be parabolic at all temperatures up to 900°C. Zirconium oxide is soluble in zirconium up to about 7 wt %. The rates of diffusion are roughly 1.5 mils/month at 500°C, and 2.0 mils/month at 650°C. An important aspect of this effect has been observed by Treco² in that 2.0% oxygen renders cold work and machining of zirconium impossible. Zirconium has also been observed to be notch-sensitive material, which precipitates rapid fatigue failure. The oxide diffusion problem is greatly reduced by operating at temperatures above 375°C. As pointed out earlier, zirconium is capable of dissolving both hydrogen and nitrogen. The solubilities of these gases are more or less interdependent, and their effects upon physical properties are very similar. The diffusion of hydrogen in zirconium is very rapid, whereas that for nitrogen is relatively slow. There appears to be no diffusion of nitrogen below 800°C. The nitrogen problem is further eased by the lack of solubility or reaction with sodium. Hydrogen, on the other hand, is readily soluble in sodium and, if present, will easily find its way into the zirconium. The hydrogen problem is important but the possibility of adding hydrogen appears less likely than for oxygen, and it appears to have a higher toleration level. However, in view of the necessity for long moderator can life (many years, if possible), means for minimizing these gaseous problems are being developed. This is more completely discussed under the section on trapping in the sodium cooling system. A final problem with zirconium is the possible carburization

in the presence of sodium and graphite. Experiments similar to those described for stainless steel showed that carburization of zirconium is not effected at 600°C. In fact, only slight carburization was observed at 750°C.

Although the initial phases of the use of zirconium as a principal material in sodium graphite reactors showed formidable problems, extensive evaluation has demonstrated its use to be basically sound. The successful operation of the SRE will be further proof of zirconium as an engineering material in this type of reactor.

C. FUEL ELEMENTS^{4,5,6}

The present SGR systems use a solid-metal type of fuel. The fuel element is probably the most important material in the reactor, in terms of total cost. Economic studies indicate fuel costs represent between 25 to 50% of the long term total cost. The choice of fuel design and material are affected by four main aspects:

- 1) the general reactor concept
- 2) the method of manufacture
- 3) performance in the reactor
- 4) the reprocessing

It is necessary, during the choice of a fuel material, to evaluate the overall process flow of these materials. Even after almost ten years of experience in the reactor business, this is a most difficult task. The evaluation is naturally based on cost. The ground rules for economic studies are constantly changing; and, as the atomic energy field changes more from government-sponsored to commercial enterprise, a constant reappraisal is required. Only recently have commercial prices been available on such processing operations as UF_6 to U ingot, and scrap recovery; while reprocessing itself is completely an AEC-sponsored activity, quotations being only recently available. This is only a very brief reference to the importance of fuel material economics and to the large topic of the relation of fuel cost to the choice of a specified material.

The technical problems involved with fuel materials are outlined as follows:

- 1) stresses due to elevated temperature, radiation damage, and accumulation of fission products;
- 2) reactions between the cladding and the fuel;
- 3) reactions between the fuel and the coolant, in case of rupture of the cladding; and
- 4) reaction between the cladding and the coolant.

Of these factors, the most predominant is the radiation damage at elevated temperature. There have been no reactors operating under the temperature conditions anticipated in the present SGR type. It is true that some reactors have a greater temperature difference across a fuel slug, but none have both a high central temperature and a high surface temperature. In order to understand the fuel element problems, let us examine the general configuration shown in Figure 10.

Figure 10. Fuel Element Design

The fuel elements are fabricated in the form of 7-rod clusters. Each rod consists of a column of 6-in. uranium slugs in a thin-walled stainless steel tube, thermally bonded by NaK alloy. The slugs are 0.750 in. in diameter, the jacket tube is 0.010 in. in wall thickness, and the NaK annulus is 0.010-in. thick. The stainless steel jacket material is Type 304 stainless steel and is closed at each end with a welded plug. The NaK bonding alloy extends a few inches above the slug column to a free surface. This space serves as a volume for the thermal expansion of the NaK and for the accumulation of fission products during irradiation. The spiral wire wrap separating the fuel rods serves two functions: (1) to permit the sodium to cool the inner surfaces of the peripheral rods and the center rod, and (2) to increase the coolant mixing by providing a more turbulent flow. With the configuration established, let us look at the operating conditions.

The surface temperature of the fuel is essentially set by the sodium temperature, if one allows for a small temperature drop across the stainless steel cladding and the NaK bond. The sodium temperature varies along the rod from 500 to 960°F. The fuel center temperature has been limited by the alpha-beta phase transformation at 1220°F. This upper limit has been based on the change in volume resulting from the phase change and on thermal cycling results indicating gross distortion effects. The dynamic variation in temperature conditions within a single rod further complicates the problem. Due to the self shielding of the fuel material, there is a maximum drop of about four-to-one in the neutron flux across a peripheral fuel rod, with the average being two-to-one. Thus, two to four times as many fission events occur in this peripheral zone. Similarly, the same proportion of both solid and gaseous fission products develops in this area. These conditions produce a fertile probability for warp, due to the nonsymmetry of stresses, temperature, and physical property changes. For this reason the original fuel design used short slugs. An alternative fuel design being studied is a large hollow slug element shown in Figure 11.

Figure 11. SRE Fuel Elements

The major problems of operating metal fuels at high surface temperature have only recently been known. Under the SGR conditions, there are four types:

- 1) Cottrell creep at relatively low temperature,
- 2) distortion due to thermal cycling,
- 3) the more severe problem of swelling due to fission gas pressure,
- 4) the relatively minor problem of wrinkling.

This last effect, due to grain size and orientation, is expected to be small, due to the flexible NaK bond; and can be easily reduced by adding a small amount of alloying agent, such as silicon. The Cottrell creep effect was observed by the British in Calder Hall. It is largely dependent on the structural load of the

fuel supporting itself in long columns. As the temperature of the fuel increases, up to 400°C, this effect reduces to zero. Although this somewhat contradicts the usual interpretation of creep phenomena, the effect appears quite real. The thermal cycling phenomenon is associated with the anisotropy of uranium and can be greatly reduced by using a randomly oriented grain structure. "Swelling" is the term used to describe the decrease in density of fuel material under irradiation. Experimental results in Great Britain have indicated changes in density in both unalloyed and alloyed uranium. An example of this phenomenon is indicated in Table V.

TABLE V
CHANGES IN DENSITY OF FUEL MATERIALS

Material	Temperature (°C)	Burnup (%)	Approximate Density Decrease (%)
U	20	1.0	2.0
U	300	1.0	5.0 to 10.0
U	500	1.0	30.0
U	800	1.0	100
Th-U (40%)	Same as above		
U-Mo (14%)	Improved by factor of 2 to 3.		

This data has been further substantiated by American results at Argonne and other sites. Paine and Kittle of Argonne National Laboratory have indicated a rather sharp increase in swelling of unalloyed uranium in the 425°C range. There are now under consideration five general techniques to minimize or eliminate this fuel material problem:

- 1) by direct restraint, using a strong can;
- 2) by use of a dispersion type fuel in a nonfissile matrix;
- 3) by internal restraint, using a cermet, such as uranium in uranium carbide;
- 4) by high strength alloys; and
- 5) by a design, such as a cored slug, to permit internal stress relief.

Each of these methods has advantages and disadvantages which will be resolved only by extensive fuel cycle analysis, overall economic analysis, and numerous experimental statistics.

As the other fuel element material problems are examined, rather standard engineering problems are encountered. In the case of reactions between the cladding or jacket, as it is sometimes called, and the fuel, the problems are temperature dependent. Two chemical reactions may occur:

- 1) solid state diffusion above 500°C , resulting in a brittle interface;
- 2) eutectic formation (m. p. 752°C) between the uranium and the iron in the stainless steel.

For these reasons, cans made of steel cannot be considered, where extended fuel temperature excursions exceeding 500 to 550°C may occur. Zirconium and zirconium alloys are being examined for the higher temperature application, but there are few data presently available. In addition, there is a possibility of mechanical interaction between the can and the fuel, resulting from repeated thermal cycling. Regarding other chemical reactions between fuel element materials and the sodium coolant, there are no major problems. All of the metallic fuels are essentially nonreactive with sodium or NaK. This is, of course, dependent on the oxygen content of the sodium. The fuel can retains fission products and also prevents the formation of small quantities of nonadherent

fuel oxides which would become particulate matter in the sodium coolant. Finally, the reaction between the sodium coolant and the Type 304 stainless steel can is again minor. This is described in the section on Sodium Cooling System Materials.

One of the prime objectives of the fuel element program for the SGR has been to develop both a uranium-base fuel and a thorium-base fuel for long burnup at elevated temperature. This has taken the form of an experimental fuel program, in which various types of potentially improved fuel materials and designs are being tested and evaluated. The present program is shown in Figures 12 and 13. The major types being studied are:

- 1) uranium alloys which minimize the effects of the α - β phase transformation and provide increased strength;
- 2) cored slugs to provide a free-volume internal expansion;
- 3) large hollow slugs for economy, symmetry of stresses and temperature; and
- 4) thorium alloy to provide a radiation-stable matrix material and eliminate low temperature phase transformations.

In each case, a complete chemical and metallurgical history is being obtained during the fabrication, in order to interpret the results of hot cell examination of the irradiated fuel. There are also numerous thermocouples inserted in the fuel for the purpose of temperature monitoring during irradiation. During the establishment of specifications for these fuel materials, it was apparent that the choice of chemical quality and surface and internal defects was a subject for extensive (but inconclusive) review. In the case of chemical quality, it is not within the realm of a relatively small study program, aimed at unknown operating conditions, to recommend a substitute for the quality of mass produced material from the AEC fuel material plants. Thus, reactor-grade quality material was accepted. However, the fabrication of the fuel indicated a need for further evaluation of such defects as seams, cracks, and internal porosity. Specific examples of these defects are being examined in the experimental fuel program.

TECHNIQUE	MATERIAL	PIECES FABRICATED	U ENRICHMENT
Powder Compacted	{ U-1.2 w/o Mo U-Large Hollow	{ 515 36	
Cast	{ U-2 w/o Zr U-1.2 w/o Mo U-1.8 w/o Mo U U-1.8 w/o Mo U-2.0 w/o Zr	{ 215 75 65 25 80 80	{ 2.78% 4.5%
Extruded	{ Th-5.4 w/o U Th-11.2% U Th-16.4 w/o U	{ 450 110 75	Max.

Figure 12. Experimental SRE Fuel Procurement

Figure 13. SRE Experimental Program

The timing of the fabrication of the fuel was another prime factor in the choice of the main fuel material. The choice had to be made in the summer of 1954. At that time, the initial cast uranium-zirconium alloys made by Argonne were showing excellent radiation stability results. However, the use of this alloy required large-scale development of homogeneous ingots and subsequent processing. There were also a few inconsistencies in these early irradiation results. The decision was thus made to use, as the main fuel, a standard, off-the-shelf material; alpha-rolled, beta-heat treated uranium.

As the fuel program indicates, there is considerable flexibility, to allow for improvement. The more advanced fuel materials planning includes uranium oxides, restrained slugs, cermetes, and gamma-stable alloys. The choice of these materials and designs is governed by extensive nuclear parameter and overall economic analyses now in progress.

D. CONTROL AND SAFETY RODS

The primary factor in the choice of materials for control and safety rods are the operating conditions, neutron flux, and ambient temperature. It has been a basic assumption in the reactor design to have a clean reactor face for ease of accessibility. This requires that there be no extensions, other than drive mechanisms, above the top shield. Thus, in the case of the control rod, the control rod material must operate at a high temperature, with the drive mechanism located at a position of somewhat lower temperature. The safety rod, however, hangs above the core (but not above the reactor floor) and is subjected to different operating conditions. In both cases, the control and safety rod materials operate inside thimbles made of Type 304 stainless steel, at various locations in the core. The thimble is sodium cooled externally and has a helium atmosphere internally.

The control rod problem is principally one of operating materials under constant dynamic load at high temperature for long periods with utmost reliability. The control rods' functions are regulatory in nature. Their purpose is to provide approximately 10% control of reactivity. There have been various designs examined, both empirically and by experiment. One of the early experimental types employed boron steel balls, flowing under gas pressure. It was discarded, principally due to the possibility of sintering or self-welding

Figure 14. Control Rod

Figure 15. Control Rod With Detail Sections

under operating conditions. The final design for the SRE consists of a poison column of boron-nickel alloy suspended on a pull tube, as shown in Figures 14 and 15. The control rod motion is obtained by a ball-nut-screw arrangement shown in Figure 16. This mechanism should be able to operate consistently at

Figure 16. Control Rod Drive

1200°F. The high temperature results from gamma heating and the relatively poor heat transfer by the helium to the sodium-cooled steel thimble. Herein lies the major control rod problem: to provide reliable, wear-resistant materials for moving parts at 1200°F.

An intensive literature search showed some materials, such as azurized steel or case hardened steel, were satisfactory for short periods, but a complete development program was required to determine performance over a long term. It was also necessary to examine the frictional principles involved; that is, should there be a soft material in contact with hard material, or should two hard materials be in contact. Ordinary lubricants could not be used. The only recommended lubricants are graphite or molybdenum disulphide. Numerous

samples of special alloys, coated, and treated materials were examined. After a compilation of the data, and mockup tests, it was decided that two hardened surfaces, of about Rockwell C-55 to C-60, were required. The MARK I final design resulted in a Haynes 25 cold-work race with Stellite No. 3 balls. The cold work is the key to the performance. The Haynes 25 race is ground to final size (early attempts to use Haynes 25 balls looked promising, but there was no process found to cold work the balls uniformly). A MARK II design is under development, in which the design allows the mechanisms to operate at lower temperatures and relieves the severe material problems.

The general problem of materials for the safety rod was not as severe. Rather than a dynamic system, as in the control rod, the safety rod has a static load over fairly long periods at high temperature. Thus, the main problem is the self-welding of the latch mechanism which releases the supporting rod. A ball-nut-screw arrangement is also employed, but it is not subject to conditions as severe as those for the control rod application. The initial attempts to solve the self-welding problem included surface treatments of the mating surfaces, such as azurizing, silver plating, and chrome plating. A final design used a recent development in tungsten carbide flame spraying. Satisfactory operation for periods of 500 hr and over have been demonstrated. In addition to the mating latch surfaces, a keyway is also designed which requires wear resistance at elevated temperatures. At first, some exotic materials were used, unsuccessfully, before it was discovered that a standard chromized drill rod proved satisfactory. A sketch of a typical safety rod is shown in Figure 17.

Now that we have examined the regulating and drive mechanism problems, let us review the control and safety materials themselves and the environment in which they operate. In both cases, the control material consists of centrifugally cast boron nickel sleeves containing 2 wt % normal boron. A typical analysis is shown in Table VI. No attempt was made to separate the boron isotopes to provide a higher cross section material. The boron nickel material represented a choice from many possible high cross section materials. It has probably the lowest cost for operation at high temperatures, but it is not without disadvantages. The main problem is the reaction between boron and neutrons; in simple form: $B + n \rightarrow He + Li$. This reaction is similar to the fission process in fuel materials, and results in gas evolution which causes

Figure 17. Safety Rod

TABLE VI
BORON NICKEL ANALYSIS

Element	%
Nickel	80 to 82
Chromium	10 to 12
Iron	4 to 5
Boron	1.5 to 2.5
Carbon	0.7 max.
Silicon	1.0 max.

some swelling. Very minute boron particles provide extensive surface area for the escape and diffusion of the gas atoms. If the boron is present in a ductile matrix in larger discrete particles, there is less surface for gas release and thus swelling is reduced. A powder-compacting technique would produce such an improved distribution. Other types of neutron absorbing materials are being examined, and will be incorporated in future tests.

IV. SODIUM COOLING SYSTEM MATERIALS

This third category of materials relates to components external to the reactor. The main components attached to the reactor core are as necessary to the successful operation of the nuclear power plant as is the reactor itself. This is true, both for quality of material and workmanship. Due to the expensive maintenance and the potential hazards associated with the failure of a component, the quality of ordinary commercially available hardware, such as valves and heat exchangers, must be raised to meet the more exacting requirements.

As may be deduced from the introductory remarks concerning a homogeneous container material system, the same sodium coolant system material is used wherever feasible, depending on the function. The valves, pumps, piping, hot and cold traps, heat exchangers, and the main reactor tank are all considered part of this sodium cooling system. The purposes of using a single material are to:

- 1) minimize the difference in physical properties, such as coefficient of expansion
- 2) reduce the welding of dissimilar materials
- 3) minimize effects of varying rates of corrosion by the coolant and erosion of the coolant by suspended particles
- 4) reduce total cost

The determining factor in the choice of these materials is the service condition. With the assumption of conservative physical property data, a maximum operating temperature of 1200°F for the sodium became the basic assumption for the choice of design and materials in this part of the system. However, it is important to note that other parts of the reactor core similarly control the peak sodium coolant temperature; namely, the fuel material, and moderator can materials. Thus, for the SRE, a 960°F maximum sodium temperature is used because of the assumed limitations of zirconium. However, the system external to the core can handle 1200°F sodium, providing suitable core components are developed. With the metallic fuels, it would be necessary, at the 1200°F sodium

temperature, to use zirconium alloy jackets, which are now only in the development stage. Nonmetallic fuels, such as oxides, may be free from this restraint. Zirconium alloy or stainless steel cans could perhaps be used for the moderator cladding. At this higher temperature, compatibility, diffusion (such as carburization), and self-welding become dominant factors which require intensive experimental determination.

A. MAIN TANK, PIPING, VALVES, AND PUMPS

A typical piping layout for an SGR is shown in Figure 18. All commercial sodium systems to date have used steel as the principal container material.

Figure 18. Sodium System

During the initial selection of steels for use in sodium systems, AISI Types 316, 347, 304 ELC, and 304 were considered for the SRE. Type 316 was eliminated, due to high cost and welding problems; also, it offered no advantages in corrosion. Type 347 was eliminated because of the weld-cracking problems which were under extensive research at the time (particularly the sigma-phase embrittling problem). When the SRE was being planned, Type 304 ELC was at

a premium price, with respect to Type 304. It also has a lower strength at high temperature. Therefore, Type 304 was the first choice. Types 304 ELC and 304 are now produced at comparable prices. Both have excellent welding characteristics.

Under today's conditions, the same choice of Type 304 would probably be made, because of superior high-temperature strength. Other steels being examined for use in SGR systems include ferritic steels of the Type A335, Grade P22. This is a 2% chromium, 1% molybdenum, ferritic steel. With this type, there are advantages of cost and of lower thermal stresses, due to a lower coefficient of expansion. This type would be a compromise on corrosion, but its strength is sufficient. The creep strength of these ferritic steels reach a maximum at about 2 to 3% chromium. Molybdenum adds the high temperature strength. The main parts of the SRE system use the Type 304 stainless steel, for the reasons given. In all cases, commercial quality specifications are used.

The welding aspect of the main piping complex is another important factor. All major piping is field-welded. In the case of Type 304 stainless steel, welding rod Type 308 is used with an inert gas root pass and shielded arc filler. Since this steel has excellent welding properties, it does not require the extensive preheating and postheating, as do the lower-alloy ferritic steels. In all welds, the inspection includes dye penetrant checks after each pass and X-ray on the finished section. Piping in the main system is Schedule 40, 6-in. diameter; and, in the auxiliilliary circuit, Schedule 40, 2-in. diameter. Flexibility to accomodate thermal expansions is achieved by bends.

In piping, tanks, valves, and pumps, the only materials which are not Type 304 are the Stellite plugs in the valves and the valve packing. The purpose of the Stellite plugs is to prevent self-welding. The valve designs incorporate either a sodium freeze seal or bellows seal. In the case of the freeze seal, the packing in an Inconel-Monel braid.

B. HOT AND COLD TRAPS

The use of hot and cold traps in the external piping system provides a means for controlling the oxygen content and some other impurities, such as nitrogen and hydrogen, in the sodium. As was described under the core

materials problems, zirconium is an excellent getter for the oxygen in the sodium. In the cold trap, shown in Figure 19, advantage is taken of the

Figure 19. Cold Trap

temperature dependence of sodium oxide and sodium hydride solubility in sodium, in order to remove these impurities at significant rates. By maintaining a volume of sodium, at a temperature slightly above the melting point, in contact somewhere with the bulk system, a low-temperature sink is provided for the impurities. The cold trap is cooled by toluene at its boiling point (231°F). This corresponds to the temperature at which the solubility of sodium oxide in sodium is at an acceptable level. The action of the trap is to precipitate sodium oxide and filter it out, by means of stainless steel wool. For example, assuming an initial oxide concentration of 100 ppm in the 52,000 lb of sodium used in the SRE primary loops, the two cold traps will reduce the oxide concentration to 10 ppm in 58 hr. These traps are available for maintenance. The anticipated hydrogen impurity level is much less than for oxygen, and has therefore not been as fully investigated.

The hot trap technique employs a different principle. This trap operates at 1200°F, a higher temperature than the normal sodium coolant temperature, and provides a large surface area of thin zirconium sheet which is exposed to the sodium. This is illustrated in Figure 20. A small stream of the primary

Figure 20. Sodium Hot Trap

sodium loop passes through the hot trap. The main part of the trap consists of inserts, approximately 1 ft in diameter and 6 in. high, stacked in a tube. The inserts are coils of 0.004-in. corrugated zirconium sheet. The total weight of zirconium in one unit is 320 lb.

C. HEAT EXCHANGERS

There are two types of heat exchangers used in the present SGR concept: intermediate, and secondary. The intermediate heat exchanger uses a shell and tube counterflow arrangement with a U-type design. The SRE intermediate heat exchangers use Type 304 stainless steel throughout, and are shown in

Figure 21. The secondary heat exchanger consists of an airblast, directed on Type 405 stainless steel fins, or the steam generator of the electric plant, as the case may be.

Figure 21. Intermediate Heat Exchanger

V. SUMMARY

The sodium graphite reactor materials problems are principally concerned with (a) shielding materials, (b) core materials, and (c) sodium cooling system materials. These are summarized as follows:

- a) Plain carbon steel is used in the thermal shield, as it provides adequate sodium corrosion resistance under low thermal stress conditions. Ordinary and heavy concrete serve as the principal biological shield materials, with design and cost the main factors.
- b) The most important materials problems in the SGR occur in the core. The sodium coolant requires high general purity, with emphasis on low oxygen content. The zirconium-canned moderator-reflector materials are subject to both external and internal reactants, in the form of oxygen, graphite, degassing products, radiation effects, and carburization. A sodium temperature limitation of 950°C, together with the present cellular design of the SRE, minimizes these interrelated effects. The use of metallic fuel elements at high central and surface temperatures may present limitations, due to radiation-induced swelling. Various alternate methods of design, and the use of nonmetallic fuel may provide greater flexibility. The use of cast boron-nickel control and safety rod materials is satisfactory for intermediate periods of use. More promising materials are being examined. The choice of materials for control and safety rod drive mechanisms that will operate at 1200°F for extended periods has presented many problems.
- c) All of the major sodium cooling system materials in the SRE use Type 304 stainless steel throughout, to provide a low cost, homogeneous system with good corrosion resistance. The hot trap in this system uses zirconium, at a high temperature, as an oxygen getter material. And the variation in solubility of sodium oxide in sodium is used, in the cold trap, as a means of minimizing oxygen concentration in the sodium.

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