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# CERAMICS FOR NUCLEAR APPLICATIONS

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## INTRODUCTION

Nuclear fission energy systems have been summarized in an earlier chapter. My purpose here is to discuss these various concepts in greater detail and to indicate where ceramic materials are used, delineating problems where those materials most probably hold solutions. Nuclear concepts discussed will be various types of reactors, both fission and fusion.

### Nuclear Fission Applications, History, and Projections

To discuss nuclear fission systems, present and future, and ceramic use in nuclear reactor technology, it is necessary first to review the mechanism by which power is obtained from the atom. The history of developments in nuclear physics before and after the first chain reaction in December 1942 are well presented by Smyth[1] and need not be given here.

The reader is probably familiar with the mechanisms of the fission processes in which energetic neutrons split heavy atoms. Additional neutrons are released as are a variety of intermediate size isotopes, electromagnetic radiation, and considerable energy. The additional neutrons sustain the process if their numbers are controlled at a steady state of production. The energy is converted to useful power within the limits of thermodynamic efficiency.

The fission waste and the electromagnetic radiations present problems of protection and material behavior.

Natural uranium contains 0.7% of the fissile isotope $^{235}U$ and 99.3% of $^{238}U$, which is nonfissile but fertile, that is, it can be used by converting it into plutonium, which is fissile.

There are two main categories of reactors: (1) thermal reactors, which use moderators to slow down the neutrons, and (2) fast reactors, which use no moderators.

In a thermal reactor, used by most nuclear power stations built so far, fissions are due to the absorption of slow neutrons. To slow down fast neutrons to thermal energies, a sufficient amount of moderator must be included in or around the core of the reactor. These thermal neutrons produce further $^{235}U$ fission necessary to start the chain reaction and also to convert some of the $^{238}U$ to plutonium. Flexibility in size is an advantage of thermal reactors.

In a fast reactor, the majority of fissions occur by interaction of the fissionable material with neutrons of high energy to "breed" plutonium because the fuel is enriched with $^{235}U$ or plutonium. A disadvantage of the fast reactor is the large amount of fissionable material necessary to attain a chain reaction, but because there is no moderator, the overall dimensions may be small. The Trinity, Nagasaki, and Hiroshima homogeneous reactors of 1945 were military weapons which released a vast amount of energy in an extremely short time.

Thermal reactors are classified either as homogeneous or heterogeneous. In a homogeneous reactor the fuel is dispersed uniformly, either in liquid or solid state, as an alloy or mixture with the moderator. In a heterogeneous system the solid fuel material is not an integral part of the moderator.

In a heterogeneous system, the solid fuel material is fabricated into various shapes (pellets, pins, etc.) which are usually clustered together in assemblies called fuel elements. The heat produced by the nuclear fission in power reactors is transferred, by a heat exchanger, to high-pressure steam used to drive a turbogenerator.

The possibilities for producing useful reactor-generated power for domestic use were first discussed by Fermi.[2]

Ceramic applications can be separated into various groups by their use in the reactor: (1) fuels, (2) ceramic control materials, (3) moderators, (4) reflectors, (5) Shielding, (6) radioactive wastes disposal, and (7) structural parts and ancillary uses.

## 1. Nuclear Fuels and Fuel Elements

A primary nuclear fuel is defined as a naturally occurring fissionable isotope. A secondary nuclear fuel is a fissionable isotope synthetically made by using the nuclear properties of a primary fuel.

Uranium-235 is the primary nuclear fuel. Of all the isotopes occurring in nature, this one is unique in that it undergoes fission with thermal neutrons and, in the fission process, liberates from one to three neutrons. Therefore, a chain-reacting system is possible.

Plutonium-239 is the most important of the secondary fuels produced by the reaction

$$ {}^{238}_{92}\mathrm{U} + {}^{1}_{0}\mathrm{n}\ \text{(neutron)} \rightarrow {}^{239}_{92}\mathrm{U} \xrightarrow[\text{23m}]{\beta^-} \underset{(\beta\text{-particle})}{{}^{239}_{93}\mathrm{Np}} \xrightarrow{\beta^-} \underset{(\beta\text{-particle})}{{}^{239}_{94}\mathrm{Pu}}. \quad (1)$$

Therefore, if a pile is made of natural uranium and a allowed to run for some time, a portion of the $^{238}$U will be converted to $^{239}$Pu, which possesses fission properties similar to $^{235}$U this then is the most important of the secondary nuclear fuels.

The other secondary fuel is $^{233}$U. This isotope can be made synthetically if $^{232}$Th is incorporated in the primary nuclear reactor as described for the production of plutonium. The reaction is as follows:

$$ {}^{232}_{90}\mathrm{Th} + {}^{1}_{0}\mathrm{n} \rightarrow {}^{233}_{90}\mathrm{Th} \xrightarrow{\beta^-} {}^{233}_{91}\mathrm{Pa} \xrightarrow{\beta^-} {}^{233}_{92}\mathrm{U}. \quad (2)$$

Inasmuch as the source of nuclear energy is within the atom itself and is independent of chemical combinations, nuclear fuels may be ceramic compounds, metal alloys, or enriched isotopes. Ceramics have the following advantages over metallic fuels: higher melting points; absence of phase changes (uranium metal has two such changes at 668 and 774°C accompanied by large volume changes); better resistance to coolant, particularly to liquid metals; and better resistance to neutron irradiation. Disadvantages of ceramics are their lower thermal conductivity, which gives rise to high internal temperatures, and their lower density of fissile atoms. Although several kinds of ceramic systems hold promise as fuel for power reactors, oxides, carbides, nitrides, and cermets have received the most attention and continue to be investigated.

Each fuel system has advantages and disadvantages and, ultimately, the service conditions dictate the choice of material. Fissile materials include uranium, thorium, and plutonium with uranium receiving the most attention. In the past 10 years, the use of plutonium as enrichment has received the most emphasis so that mixtures of uranium with plutonium, either for the oxide or carbide systems, are being investigated more earnestly. The primary fuel design considerations are dimensional stability, compatability, and reactions between core, cladding, and coolant; fuel relocation; solid and gaseous fission product accommodation; fission product and plutonium migration; fuel swelling and thermal expansion. All of the ceramic fuel materials are prone to the same kinds of irradiation effects and the basic problems are essentially the same for each fuel type. Breeder fuels must withstand extremely high neutron fluxes ($3 \times 10^{23}$ neutrons/cm$^2$) and prolonged stays in the reactor to achieve high burnups.

Uranium carbide or mixtures of uranium carbide and plutonium carbide or nitride fuels have application in liquid-metal-cooled thermal and fast reactors. In common with oxides, the carbides have the advantage of a high melting point and a good dimensional stability. However, the thermal conductivity of the carbides is 4 to 10 times higher than that of uranium dioxide or mixed oxide fuels. Uranium and plutonium nitrides are considered by many investigators as having the most promise as advanced fuels because they possess all of the advantages of the carbides and none of their disadvantages (no carbon transport problems, but higher thermal conductivity).[3] There are several ways to take advantage of the higher thermal conductivity property with fuels: larger diameter fuel rods, higher specific power operation, and lower fuel temperatures to decrease fuel redistribution, swelling, and fission gas release.

In the United States today, four principal methods are used to synthesize and fabricate uranium-plutonium carbide fuel for fast reactor applications. The United Nuclear Corporation synthesized powder by the carbothermic reduction of blended $UO_2$ and $PuO_2$. The resulting carbide powder is pressed and sintered into dense pellets by using a nickel sintering aid. Atomics International has investigated the the potential of "tungsten-doped" carbides. Their fabrication process is based on directly arc-casting carbide pins. At Westinghouse Atomic Reactor Division, mixed carbides are synthesized by arc melting, followed by powder processing to form pellets. In the Westinghouse concept, chromium carbide is added as a stabilizer for hyperstoichiometric fuel compositions. The process used by the Los Alamos Scientific Laboratory is based on direct combination of uranium, plutonium, and carbon in an arc melter, followed by conventional powder processing to form pellets.[4]

Each of these different processing methods results in a slightly different fuel composition, each having its own characteristics. The different processes selected by the various organizations reflect differences in opinion about the relative importance of carbide fuel fabrication economics, fuel swelling during irradiation, and fuel compatibility with the clad.

Compatibility of the fuel with the clad is an important reason for using carbide fuel. Stainless steel clads react with free uranium and plutonium to form low melting eutectics. For example, the $Pu-Pu_6Fe$ eutectic melts at 410°C. Therefore, significant amounts of metallic actinides must not be present in the fuel. Alternatively, hyperstoichiometric carbide fuels can seriously carburize the clad. For compatibility, a single-phase mono-carbide fuel is preferable, particularly for high performance applications that involve a sodium thermal bond.

The literature on nuclear fuels, fuel technology, and materials is the fastest growing section of this discipline and is too extensive to present in detail here. Ceramic nuclear fuel is among the fastest growing segments of the industry. It became a $73 million per year business in its first decade and is projected to exceed $540 million annually before the end of its second decade.

In reactor fuel elements, cermets find their major role in dispersion or matrix units. Uranium oxide, thorium oxide, or other fissionable or fertile materials are dispersed as pyrolytic carbon-coated microspheres in a continuous matrix of a nonfissile material having a low absorption cross section for neutrons, that is $UO_2$ in aluminum - U.S. Army Power Reactor APR-1.[5]

The concept of forming a refractory coating on fuel particles has attracted considerable attention. Advantages include preventing reaction between the fuel and the matrix or atmosphere, and minimizing the escape of fission fragments. Pyrolitic carbon coatings on $UO_2$, UC, $UC_2$, $UO_2$+$ThO_2$, and $UC_2$+$ThC_2$ particles dispersed in graphite have received extensive attention.[6] Such a concept is to be used in the high-temperature gas-cooled reactor, HTGR.

A unique method of producing solid solution (U,Zr)C powder for ultimate fabrication into dense monolithic carbide elements has been described by Schell.[7] The resulting complex cross section monolithic carbide elements contain only (U,Zr)C solid solution.

The first commercial nuclear electric power plant (60,000 kW) in the United States was built at Shippingport, Pennsylvania, near Pittsburgh in 1957. There are now 54 larger versions of this original plant producing 6% of our electricity. By the end of 1974, 59 plants are to be on line. It is believed that 140 nuclear plants will be operational by 1980, producing 20 to 25% of our power. By 1990, 45% of our power will be nuclear, and by 2000, 60%. Within the next 26 years,[8] 500 new breeder-type plants are projected. In the next decade, 100 conventional nuclear plants are planned to which the United States is committed at a cost of \$100 billion. Current goals, proposed by the USAEC, are to spend \$10 billion for federal energy research and \$5.1 billion to develop a breeder reactor for power plants. President Nixon stated in his Energy Message to Congress on June 4, 1971, that his program included "a commitment to complete the successful demonstration of the liquid-metal fast breeder reactor by 1980." If these predictions prove accurate, the United States will need 33 times the uranium enrichment capacity it now has by the end of this century, and will need astronomical increases in our capacity for fuel recycling.

In the belief that our supply of uranium could not stretch much beyond the year 2000 unless we used a breeder reactor with a 40-fold fuel economy,* work began in the 1960's to develop a large-scale model that would produce power. The first fast reactor produced ~200 kW in December 1951 near Arco, Idaho, EBR-1.

The reactors in commercial service today use <1% of the energy in naturally occurring uranium (thermodynamic efficiency ~32%). Heavy water reactors produce about twice as much electricity/kilogram of uranium as light water reactors. There is considerable debate as to the extent of our uranium reserves (present extraction cost is ~$8/1000kg an increase to $15/1000kg would only raise the price of a one-half cent). This most increase could extend our needs by using light water reactors to the year 2100, thereby giving us more time to develop the breeder.

There are three promising types of breeders: the LMFBR, a liquid-metal-cooled breeder, the MSBR molten salt breeder, and the gas cooled breeder, GCFR.

The breeder offers other advantages besides ensuring a much longer supply of nuclear fuel, projected to double our plutonium inventory every 10 to 15 years or by 80,000 kg by the end of the century. The breeder is more efficient than the conventional reactor (~40% thermodynamic efficiency). and converts more of the nuclear heat to electricity. Therefore, it produces less heat loss and less radioactive waste; the waste being difficult and dangerous to dispose of.[9] Also the breeder operates at much lower pressure so there are fewer chances for leakages of radioactive gases.

Many foreign countries have breeder-reactor programs. Because of their past experience and present activities the United Kingdom (Dounreay Prototype Fast Reactor 250,000 kW electric), France (Phoenix - 250,000 kW electric), West Germany (KNK and SRN - 300,000 kW electric), and the USSR (BN-350- 150,000 and BN-600- 600,000 kW electric) are considered the leading nations, other than the United States, in breeder reactor development.[10]

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*The $^{238}$U in the reactor captures neutrons becoming $^{239}$U as indicated in Eq. (1) on page 4.

Cumulative fuel cycle costs, currently \$360 million annually, are estimated to total \$14 billion by 1980; the annual expenditure being \$2.4 billion; accumulated sales of uranium ore, approximately \$6.7 billion; uranium enrichment in the AEC's diffusion plants, approximately \$600 million annually with the potential for an additional \$400 million in the rest of the free world.

In the fuel-cycle phase where enriched uranium is fabricated into the finished reactor core, annual fabricating costs for 1970 were approximately \$82 million, and \$100 million in 1972, with \$200 million projected for 1980, for only initial fuel supplied. Annual replacement fuel sales should be about twice that of the initial fuel, or up to nearly \$300 million by 1980 from a 1970 value of \$5 million. Nuclear power plants have a 30-year life expectancy, whereas a fuel charge has a 3-4-year life.

The 1971 requirement for uranium concentrate was 8200 kg (9,000 tons) valued at \$144 million. This should rise to an annual $30.9 \times 10^3$ kg (34,000-ton) requirement valued at \$544 million by 1980 with the cumulative total being $189 \times 10^3$ kg (208,000 tons) valued at \$3.3 billion.[8]

## 2. Ceramic Control Materials

Control materials are used in nuclear reactors to absorb neutrons and to control reactivity. Their primary requirement, therefore, is that they have a high absorption cross section $\sigma_a$, for neutrons. The fact that control materials have high cross sections means that they are highly susceptible to reaction and potential damage from neutrons. Such materials in the form of rods, when placed within a reactor core, can completely stop a chain reaction by capturing the neutrons and making them unavailable to the fuel.

For many years a principal control material has been the alloy of cadmium-indium-silver. In the past two years, the cost of this alloy has risen ten-fold intensifying the fabricators' search for cheaper replacements ($B_4C$, pyrohafnates, and rare earth mixtures). Although they have effective cross sections, their significantly lower density is cause for concern because of their slower drop rate which results in increased "scram times."[11]

In addition to alloys, dispersions (such as $B_4C$ in aluminium or boron in graphite) and as a monolithic ceramic ($B_4C$ hot pressed pellets) are utilized in control rods. The monolithic or pure ceramic materials have little structural strength and must be supported by the control rod components. Current designs have pellets in water-cooled stainless steel tubes or tubes filled by vibrational compaction of sized $B_4C$ powders.

Desirable properties of control materials include high cross section for neutron cross section, dimensional stability at high temperature, good resistance to radiation damage, ease of fabrication, mechanical stability, and decay after neutron absorption to a daughter material also possessing a high cross section for neutron absorption.

Currently, the limiting factor in control rods in the fast flux test facility (FRTF) is the tendency for the 316 stainless steel tubes that contain the control material to bow as a result of irradiation damage in the steel. The bowing is usually limited to $\sim$100 full-power days $\sim$3mm in $\sim$2m before replacement must be made.[12] The swelling of the $B_4C$ pellets has been accommodated by allowing a diametral gap between the pellet and the tube wall. The helium gas evolved by the n,$\alpha$ reaction has also been successfully accommodated by providing a plenum. $B_4C$ is able to retain a high percentage of the generated helium, however.[13]

Table I lists common forms in which ceramic materials are used.

TABLE I[14]

COMMON CERAMIC CONTROL MATERIALS

| Material | Melting Point | Macroscopic Cross Section | May be Used |
|---|---|---|---|
| $B_4C$ | 2450°C | 83.6cm$^{-1}$ | in Al |
| $B^{10}_4C$ | 2450°C | 443.8 cm$^{-1}$ | in SS<br>in graphite |
| $HfO_2$ | 2130°C | 2.9 cm$^{-1}$ | in Zr,Ti,SS |
| $Gd_2O_3$ | 2330°C | 1134.4 cm$^{-1}$ | in Zr,Ti,SS |

| | | | |
|---|---|---|---|
| $Eu_2O_3$ | 2050°C | 116.8 $cm^{-1}$ | in Zr,Ti,SS |
| $Sm_2O_3$ | 2300°C | 141.1 $cm^{-1}$ | in Zr,Ti,SS |

Ceramic burnable poisons are similar to ceramic controls, but, as the name implies, they are designed to burn out at the same rate as the fuel to maintain a constant reactivity of the core. The tendency of reactor design is to have long core life between reloadings. Unless some of the reactivity caused by $^{235}U$ loading is counterbalanced by burnable poisons, reactor control becomes difficult. To achieve a 10,000-life, or even 5000-h, the core should contain some form of burnable poison.[15]

The most common element used for this purpose is boron 10, which has a 4020-b cross section and occurs in natural boron as about 20% of the total boron. $B_4C$ will retain the burnout products of lithium and helium and has good thermal stability to it's 2400 ± 50°C melting point. It reacts with water at >400°C, but can be lowered by reducing the surface area, that is, hot pressed $B_4C$ >95% theoretical density has good resistance to 2000 psi water at @420°C, and can withstand a burnup of 15 at.% before physical degradation begins.[16] Other boron compounds that can be used as burnable poisons are ZrB, $ZrB_2$, $ZrB_{12}$, chromium borides, calcium borides, $UB_4$-$CeB_4$ solid solution, BN, and the rare earth o oxides of europium, samarium, gadolinium, and dysprosium.

## 3. Moderators

An integral part of a thermal reactor is the moderator, which is used to slow down the neutrons given off by $^{235}U$ so that the chain reaction can continue more efficiently. Slow neutrons are much more effective in causing $^{235}U$ to fission than are fast ones. The moderator is usually a lightweight material having atoms which neutrons can hit, causing the neutrons to lose part of their energy. The requirements for moderators are (1) that the material has a low-neutron capture cross section, so that when neutrons interact with the atoms of the moderator there is little probability that they will be captured and (2) that there be a comparatively large decrease in the neutron energy from about 1 MeV to about 0.025 eV per collision with the moderator atoms, which measures the efficiency of the material. Light elements are better, particularly beryllium, carbon, oxygen, hydrogen and deuterium oxide (heavy water). The advantage of heavy water for low-temperature reactors is obvious, whereas high-temperature reactors make use of purified graphite or beryllium oxides. In general, graphite is favored because it has better thermal conductivity at higher temperatures and is second only to beryllium oxide in its moderating ability. Care must be taken in fabrication to produce a random crystal orientation because individual crystals have a marked anisotropy in many properties. Thermal expansion is a particularly significant factor because unequal dimensional changes can disrupt close structural tolerances. By virtue of its use, a moderator is subjected to severe radiation damage; this factor must be considered in design.

## 4. Reflectors

Materials used for reflectors must possess a highly scattering cross section, $\sigma_s$, over a broad neutron spectrum. Reflectors are designed to conserve neutrons by reflecting them back into the core, thereby preventing escape and, in turn, cutting down the shielding requirements. A good moderator is a good reflector. Because $\sigma_s$ will vary with neutron energy, different materials are required for neutrons of various energies. For fast reactors, tungsten carbide or thorium oxide might be used, whereas for thermal neutrons a wide variety of compounds may be used; such as the alkaline earth oxides; stabilized zirconia and zirconates of thorium, beryllium, calcium, and magnesium. Norton lists a large number of other high melting point compounds suitable for thermal neutrons.[17]

## 5. Shielding

Neutron shielding is best achieved by materials with high absorption cross sections, particularly if there is a space limitation where the classic, but relatively expensive, poison and control materials are used. In land based tests, or power reactors where there are no shielding mass, limitations, concrete filled with high neutron absorbing materials (such as baryte, iron ore, colemanite, and ferro phosphates) is generally used. Gamma radiation is best absorbed by dense materials of heavy atoms such as lead or steel.

Glass with densities up to 621 $kg/m^3$ (388 $lb/ft^3$) have been developed that accomplish the double task of permitting visibility while absorbing high-energy electromagnetic radiation to protect observers. These glasses contain a maximum of the heavy elements, that is, $PbO$, $Ta_2O_5$, $Nb_2O_5$, $WO_3$, etc. Shielding glasses are generally supplied in three types: a very high-density lead glass with a correspondingly high absorption coefficient (some 27.9 $mm^{-1}$ for 1.0 MeV gammas), an intermediate density lead glass with a still significant absorption coefficient, and a cover glass which is essentially a plate-glass composition with low absorption coefficients near those of aluminum. The glasses are generally cerium-protected against radiation coloration. The amount of cerium present determines the extent of protection against irradiation coloration and is limited by the yellow color it imparts to the glass.[18] To minimize reflection between the multiple glass layers, the gap between them is generally oil filled.

In cases where close inspection or microscopic examination is necessary in a high radiation environment, shielding glass cannot solve the problem of personnel protection, and periscope or microscope-periscope combinations are used so that personnel can operate from behind protective walls. Again, the glass optics have been especially formulated using cerium to prevent coloration without causing an objectionable amount of intrinsic yellow color. Cerium is added in sufficient quantity to protect glasses from coloration at the $10^6$-R target does.

## 6. Radioactive Waste Disposal

A vexing problem in nuclear technology is the disposal of atomic wastes because many of the by-products of nuclear fission have exceedingly long half-lives (28 y for $^{90}Sr$ and 24,300 y for $^{239}Pu$). The annual United States generation of 64.3 $m^3$ (17,000 gallons) of highly radioactive wastes is expected to jump to 3785 $m^3$ (1 million) gallons by 1980 and $17.4 \times 10^3 m^3$ (4.6 million gallons) by the turn of the century The stored (an interim measure) volume of liquid wastes by 1980 may reach 3785 to 37,850 $m^3$ (1 to 10 million gallons). This waste will contain >10 billion curies of fission products.[19] Ceramic technology has played a significant role in investigating solutions to these problems. Safe disposal requires that wastes be converted to inert solids and placed in repositories where long-term isolation with minimum surveillance can be assured.

The major source of high activity wastes is the Purex method of solvent extraction fuel reprocessing used in the United States, England, France, and elsewhere and consists essentially of nitrate salts in nitric acid (usually ∿15 elements appear in significant concentrations).

Solid form disposal techniques receiving extensive attention are calcines prepared in such equipment as fluidized bed, pot, or spray calciners, and ceramic forms that can be prepared by several techniques. The best developed processes for converting liquid waste to ceramic forms are[20]

(1) Phosphate glass. Orthophosphoric acid is added to acqueous waste; 90% of the liquid is evaporated, fed continuously into a platinum melter, converted into glass at 1100 to 1150°C and poured into mild steel storage pots Brookhaven National Laboratory (BNL).

(2) Spray solidification. Chemical additives and orthophosphoric acid are added to waste which is then dewatered and denitrated in a spray calciner. The output is continuously fed into a platinum melter at 1050°C and the resulting glass is poured into mild steel storage pots Battelle Northwest Laboratories (BNW).

(3) In-pot melting. Calcined waste is mixed with a borosilicate glass frit and fused directly in 18-8 stainless steel pots at 900°C (BNW).[21]

(4) Fingal and rising level glass. Waste and a silica-borax slurry are fed continuously into a 25-20 stainless steel pot in which evaporation, denitration, and fusion to a glass at 900-1000°C occur simultaneously in separate zones. Slurry is added to glass until the glass level reaches the top of the pot when feed is stopped, the pot is stopped, the pot is heat treated, and se led for storate. Oak Ridge National Laboratory (ORNL) England.

(5) Pot Vitrification. Waste and chemical additives are sequentially evaporated, calcined, and melted in Inconel melter at 1100-1150°C and drained into a mild steel pot for storage. The Inconel melter is replaced after 20 cycles and is used as a storage container when the final processing cycle is completed (France).

Natural high exchange clays, such as montmorillonite, have also been used to absorb the alkaline earth fission products onto their surface. After all available exchange sites have been exhausted, the clay is heated to 900°C for fixation. This process has been largely supplanted by the aforementioned processes. Sorption on synthetic zeolites is also being investigated. A process where various molten salts melting in the 500-to-800°C range are used as oxidizers to decompose, combust, and solidify radioactively contaminated carbonaceous wastes for disposal is also being investigated at the Battelle Northwest Laboratories and by the Atomics International Division of North American Rockwell.

Entrapment in other materials, such as concrete, tar, and sulfur, is also employed for low-level wastes.

## 7. Structural Parts and Ancillary Uses

### a. Structural parts

Though the discussion so far has concentrated on the materials integral to neutron production, there are many other uses for ceramics in a nuclear power plant. Alumina is used for many structural parts, such as chemical plumbing for pump plungers, seals, and glands; electrical and thermal insulation as thermocouple protection tubes; fiber insulating blankets for in-core duct work (in HTGR); piezoelectric and ferroelectric ceramics for transducer gauges, etc.; ceramic-metal seals for electrical feed-throughs; porcelain-enameled wire for electric motor windings, etc. All of these applications would be under heavy irradiation and high temperature, and subject to possible coolant penetration. Ceramics play the key role in these applications because their uses will play an important part in the development of a high-performance breeder.

### b. Ancillary Uses

Dosimeters. The change in the optical properties (absorption and fluorescence) of glass with irradiation is used to measure radiation dosage. Dosimeters are used for personnel protection in nuclear industry, in situ dosimetry in cancer radiotherapy, etc. Special glass compositions are also used in Cerenko counters for high-energy spectrometry, scintillators for neutron detection, and secondary neutron standards.

## Nuclear Metallurgy

Ceramic refractory materials are as essential to the nuclear metallurgy industry as they are to the steel industry, and are used widely.

## Metal Production

The first gram quantity of plutonium metal was produced by Baker[22] in April 1944 by the $PuF_4$ bomb reduction. The process used the thermite reaction of a metal halide (calcium) with $PuF_4$, boosted by $I_2$ (it makes more fluid slag) in a rammed fused magnesite crucible fired at 2200°C in a graphite mold. The reaction takes off at 975°C in an argon atmosphere leaving the molten metal as a button free of slag in the bottom of the crucible. The bomb reduction of $UF_4$ using calcium or magnesium in an identical reaction was developed at the Ames Laboratory, also using Mg bomb liners.[23,24]

Magnesia has been replaced by $MgF_2$ recovered as a by-product of the reduction in large scale metal production.[27] Reductions are carried out on a large scale (up to $1.4 \times 10^3$ kg or 1.5 tons) to yield large castings that do not require remelting before further fabrication.

Most of the molds used to vacuum-cast uranium and uranium alloys are machined from electrode grade graphite. Because the molten metal tends to react to form carbides and pick up free carbon from the graphite, it has been necessary to develop coatings to prevent these reactions. Coatings presently in use, depending on the metals to be alloyed, are magnesia, alumina, zircon, zirconia, and combinations of these compounds. The coatings may be applied to the crucible/mold surfaces by brushing or cold spraying, and then baked or fired before use or by flame or plasma spraying to eliminate the prefiring step. In connection with these foundry applications, molds, stopper rods, covers, and impingement plates, thermocouple sheaths and other insulator spacers are also used. Alumina, magnesia, zirconia, calcia, thoria, urania, and calcium fluoride are also used as melt containers and molds for various nuclear metals and their alloys.[28]

High-purity plutonium production by electrowinning or electrorefining was developed by using ceramic refining cells. The cells contain the molten salts of NaCl, KCl and 2 mol % of $PuCl_2$, $PuF_3$, or $PuF_4$ at temperatures up to 850°C and must therefore have high thermodynamic stability to minimize the possibility of reaction between the container (cell) and the molten salts, or the molten plutonium, or both. In addition to the cells, stirrers and anode sheaths are also required of ceramic. Initially, impervious $Al_2O_3$ cells were used but eventually were supplanted by an impervious MgO developed at Los Alamos. The plutonium produced using these ceramics[29] is the highest purity ever developed and is currently used by the National Bureau of Standards as a certified analytical standard. Only plutonium of this purity can be considered as fuels for implanted biomedical aids such as heart pacers.

Although advanced alloys of other metals offer a continued challenge for refractory melt containers and molds, there are few requirements not being met by ceramic refractory materials. In uranium production no material has yet been developed that is free of carbon, inducts, causing minimum total contamination to the melt. A mold material, cheaper than graphite, unreactive with uranium, and capable of being produced to close tolerances has not yet been developed.

## Direct Energy Conversion

In this concept a self-heating emitter of a fissionable material ionizes a vapor, usually cesium, causing a flow of electrons in the plasma between the hot emitter and the cooler collector, resulting in an open circuit voltage. High purity, impervious alumina provides the insulator between the collector and the base. Requirements for this development have resulted in irradiation resistant ceramic-to-metal seal combinations capable of operating continuously at $1400^\circ$C in the extremely corrosive cesium plasma.

## Nuclear Fusion Applications

Although fission reactor systems can be used to supply the world's energy needs for centuries, they can do so only at the expense of creating vast amounts of radioactivity, some of which could escape into the environment. They also introduce the threat of catastrophic accidents. The fusion process seems most likely to meet all practical, economical, and environmental requirements because it uses cheap, inexhaustible fuel,[illegible] has no radioactive waste products, produces no chemical pollution, and has high thermodynamic efficiency. Two versions of fusion power generation drive this field: magnetic confinement and laser-pellet concepts.

It is generally agreed by the entire scientific community engaged in fusion reactor studies that there exist two major areas with large uncertainties confronting the fusion program: plasma physics and materials technology.[30]

Schematic diagrams of plasma confinement and laser-initiated nuclear fusion systems arerepresented in this book as Figs. 5A and B in the chapter on "Ceramics in Advanced Energy Systems" by Johnson, together with an explanation of their operation, and will not be repeated in this chapter.

The four systems being actively studied today are (1) theta pinch, (2) Tokamak, (3) mirror, and (4) laser. The laboratories investigating these systems and problem areas where ceramics may provide solutions are shown in Table II. Laboratories other than those listed, are also involved in various aspects of insulator research and development: Brookhaven National Laboratory, Atomics International, Battelle Northwest Laboratory and Argonne National Laboratory.[31]

It is difficult to specify exact requirements for insulators to be used in these applications because the conceptual designs are in a more or less constant state of change, and because the designs can be adapted, to some degree, to match the characteristics of available insulators. Guideline environmental conditions of operation, geometric description, electrical requirements, desired lifetime, etc., of insulators in the theta pinch and mirror systems have been described in detail by Krakowski and Clinard[32] and by Van Konynenburg,[33] and are too extensive to be treated in this discussion A more general description of insulator requirements for the three systems, excluding lasers, has been given by Bates[34] and is recommended for further reading.

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* Uses heavy forms of hydrogen, deuterium, and tritium. Tritium can be "bred" in thereactor by neutrons produced in the fusion process. The deuterium comes from water - a gallon of which contains 0.123g (1/230th of an ounce), which costs 4¢ to extract.

## TABLE II

### FUSION SYSTEMS, STUDY LOCATIONS, AND CERAMIC APPLICATIONS

| System | Investigating Agency | Ceramic Applications for Current Conceptual Designs |
|---|---|---|
| Theta Pinch, RTPR | Los Alamos Scientific Lab. | First wall insulator, inner blanket insulation, implosion heating coil insulation, compression coil insulation. |
| Tokamak | Oak Ridge National Lab; Princeton Plasma Physics Lab.; Univ. of Wisconsin; Overseas Labs. | Insulator baffles blanket. Neutral beam injector. Magnet & power lead insulator. |
| Mirror | Lawrence Livermore Lab. | Beam injector insulator. Injector power supply. Superconducting magnet insulator. Direct converters. Coolant channel insulation (optional). |
| Laser | Los Alamos Scientific Lab. | Laser Windows |
| | Lawrence Livermore Lab. | Target Materials |
| | KMS Fusion Lab. | |
| | Overseas Lab, | Laser Windows |

Proposed ceramics now being investigated for these applications are alumina of various purities, quartz, SiC, $Si_3N_4$, porcelain enamels, and cermets.

In the laser-induced fusion concept,[35] many materials problems are similar to those of the confined plasma systems relative to containment, insulation, etc. Additionally, however, the megajoule powered laser beam poses difficult window requirements, in dielectric breakdown, optical absorption and reflection, elimination of internal defects, surface finish, etc. Nd-glass and alkali halide windows are being investigated, but as expected, almost all of these developments are of a proprietary nature and are difficult to document. Ceramic materials and technology will doubtless be used in the target pellets about which practically no information is currently available. Much of the information developed on ceramic materials used in fission systems will be applicable to fusion systems, but many of the projected requirements will need considerable extension of the state-of-the-art.

## Conclusion

The purpose of this review is to indicate the wide variety of applications for ceramic materials in the nuclear industry. The ceramist scientist or engineer needs to understand that dramatic transitions are occurring in the nuclear energy field and he needs to understand the challenges they present. The ceramist's creative and inventive instinct is essential to the resolution of the problems associated with nuclear energy research and development. Many more ceramists should be at the front of this challenge. They must, and will, play a major role in establishing the deep and detailed technical understanding of the ceramic materials on which the economic future of nuclear power rests. The tasks are difficult and complex, requiring unprecedented interdisciplinary technological efforts and disciplined sound engineering developments to assure success.

The difficult challenge of realizing the full potential of nuclear energy for the betterment of mankind lies ahead of us. The breakthrough of technological barriers separating us from essentially limitless supplies of energy demands all the initiative, strength, and capabilities we can muster.

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