Gas-cooled Reactors of a Mixed Fuel-Moderator Type

By G. C. J. Dalton*

Reactors of this type may use any of the moderators, graphite, beryllium oxide or beryllium, and one of the coolant gases hydrogen, helium, carbon dioxide, nitrogen or other suitable gas to be selected on the grounds of compatibility with the moderator and circuit materials and engineering suitability. The uranium233-thorium fuel cycle is the most suitable because the conversion factor possible with this system is greater than with the plutonium-uranium238 cycle. Because there is no necessity for lumping the fuel, the moderator may be used to dilute the fuel and to increase the surface available for heat transfer to the coolant. This dilution should also improve the burn-up possible before the fuel element suffers physical disintegration. The fuel and fertile material may be mixed uniformly with the moderator, or may be in a thin, enriched layer near the surface of the element. The fuel elements may be rods, spheres, plates, or bricks, pierced by coolant channels. Research will be directed towards the production of a high integrity fuel element, the solution of problems of compatibility of gas and moderator, control of fission products and reprocessing and refabricating fuel elements. It is expected that this type of reactor will have particular application in the power range of 5 to 50MW.

INTRODUCTION

Mixed fuel-moderator reactors have been discussed for many years, but so far none has been built. The advantages of the system are the dilution of the fuel with moderator to give a high heat rating, and the prospect of a burn-up of several times the initial fuel investment before radiation causes structural damage to the fuel element.

Proposals have ranged from homogeneous mixtures of all the fuel and all the moderator, to systems in which just enough moderator is mixed with the fuel to give an adequate heat transfer surface. Arrangements include systems of fuel elements in the form of spheres, cylinders, or plates, with the fuel distribution varying from homogeneous mixtures to arrangements with the fuel in the form of highly enriched zones near the surface of the fuel element. Daniels (1956) proposed a system consisting of blocks of graphite moderator containing fuel plugs and separate holes for the helium coolant.

Probably the main reasons why such reactors have not yet been built include the fact that the only moderator available in quantity has been porous graphite, and this has led to a coolant circuit contaminated by fission products. The problems and safety considerations associated with this have been sufficient to put several of such schemes on the shelf for future reference. Another technical problem which has appeared very formidable is that of changing fuel elements in this type of reactor.

Unfortunately, much of the work being done in the U.K. and in the U.S. is covered by security, and it is therefore not possible to discuss this type of reactor, except in very general terms.

FACTORS INVOLVED

Moderators

The ideal moderator should be impermeable to fission product gases, compatible with a suitable coolant gas, and not susceptible to radiation damage. It should also retain its strength at high temperatures, be resistant to thermal stresses, and have good nuclear properties. It

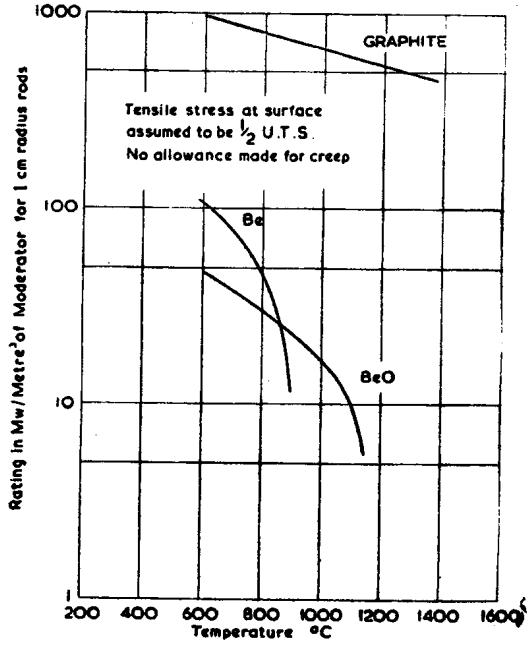

FIGURE 1: Comparison of moderators for mixed fuel moderator systems on the basis of thermal stress.

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should also be cheap and easy to fabricate. From the processing point of view, it should be easily separated from fission products and, unless very cheap, easily reconstituted.

In fact, the three moderators, graphite, beryllia and beryllium metal are all far from the ideal. Graphite has many excellent high temperature properties but unfortunately is not impermeable to fission product gases. Beryllia is also slightly porous, and only beryllium offers the possibility of complete containment of fission products.

To be resistant to thermal stress a material must have any or all of the following properties: a very low thermal expansion coefficient, a very low elastic coefficient, an exceptionally high thermal conductivity.

Figure 1 shows that graphite has much better thermal stress resistance than beryllia or beryllium, because graphite has a very low elastic coefficient.

Graphite and beryllia exhibit very little creep. Beryllium has some ductility at certain temperatures, but in general all these moderators must be treated as brittle materials.

Fabrication is difficult for all three materials. Graphite is still difficult to join, and beryllia can be fabricated only in relatively simple shapes of sizes of the order of linear length of 10cm. Beryllium can be formed by powder metallurgy into useful fuel coverings, but must be regarded as a difficult material to work. The toxicity of beryllia and beryllium add to the difficulties of their fabrication.

Estimates of costs are graphite £600/ton, beryllia £20,000/ton and beryllium £80,000/ton. One may expect the cost of beryllium and beryllia to come down, but it is clear that these materials must be used sparingly, and that recovery after processing is necessary. Fortunately (Figure 2) the size of a beryllia or beryllium reactor of conversion factor near 1.0 is very much smaller than that of equivalent graphite piles.

Mixtures of moderator materials may also be considered, e.g., graphite giving a strong structure, containing plugs of beryllia with the fuel mixed with either moderator or in a separate zone.

All these moderator materials should be stable under neutron irradiation, although there is some doubt about beryllium above 600°C. The (n, 2n) and (n,α) reactions produce helium within the beryllium and may cause damage. At lower temperatures, this does not appear to be serious. Samples from the reflector of the United States M.T.R. reactor showed no damage after an irradiation of $\sim 10^{21}$ nvt. fast neutrons. Because of the dependence on energy of the neutrons—(n, 2n) threshold $\sim$ 1.85 MeV, (n, α) threshold $\sim$ 0.7 MeV—this evidence must be treated with caution.

Wigner effects in graphite are not expected to be serious above 600°C but at the cold end of the reactor must be allowed for. The equivalent beryllia effects are not so well known.

At room temperature the thermal conductivity of graphite is reduced by a factor of 30 by neutron irradiation, but this effect is greatly reduced at 150°C (to a factor of 4) and has largely disappeared at 600°C. The thermal conductivity of beryllia and beryllium is apparently not greatly affected by irradiation. The thermal conductivity of beryllia decreases considerably with temperature.

Neutron economy

Although graphite has a much lower absorption cross section (3.8 millibarns, as compared with $\sim$ 10 mb for beryllia and beryllium) beryllium is a more efficient moderator, has a higher scattering cross section (6b compared with 4.8b for graphite) and has a higher density of atoms per cubic centimetre. Consequently, graphite systems have a much greater

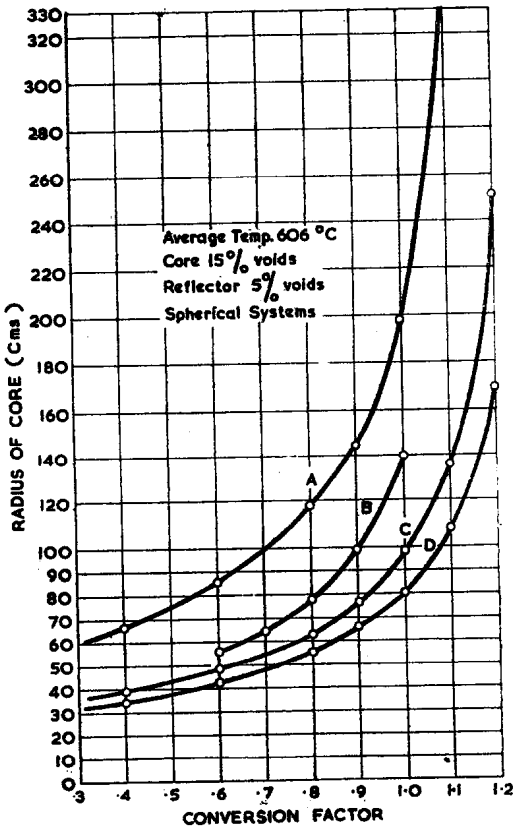

FIGURE 2: Comparison of core sizes for various moderators.

Allowance for poisons, 0.05 x absorption in U233.

Allowance for burn-up control, 0.05 x absorption in U233.

A—Graphite R = 4,000; graphite reflector.

B—Equal volumes BeO and graphite. U:BeO: graphite = 1:1520:1710. Graphite reflector.

C—U:BeO = 1:2400. BeO reflector.

D—U:Be = 1:3000. BeO reflector.

physical size than beryllia or beryllium systems of the same conversion factor. This may not be important for very large systems of power greater than 200 MW, but for systems in the 5-50 MW range the graphite systems are inconveniently large (Figure 2). The curve for a mixed beryllia-graphite system shows a considerable reduction in size from the pure graphite system.

In addition to the greater compactness as a moderator beryllium and beryllia offer the possibility of neutron "enhancement" by the (n, 2n) effect. This "enhancement"—defined as the number of neutrons reaching thermal energies less the number of fission neutrons—may be as high as 8 per cent. for beryllium metal.* This "enhancement" is over and above the losses due to the (n,α) reaction in beryllium. This latter reaction produces lithium 6, which has a high cross-section and which therefore may be assumed to absorb one neutron for every lithium 6 atom produced. If this estimate proves correct, the neutron "enhancement" in beryllium and beryllia systems will have a considerable effect on the conversion factors. Systems of only 6ft. in core diameter with conversion factors of up to 1.1 or 1.2 may be possible.

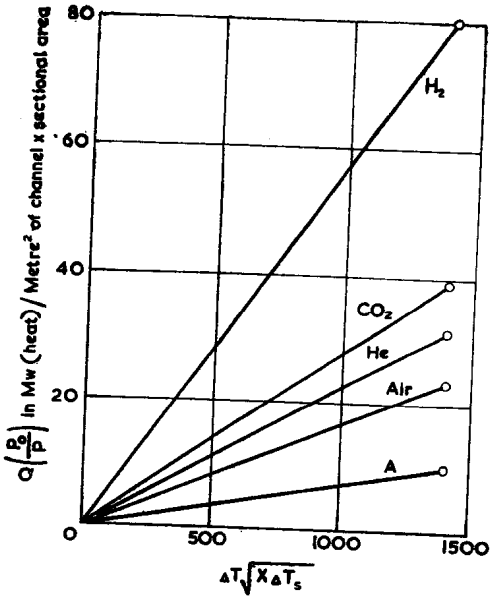

FIGURE 3: Comparative cooling properties of gases at 600°C. (Note that Q is not sensitive to absolute temperature.)

Q = heat output in MW (heat) sq. metre of channel x sectional area.
P_o = Atmospheric pressure.
P = Mean pressure of coolant gas.
ΔT = Temperature rise of gas across core.
ΔT_s = Surface temperature - gas temperature (heat transfer).
X = Pumping power used in core / Heat output of core

*I am indebted to Dr. J. Thompson, of the A.A.E.C. Research Establishment, for this figure.

The stability of the moderator which is producing helium at a rate of about 0.8 atoms helium per fission is a matter requiring experimental investigation.

The most attractive fuel cycle is the U233-thorium cycle. The high η value and its constancy with rise in temperature offer better neutron economy than the Pu239-U238 cycle. This latter cycle allows only a poor conversion factor because of the rapid increase in the proportion of non-fission capture of plutonium with temperature. Nevertheless, if plutonium becomes very plentiful as a result of building large natural uranium power stations, plutonium fuel may become available to be burnt in low conversion factor systems.

Coolants

Figure 3 shows the relative merits of coolants from a purely heat removal point of view.

The best coolant from an engineering point of view depends in a rather complex manner on the operating temperatures, the relative merits of total heat output and plant size and complexity, and whether the system has an isolated primary circuit or a single closed cycle gas turbine system. Fortescue (1953) gives the relative bulk of circulating equipment for a constant ratio of heat output/pumping power as:

Gas	Relative circulator volume (isolated primary circuit)	Total heat output
Helium	1	1
Carbon dioxide	.13	1.39
Nitrogen	.11	.78
Hydrogen	5.3	2.64
Argon	.033	.33

For a combined coolant-gas turbine system the heavier gases are preferable, because of their smaller bulk for a given power.

Hydrogen is clearly the most effective coolant, but has the disadvantage of requiring very large circulating equipment. Argon is a poor coolant, and gives only a limited heat output from the reactor, but has advantages in the matter of turbine size over hydrogen and helium. Carbon dioxide, with better heat removal than helium and only one-tenth of the circulator plant size, is an excellent choice when permitted by compatibility and corrosion criteria.

Nevertheless, because many projects have foundered on the problems of corrosion and coolant stability, it is probable that the final choice will be made on stability and compatibility grounds rather than on heat removal. Helium is clearly the most attractive from this point of view. Its disadvantages are its availability, cost, and the bulk of circulating equipment.

Compatibility and stability

From information available—

Carbon dioxide appears to be satisfactory with graphite up to 500°C.; with beryllia to

1200°C (probably no limit if dry); with beryllium up to 600°C.

Hydrogen appears to be satisfactory with graphite up to 900°C; with beryllia to 1200°C in absence of radiation (probably no limit if dry); with beryllium up to 900°C.

Nitrogen appears to be satisfactory with graphite up to 2500°C; with beryllia at least to 1000°C (probably no problem); with beryllium up to 600°C.

Air appears to be satisfactory with graphite up to 350°C; with beryllia up to 1500°C; with beryllium up to 500°C.

Activity of coolant gases

Hydrogen, helium, carbon dioxide, oxygen do not become significantly active.

Nitrogen has an (n,p) reaction, giving C14 with a reaction cross-section of 1.75b for thermal neutrons.

Argon becomes active but its activity is short-lived ($\sigma a = 0.62b$, half-life = 110 minutes). It is unlikely to be used unless the compatibility problems of the other coolants are insoluble and helium is unavailable.

Other more complex gases with excellent heat removal properties because of their high specific heat, such as methane, ethane, carbon tetrafluoride and benzene have been proposed, but it is probable that the above list contains the likely coolants for any high temperature system.

Coolants with an appreciable cross-section, such as nitrogen and argon, introduce a stability problem because the loss of coolant increases reactivity. In systems operating at 15 atm or more and a coolant voidage of 20 per cent., this increase could be of the order of 1 per cent.

Burn-up control

At 50b of stable poison per fission, 100 per cent. burn-up represents about 5 per cent. loss of reactivity due to poisons.

At a flux of 5×10^{13} the Xe and Sm poisoning causes a loss of reactivity of about 6 per cent. The problem of supplying this control is difficult and cumbersome. Burnable poisons may be used to avoid some of the awkward mechanical controls.

Temperature effect

The $\frac{1}{v}$ effect gives a reduction of fission and absorption cross-sections in the ratio of 1 at thermal (.025eV) to $\frac{1}{\sqrt{3}}$ at 606°C (.075 eV). Hence, one major difficulty will be the problem of starting from cold, and another the shutting down for a time sufficient for the temperature to fall.

Stability

In general, it is expected that these reactors will have a negative temperature coefficient. Nevertheless, this is by no means certain and more detailed work will be necessary to determine this point. Many temperature effects may be measured on a zero energy assembly, but some effects, e.g. the effect of the sharp fall in the absorption cross-section of Xe and Sm, will require special treatment. (Figure 4.)

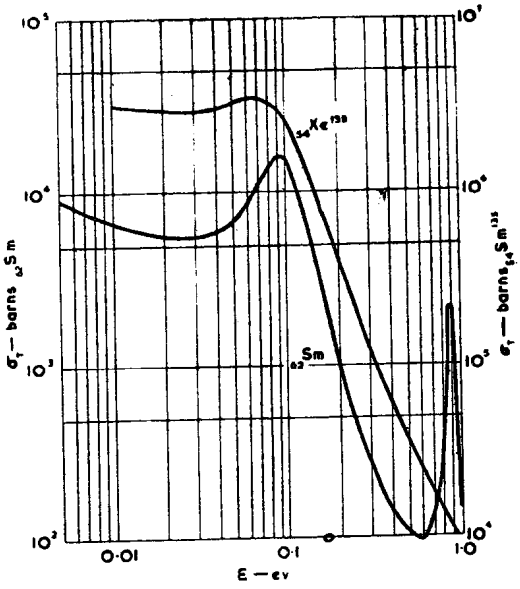

FIGURE 4: Change of σ_T of Xe and Sm with energy.

The resonance of plutonium at 0.3 eV can have an effect on systems burning plutonium.

The provision of sufficient control to deal with all these effects and to operate these controls at a temperature of the order of 350°C (cold part of the reactor) through a pressure shell, poses a challenging design problem.

Problems of an active circuit

The coolant circuit may become active because of induced activity in the coolant itself as in the case of nitrogen or argon or by the release of fission products from permeable fuel elements.

In the case of argon the problem is largely a matter of shielding and of leak control and ventilation. Because of the short half-life of the induced activity, the problems of maintenance and fuel changing, although difficult and expensive in "cooling" time, are within the bounds of practicability. The effect on stability of loss of coolant has already been mentioned. Nitrogen presents a more difficult problem because of the long-lived (half-life 5×10^3 years) and toxic carbon 14 formed. The destination of this product in the circuit is uncertain, and adequate leak control and ventilation will be necessary.

A more serious problem in systems using permeable fuel elements is the release of fission products. At high temperatures many of these will certainly escape into the coolant because some 40 per cent. of fission products pass through a gaseous phase. Sufficient work has not yet been done to determine the ultimate

destination of these fission products, and this in itself represents a long and difficult research task. As an example, consider only one fission product, xenon 137, which comprises 5.7 per cent. of the fission product yield. Much of this will escape from the fuel element and decay (3.9 minute half-life) to caesium 137 (half-life 33 years). There is a strong probability that this will deposit on heater-exchangers, blowers and ducting. In decaying to barium 137, a 0.66 MeV gamma radiation is emitted. Preliminary calculations indicate that heat exchangers, compressors, etc., will be equivalent to a source of the order of 10^3 curies for this one fission product.

It is clear that the maintenance of equipment under these conditions will present formidable problems to the engineer. It is relevant to note that the estimates of maintenance of an active circuit of a proposed homogeneous aqueous reactor have proved unexpectedly high (~ 4.1 mill/kWh) in a reactor specifically designed for such maintenance. Further, the additional capital cost due to the need for an active circuit may be serious.

It should not be assumed that these costs are strictly applicable to a gas-cooled system, and a great deal of detailed design will be necessary before reliable cost figures will be available. Nevertheless, it should be noted that systems confidently expected to produce power at 6-7 mills/kWh in 1955, are now being costed at 19.4 mills/kWh. (Anon 1957.)

Schemes have been proposed for the continuous removal of fission products released into the system, but the information available does not indicate that an advanced stage has been reached in this research. At this stage it is only possible to say that it is highly desirable to reduce fission product activity in the coolant to a minimum, either by a reduction of moderator porosity or by scavenging techniques.

Site safety should be mentioned in this respect. It is unlikely that safety committees will be happy with pressure vessels containing megacuries of activity in an unfixed state.

POSSIBLE REACTORS AND THEIR APPLICATION

For the few large central power stations near the major cities of Australia, the normal development of the U.K. and U.S. systems burning near-natural uranium will have an application for many years. Nevertheless, Australian requirements are mostly for units in the 5-50 MWE range—a range for which the type of reactor being discussed is most suitable.

Assuming feasibility problems are solved, economic factors will decide the choice of cycle from:

- primary gas coolant—secondary steam,
- primary gas coolant—secondary gas turbine, or
- single circuit gas coolant—gas turbine system.

In all cases it is desirable to have high integrity and high burn-up fuel elements as a unit of this size would not warrant a large scientific maintenance staff.

Two examples of possible reactors are:—

(1) A system consisting of cylindrical fuel elements made from a mixture of U Be_{13}, Th Be_{13} and canned in beryllium. The fuel zone may be either a very enriched annulus, or a homogeneous mixture of the fissile and fertile components with beryllium. The secondary cycle is steam, which may operate with air condensers if cooling water is expensive. W. R. Wooton (private communication), of Babcock-Wilcox, has proposed a 10 MWE unit to match such a reactor.

This example is very conservative, giving only about 1 MW heat per kilogram of fissile material. Much higher outputs are possible using higher gas pressures, smaller fuel elements and a higher coolant voidage fraction.

System—U : Th : Be 1:40:4000.
Reflector—graphite or beryllia.
Cylinder—163cm. diam. × 140cm. high.
Coolant—helium or carbon dioxide.
Heat output—~ 30 MW
Coolant voidage—17.5 per cent.
Mass beryllium (core)—4 tonnes.
Mass uranium—~ 26 kg.
Fuel element diameter—4.5cm.
Coolant pressure—15atm.
Coolant inlet temperature—250°C.
Coolant outlet temperature—500°C.
Beryllium surface temperature—600°C.
Beryllium centre temperature—630°C.
Thermal stress at surface—½ ultimate stress at 600°C.

Pumping power in primary circuit (say 1/3 in core, 2/3 in circuit)—~ 10 per cent. electrical output.

Burn-up— < 100 per cent. probably determined by endurance of fuel element.

(2) A reactor capable of supplying a gas turbine must deliver gas at at least 650°C. A possible system is a beryllia moderated and reflected reactor of 180cm. core diam., 15 per cent. voids and a heat output of 50-60 MW in the form of hot helium gas at 15 atm. pressure. For a gas temperature of 650°C a beryllia surface temperature of 850-900°C will be necessary if the fuel components and channel diameters are not to be inconveniently small. An assembly of plates with plates of the order of 1-2 cm. thickness may be preferable to a rod system in this case.

Larger outputs and high ratings are possible with systems of a higher channel voidage and a larger core cross-section.

Processing and economy

Only a limited mention will be made here of processing requirements. For processing purposes the mixing of fuel and moderator is a disadvantage, and an effort will be made to produce a fuel element in which the fuel and moderator are easily separable by mechanical means before processing. This applies particularly in the case of beryllium and beryllia moderated systems, where the value of the moderator makes recovery essential.

Finally, the economy of processing and fuel changing depends strongly on the in-pile life of the fuel element. The development of a high integrity fuel element of high burn-up—perhaps several times the initial investment of fuel—is the most important factor in the future of this type of reactor.

CONCLUSION

This paper does no more than outline some of the possibilities and problems of this type of reactor. Extensive irradiation tests of possible fuel element materials will be necessary before any may be selected with confidence for a reactor experiment. Corrosion and stability problems will have to be eliminated or solved, and fuel removal and processing will need to be examined in detail before any reliable cost assessment may be made.

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