Liquid Metal Fuel Reactors

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The advantages and disadvantages of a liquid metal fuel reactor system are outlined in relation to other possible power reactors. Liquid metals which are possible fuel carriers and coolants are discussed, and in particular a comparison is drawn between a solution-type fuel using bismuth and a dispersion-type using sodium. The reasons for the choice of a sodium dispersion fuel for the A.A.E.C. research project are presented and the problems posed by such a reactor system are summarised.

INTRODUCTION

Present trends of development in nuclear power reactors are towards higher operating temperatures to improve thermal efficiency, and towards more irradiation-resistant fuels to achieve increased reactor reliability and greater fuel utilisation.

"High temperature" operation requires definition. Considering existing large-scale power reactors, it may be said that the upper temperature limits are about 300°C for pressurised water units and 450°C for gas-cooled systems. Temperatures of 500°C and up can be regarded as high in nuclear reactors.

Operation at temperatures higher than 500°C should be possible with two types of coolant—gases and liquid metals. The Australian Atomic Energy Commission's research program has as its basis the study of such high temperature systems applied to thermal reactors. Advanced types of gas-cooled reactors are discussed by Dalton (1958); this paper considers the preferred alternatives among the possible liquid metal systems.

LIQUID METAL COOLANTS

The necessity for liquid coolants to operate at high temperatures has arisen in many instances before the advent of nuclear power. Classical examples are the General Electric Company's mercury boiler for increased efficiency of power generation, dating from 1922 (Hackett, 1942), and the use of liquid sodium to cool aircraft exhaust valves since 1928 (Heron, 1928).

Ideally, a liquid coolant for a thermal reactor should have low capture cross-section for thermal neutrons, a low melting point, high boiling point, and a low vapour pressure in the operating temperature range. It must be compatible with constructional and container materials which it contacts in the reactor core and external circuit, so that negligible corrosion or mass transfer of these materials will occur. It is desirable for the coolant to have good radiation stability and a low level of induced radioactivity after neutron irradiation to avoid the necessity for heavy shielding of circuit components external to the reactor core, and to minimise health hazards should leaks develop. Axiomatically, the coolant must have good heat transfer characteristics, i.e., high specific heat and thermal conductivity, coupled with low viscosity.

A number of liquids are available for high temperature operation, and include organic compounds, fused inorganic salts and liquid metals. Organic liquids such as "Dowtherm" (diphenyl-diphenyl oxide eutectic) have acceptable heat transfer properties, but only a small useful temperature range and indifferent radiation stability. Fused salts introduce problems of high neutron capture irradiation instability, and corrosion of container material.

The heat transfer coefficients of liquid metals are high, thus allowing high heat ratings per unit of reactor core volume or, alternatively, per unit of fissile material invested. Liquid metal systems need not be pressurised, giving considerable advantages in engineering design. Liquid metals are stable under heat and irradiation to the extent that, being elements or mixtures thereof, they do not decompose, polymerise or otherwise suffer in a reactor environment.

All liquid metals present some problems of compatibility and containment when considered for reactor service, and this factor, plus the effect of the coolant on the neutron economy of the reactor, constitute two of the main criteria in deciding the metal to be used.

Unfortunately, there is no one liquid metal which combines all the desirable attributes for a reactor coolant. Consequently, some compromise must be reached in making a selection, and other aspects of the reactor design so affect this selection that a number of research projects on nuclear reactors utilising different liquid metal coolants are in progress in the Western world (Williams, 1954; Siegel, 1955; Hoffman, 1956; Abraham 1957).

LIQUID METAL-COOLED AND LIQUID METAL-FUELLED SYSTEMS

Liquid metal coolants offer the opportunity of operating reactors with conventional type fuel elements at high temperatures and under conditions of high fuel rating. The additional step of mixing the fuel intimately with the coolant increases the possibilities of high ratings, and also removes the problem of the irradiation behaviour of fuel elements.

The desire to develop reactors having high reliability and good fuel utilisation has focused attention on irradiation damage in fuels since

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the successful operation of the earliest reactors. Under reactor irradiation, unalloyed uranium may distort by surface wrinkling, growth and swelling, causing failure of the fuel element of which it is a constituent (Ball, 1955). Heat treatment and alloying have been developed to minimise such damage, and present trends in ceramic fuels indicate that even greater resistance to irradiation damage is forthcoming. However, all solid fuel reactors are liable to the disadvantages of fuel element failure, and any system which overcomes these problems has potential advantages in reliability and high fuel burn-up.

One method of forming a "high integrity" fuel is to disperse fissile material in a matrix which is known to be stable under irradiation. This allows high heat output per unit of fuel, because the matrix may be used to extend the heat transfer surface of each fuel particle. The fuel for the Australian reactor HIFAR is of this type, being an enriched uranium compound dispersed in aluminium. Power reactors of high fuel rating may be built, using such fuels and suitable coolants, e.g., the U.S. Navy submarine "Sea Wolf" uses such a fuel in stainless steel, cooled by sodium (AECL CRR-590). However, so much parasitically neutron-absorbing material has been added to this type of fuel that a reactor using it is unlikely to be economical for civil purposes. An alternative is to disperse fuel in part of the reactor moderator. This achieves a high fuel rating by dispersing the heat source, but the irradiation stability of such dispersions in moderators suitable for high temperature operation (graphite, beryllium, beryllium oxide) has yet to be studied. The Australian high temperature gas-cooled reactor project involves studies of this type of fuel (Dalton, 1958).

In general, high temperature operation aggravates the problems of fuel element damage, and to utilise the advantages of liquid metal cooling it would be highly desirable to eliminate them altogether. A possible method is to use fuel intimately mixed with the coolant so that mechanical irradiation damage is either absent, as in a fuel solution, or of no consequence, as in a fuel slurry.

A variation of the latter approach is to replace a conventional fuel element by a "can" filled with liquid metal fuel solution or slurry. The first alternative, a canned liquid metal solution, is not attractive because of the very dilute nature of the available solution-type fuels. The "canned" slurry fuel element has been discussed widely, but has not been investigated seriously for thermal reactors because of unfavourable neutrol economy. However, should circulating fuel systems prove technologically not feasible, the concept of a canned slurry fuel, liquid metal-cooled, should receive further attention. It may be particularly suitable for a fast reactor.

Circulating combined fuel-coolant systems of several forms have been proposed and in some cases investigated. The aqueous homogeneous reactor is such a system, in which the fuel-coolant is also the moderator (AECD.3646, 1955). This reactor suffers from two disadvantages—the danger of a highly-pressurised and extremely radioactive circuit, and the corrosive nature of the liquid. Solutions of uranium salts in fused salt mixtures have been proposed also, but neutron capture in the salts and the compatibility of the liquid with constructional materials pose severe problems.

Circulating fuel systems have potential advantages in elimination of the charge-discharge problems of solid fuel reactors, and in the possibility of continuous de-poisoning and processing of the fuel. Such advantages would be lost in liquid metal systems employing canned fuel, with the exception that venting of the cans might allow continuous removal of xenon from the core.

The choice of liquid metal for a circulating fuel system is much more limited than for a liquid metal-cooled reactor, because an extra criterion—solubility of the fuel—must be imposed.

POSSIBLE LIQUID FUELS

Four types of liquid metal fuels may be postulated. These are:—

(i) Low melting point alloys rich in uranium (or plutonium);

(ii) solutions of uranium or plutonium in acceptable coolants;

(iii) slurries or dispersions of fissile material (metal or compounds) in equilibrium with solutions, i.e., added in quantities exceeding the saturation solubility; and

(iv) slurries or dispersions of fissile material (metal or compounds) with negligible solubility in the coolant.

No low melting point alloys with high uranium or plutonium contents and reasonably low thermal neutron capture cross-section are known. Consequently, the other attractive liquid metal fuels are those based on low melting point metals with good heat transfer and nuclear properties. Liquid uranium and plutonium alloys may find application in the future as fast reactor fuels, where the range of possible alloying elements and container materials can be extended considerably because of the decrease in their neutron capture cross-sections with rising neutron energy.

Metals which are possible thermal reactor coolants, and which may be considered for suitability as fuel carriers, are listed in Table I. Water and some unsuitable liquid metals are included for comparison purposes; the undesirable properties which cause their rejection are underlined.

Only three metals, lithium, lead and bismuth, have appreciable solubilities for uranium, and of these bismuth is outstanding in both this property and low neutron capture. It may be possible to increase the solubility of uranium in lithium and in lead by additions of further alloying elements, and work is proceeding on these lines in Great Britain. The

TABLE I*

Metal	Melting point °C	Boiling point °C	Density gm/cc at 500°C	Thermal conductivity cal sec^{-1} cm^{-1} °C^{-1} at 500°C	Viscosity Centipoise at 500°C	Heat capacity cal gm^{-1} °C^{-1} at 500°C	Thermal neutron capture Cross-section (barns)	Solubility for Uranium	Remarks
Bismuth	271	1477	9.75	0.037	1.84	0.0365	0.015	1000 ppm (300°C) 3.7% (650°C)‡	Attacks ferrous alloys
Gallium	29.9	1983	5.8	0.08 (30°C)	0.81	0.082 (to 200°C)	2.2	Very low	Expensive and attacks all known containers
Lead	327.4	1737	10.39	0.037	1.85	0.037	0.2 (lead 208, 0.00045)	Less than 1000 ppm (600°C)†	Cheap, readily available, but attacks most containers
Lithium	179	1317	0.48	0.09 (230°C)	0.455 (280°C)	1.0	65 (lithium 7, 0.033)	Approx. 1500 ppm (500°C)	Relatively scarce and expensive Attacks most containers
Magnesium	651	1103	1.6	—	—	0.317 (650°C)	0.3	500 ppm (650°C)	
Mercury	-38.9	357	12.88 (300°C)	0.03 (200°C)	1.01 (200°C)	0.032 (300°C)	430	—	
Potassium	63.7	760	0.72	0.090	0.17	0.183	2.5	Negligible	
Rubidium	39.0	688	1.475 (at M. pt.)	0.07 (at M. Pt.)	0.32 (220°C)	0.09 (at M. Pt.)	0.56	—	Scarce and expensive.
Sodium	97.8	883	0.83	0.160	0.239	0.302	0.45	Negligible§	Cheap, readily available. Relatively easy to contain
(Water)	0	100	1.0 (0°C)	0.001	0.259 (100°C)	1.0 (0°C)	0.6	—	

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* U.S.A.E.C. Liquid Metals Handbook, 2nd Edition

† Teitel, R. J. 1952

‡ Teitel, R. J. Gurinsky, D. H., and Bryner, S. J. 1954

§ Mogard, H. 1955

solubility of plutonium will not be considered further, because supplies of this element are not likely to be available in Australia in the near future. Also, plutonium fuelled systems have lower possibilities in conversion factor, which is likely to affect their economy adversely.

The thermal neutron capture cross-section of lithium is so unfavourable that this metal would have to exhibit outstanding advantages in other properties to make it attractive for thermal reactors. Similar reasoning applies to lead, its cross-section being much lower than that of lithium, but still high compared with bismuth. In both cases these elements occur in nature as mixtures of isotopes with widely differing nuclear properties, and an interesting possibility is the use of the low capture cross-section isotopes lithium 7 and lead 208 as reactor coolants. Present isotope separation processes are not likely to produce lead 208 economically, but the greater ease of isotope separation with light elements may mean that lithium 7 is a feasible coolant in the future.

Both lead and lithium dissolve many other metals, and consequently pose compatibility problems with container materials, but these difficulties appear not insuperable (Hoffman, 1955, 1956). However, bismuth also exhibits slight solubility for many other metals, and it should be emphasised that the magnitude of the corrosion problems in the use of this metal was not apparent from a study of its properties, but emerged as a result of a considerable research program (Weeks, 1955).

Liquid fuels of higher uranium content may be made by adding amounts of fissile material exceeding the saturation solubility in bismuth and lead. In both cases uranium forms an intermetallic compound with the coolant (UBi_2, UPb_3) and the result is a slurry of the relevant compound in equilibrium with liquid metal solution. The solubility of uranium in the liquid phase increases rapidly with rise of temperature in both systems, and this causes difficulty when the slurry flows in a circuit of varying temperatures, as would occur in a reactor and heat exchanger combination. Small particles dissolve preferentially in the hot region because of their greater surface to volume ratio, and precipitation in the cold region tends to occur on the remaining coarse particles. The result is particle growth to such an extent that deposition and eventual blocking ("plugging") of the circuit occur. Any system containing particles in equilibrium with a solution is prone to this effect, which is a case of "temperature gradient mass transfer." For example, the system thorium-bismuth has been investigated for possible use as a neutron absorbing blanket for thermal reactors, but the thorium content required is such that a slurry of $ThBi_2$ in bismuth is necessary. The alloy systems thorium-bismuth and uranium-bismuth are very similar, and particle growth of the $ThBi_2$ is proving a serious problem in such a blanket design (Barton, 1957).

The final alternative for a liquid metal fuel is a slurry in which the dispersed fuel has negligible solubility in the liquid, so that temperature gradient mass transfer and consequent particle growth is also negligible. Such slurries could be based on any of the preferred liquid metal coolants, and could contain insoluble uranium compounds such as UO_2, UC or UBe_{13}, or uranium metal where this is insoluble. Again bismuth appears most attractive because of its favourable cross-section, but most of the problems associated with this metal are with container materials, and if these can be solved, it appears more logical to use bismuth as a solution type fuel rather than in a slurry. It remains, then, to consider whether a slurry formed with any other liquid metal has sufficent advantages to compete with a bismuth solution.

Sodium and potassium have negligible solubility for uranium, and hence could be used as slurry vehicles for the metal or its compounds. Potassium has poor nuclear properties, and is more difficult to contain than sodium, and its only advantage appears to be a lower melting point. Alloys of potassium and sodium (NaK) have been used (Trocki, 1955) as reactor coolants, the potassium being added merely to lower the melting point of the coolant.

The thermal neutron capture cross-section of sodium is high compared with that of bismuth, but reference to Table I indicates that the heat transfer properties of sodium are much better than those of bismuth. This means that less sodium coolant is required for a given heat output, i.e., the poor nuclear properties of sodium are partly offset by its good thermal properties. In an insoluble slurry type fuel, the fissile material concentration is not dependent on a metallurgical property of the coolant. A liquid metal fuel could be made, based on sodium and containing uranium, in which the neutron losses to the coolant would be no greater than in a bismuth solution fuel, and in which a similar fuel rating could be achieved.

COMPARISON BETWEEN BISMUTH AND SODIUM AS FUEL CARRIERS

Metallurgical aspects

A considerable amount of metallurgical research has been devoted to the problem of finding suitable container materials for a bismuth-uranium fuel solution. Workers at Brookhaven National Laboratory (Weeks, 1955) have shown that the most promising and readily available constructional materials for bismuth circuits are steels containing 2.5 — 5 per cent. chromium and some molybdenum, but low in nickel and carbon. However, the extent of corrosion and mass transfer in these materials has led to investigation of corrosion-inhibiting additives in the bismuth. Additions of magnesium as a deoxidant and zirconium as an inhibitor have been found beneficial, and the mechanism of inhibition has been proved to be the formation of a film of zirconium nitride on the surface of the steel, the nitrogen being an inherent impurity in all steels. Horsley, working at Harwell, has shown that deliberate addition of further nitrogen to the steel prolongs the inhibition of corrosion (Horsley, 1957). However, the effects of high flux irradiation on the inhibition mechanism, and on the long-term corrosion resistance of a reactor circuit, are still unknown. Research at Ames Laboratory (Fisher, 1956) has shown that bismuth can be circulated for long periods at high temperatures (1000°C) in loops of pure tantalum, but this metal is expensive, difficult to fabricate, and has an undesirably high thermal neutron capture cross-section (22 barns). Generally, it appears that the problem of circulating bismuth for long periods under reactor conditions in materials which have desirable engineering, nuclear and economic characteristics, has not yet been solved.

On the other hand, sodium and NaK have been circulated in loops and in reactor circuits for long periods at high temperatures with little difficulty. Several reactors have been built and are operating (Siegel, 1955, AECL CRR-590) using sodium as coolant and stainless steel of the 18/8/1 type as constructional material. It is recognised that oxygen as an impurity has a profound effect on the corrosion of many metals by sodium (Liquid Metals Handbook, 1955), but adequate measures for control of oxygen content in sodium as a coolant have been developed. Thus, on the basis of compatibility with containers, sodium has distinct advantages over bismuth.

Engineering aspects

An important factor in a liquid metal fuel reactor design is the total investment of fissile material in the reactor circuit. Because fuel must pass through heat exchangers, pumps, processing vessels, and other components external to the reactor core, the total amount of fuel in a system based on bismuth may be three to five times that in a reactor core (Dalton, 1957). As the fuel rating of the reactor in megawatts per kilogram of fissile material must be considered on the basis of total fuel investment, the calculated rating of the core must be divided by the ratio of total hold-up to core investment.

In the case of sodium, a fuel slurry having neutron losses corresponding to those in a bismuth solution must have a higher uranium content. In the reactor core this is satisfactory for heat removal because of the high specific heat of sodium, but in the heat exchangers this same property makes necessary a greater heat transfer surface per unit of fuel than is required for the bismuth solution. As a result, the minimum possible volume of heat exchanger for a given power output contains more fuel for a sodium slurry than for a bismuth solution. Advances in heat exchanger design may reduce the actual value of this extra fuel hold-up, but preliminary calculations (Berglin, 1957; Dalton, 1957) indicate that the ratio of total investment to core investment for a sodium slurry system may be between 10 and 15. Thus, the overall fuel rating in a sodium system is probably inferior to that in a bismuth system.

A circulating liquid metal fuel solution would be homogeneous liquid initially, although fission product release would be expected to produce some solids (e.g., halides) and gases (e.g., xenon, krypton). Continuous fuel processing outside the reactor core would keep these to low equilibrium values. On the other hand, a sodium slurry presents new problems in the circulation of the fuel itself, because there is little experience of maintaining dispersion in light liquid systems containing fine dense particles. Possible fuel additions are discussed later, but the density range of these is between 4.4 gm/c.c. and 19 gm/c.c., whereas the density of sodium at 500°C is 0.8 gm/c.c. The problems of maintaining dispersion in such liquid-solid systems are being studied by Cairns (1957), using tungsten and other powders in water as analogues for uranium and other fuel additives in sodium. Initial results indicate that uniform dispersion can be maintained in circulating systems at reasonably low velocities of about 6.5 ft./sec. for tungsten particles smaller than 10 microns diameter in water.

This analogue work is to continue, because it is necessary to investigate the behaviour of such a dispersion under all conditions likely to be encountered in a reactor circuit. The behaviour of a reactor, fuelled by uranium particles dispersed in sodium has been studied theoretically by Dalton and Thompson (1957), who concluded that short-term variations of fuel concentration by more than 2 per cent. in the circulating fuel/coolant would have serious consequences in the form of power and temperature transients. Thus, the conditions necessary to ensure uniformity in the fuel feed must be established.

Work with aqueous suspensions may not give adequate information on the sodium based systems, because the wetting characteristics are different, and ionic phenomena do not occur in sodium. Thus, similar studies of particle behaviour in flowing sodium must be undertaken. Small loops to do this, using electromagnetic pumps, are being built as part of the metallurgical research program at Lucas Heights, and a larger loop has already been constructed by the chemical engineering section.

Sodium has several minor advantages over bismuth in the engineering of reactor circuits. The lower melting point of sodium results in less difficult "heat tracing" of circuits to ensure that all regions are molten at start-up. Heat tracing for bismuth circuits must be carefully designed because the metal expands on solidification and may fracture pipework if allowed to freeze indiscriminately. Another factor in favour of sodium is its high electrical conductivity, enabling it to be pumped by electromagnetic means far more readily than can bismuth. Finally, the sheer weight of large volumes of bismuth constitutes a disadvantage in necessitating heavy pipework and supports, whereas sodium is lighter than water.

Chemical aspects

There is no clear distinction between chemistry and metallurgy when considering phenomena in liquid metals. However, certain problems of both the liquid fuels may be termed chemical.

The bismuth fuel solution under study in the U.S.A. (Williams, 1954; Weeks, 1955) contains 600 ppm of uranium. Under reactor conditions, and operating on either U233 or U235, other non-fissile uranium isotopes would be produced and would build up in the fuel, thus limiting the fissile content if the saturation solubility is not to be exceeded. This problem does not occur in a slurry fuel, because the allowable concentration does not depend on solubility.

Continuous chemical processing of bismuth-based fuel is being studied, using molten salt contacting techniques, which are promising (Dwyer, 1955). No simple system of continuous processing for sodium slurries has been devised to date, and it may be necessary to separate fissile material from coolant before processing. A relatively simple means of separation exists in the hydroclone (Berglin, 1957), and work on this principle is proceeding in the A.A.E.C. research laboratories.

A further point favouring bismuth fuels from the processing viewpoint is the possibility of a simple operation to replenish the fuel with uranium produced in a neutron-capturing breeder blanket surrounding the reactor core. If a thorium-bismuth slurry (containing $ThBi_2$) is used as blanket, U233 is produced in the $ThBi_2$ particles. Raising the temperature of the alloy to the point at which all thorium and uranium is in solution, and cooling to reconstitute the slurry, results in significant concentration of uranium in the liquid phase, which could be further concentrated and added to the core (Dwyer, 1955). The particle growth problems of a thorium-bismuth alloy have been described above, but there is considerable incentive to solve them.

Finally, the effects of neutron irradiation on the coolant itself lead to chemical problems in the case of bismuth. The product of neutron capture in bismuth is polonium, a highly active alpha emitter, which would constitute a severe health hazard in bismuth circuits. Neutron irradiation of sodium produces the isotope Na24, a short-lived gamma emitter, which decays to magnesium 24; this is no problem and may even be beneficial is some reactor circuits as magnesium is a powerful deoxidant. In the sodium slurry liquid metal fuel other elements present are likely to have higher affinity for oxygen.

Summary—the Australian L.M.F.R. project

Of the two most promising liquid metal fuels, the bismuth solution has been chosen by workers in both the U.S.A. and England on the grounds of low thermal neutron capture cross-section, ease of handling in comparison with a slurry, and the promise of simple high temperature chemical processing. Unfortunately, the problem of corrosion of structural materials by liquid bismuth has proved much more difficult than was anticipated and, indeed, may prove insurmountable.

Liquid sodium has some disadvantages when compared with bismuth, in that a fuel slurry is more difficult to circulate than a solution, and the higher neutron absoption leads to larger fuel investments in the reactor system as a whole. However, the fact that sodium has extremely low solubility for most container materials gives promise that the compatibility problems of a sodium-based system will be less difficult to solve. The sodium slurry system for thermal reactors is not being studied elsewhere, and for these reasons it was selected as a suitable project for the Research Establishment of the Australian Atomic Energy Commission.

There are many possible combinations of moderator and fuel additions for use with a sodium coolant, and it is not possible to study all of these with the limited staff and facilities available. Thus, some elimination was necessary, although it is realised that in the process some promising combinations may have been deleted. The choice of materials for detailed study is outlined briefly in the following sections.

THE CHOICE OF MODERATOR

For high temperature service, possible reactor moderators are graphite, beryllium and beryllium oxide.

The smaller critical sizes of reactors using beryllium and its oxide as moderators give these materials advantages over graphite in permitting lower fuel inventory and power output. Further, there is a reasonable possibility of making the fuel/coolant compatible with beryllium or its oxide, whereas graphite and sodium are incompatible. In the only sodium-cooled graphite moderated reactor built to date, it was necessary to can the graphite in zirconium (Siegel, 1955), because the available graphites were all penetrated and disintegrated by liquid sodium.

Beryllium exhibits a nuclear property which enhances its value as a moderator, but which may have serious mechanical consequences. Two nuclear reactions occur in beryllium under fast neutron irradiation; these are the n, α and n, 2n reactions, both of which produce helium as one end product. Calculations based on the available data shows that a "neutron enhancement" of several per cent. is possible by the n, 2n reaction in a beryllium moderated system ("Beryllium," ASM, 1956), and this increases the possibility of breeding on the Th232-U233 cycle in a uranium-sodium-beryllium reactor. On the other hand, the effect of the helium atoms on the beryllium is not known. Calculations by the author, and later by Hickman (1958), have indicated that at high temperatures the beryllium may swell as a result of helium bubble formation. Experimental irradiations of beryllium at high temperatures in HIFAR are being undertaken, and will provide both fundamental and technological data on this problem.

Beryllium oxide has several disadvantages compared with metallic beryllium; the neutron enhancement by n, 2n reaction is likely to be less, the material is more difficult to fabricate into shapes of low porosity and the thermal shock resistance is inferior.

The mechanical effects of helium production on beryllium oxide are difficult to predict. On one hand, the relatively open lattice structure of the oxide may accommodate helium atoms more readily than the almost close-packed metallic lattice; and, on the other, the oxide must be considered as a brittle material, so that sufficient internal strain set up by the presence of many foreign atoms would cause cracking and possible crumbling.

Beryllium metal has been chosen as a moderator material for initial investigations on the feasibility of a sodium-based system, because it has the best combination of nuclear properties, it is relatively easy to fabricate, has good resistance to thermal shock, and it is negligibly soluble in sodium. The disadvantages of the metal, compared with its oxide, are mainly the temperature limitation imposed by the low creep strength of beryllium at high temperatures, and corrosion problems caused by oxygen in the sodium. Beryllium oxide corrosion in sodium is being studied in France (Grison, 1956), and the results of this work are of continuing interest to the Australian group.

POSSIBLE FUEL ADDITIONS TO SODIUM

The fissile material may be uranium metal or any compound capable of being prepared as a fine powder which is wetted by sodium, and which does not contain elements of unacceptably high thermal neutron capture cross-section. Because fission of uranium atoms in a compound will ultimately release the other combined atoms, these must not be of a substance which will cause corrosion of the moderator or circuit materials. Three uranium compounds have acceptable nuclear properties, are readily prepared and readily wetted by liquid sodium. These are UO_2 UC and UBe_{13}.

The particle size of the powder is determined by two factors:

(i) To enable fission products to escape from the particles, with the attendant possibility of continuous de-poisoning of the reactor, particles less than 10 microns in diameter are desirable (Hickman, 1958).

(ii) To mantain uniform dispersion, it is desirable to have particles with minimum settling velocity, i.e., as small as possible.

Consideration of settling rates of metallic uranium in sodium led to the choice of particles of less than 10 microns for initial studies (Kelly, 1958).

Uranium metal may be prepared in the form of particles smaller than 10 microns by repeated hydriding and dehydriding of massive uranium (Kelly, 1958). Uranium powder is wetted by liquid sodium at approximately 400°C, and does not de-wet on cooling (Taylor, 1955). Uranium is not corroded in liquid sodium provided that the oxygen content of the liquid metal is kept sufficiently low (Mogard, 1955). A disadvantage of elemental uranium as a fuel is its density, 18.3 gm/c.c., compared with that of sodium at 500°C (0.83).

Uranium oxide UO_2 has the advantage of being readily prepared as a fine powder. It is wetted by sodium at about 380°C (Taylor, 1955); a dispersion of UO_2 in sodium is being studied by Abraham (1957) at Argonne National Laboratory as a possible fast reactor fuel. A possible disadvantage of UO_2 is the liberation of oxygen to the system by fission of uranium. The most serious compatibility problems of sodium are those involving oxygen as an impurity, and this is particularly true of circuits in which metals with a high affinity for oxygen are present, e.g., beryllium. The influence of oxygen released from UO_2 on the corrosion of beryllium in sodium is to be investigated by irradiation tests in HIFAR.

Uranium carbide may have a similar disadvantage; if carbon is released by fission, it will undergo mass transfer in sodium to any metal forming a stable carbide. This is an example of "concentration gradient mass transfer," in which even minute solubility of a substance in a liquid metal will cause appreciable transfer from a "source" to a "sink" in a circuit, even in the absence of a temperature gradient. No work is being done on uranium carbide because it does not possess any outstanding advantages over uranium metal or UBe_{13}.

The most interesting fuel addition is UBe_{13}, the inter-metallic compound formed between uranium and beryllium. It has the advantage of low density (4.4 gm/c.c.) compared with uranium metal, and this reduces settling out problems of the dispersion. The compound is relatively easy to prepare by heating finely-divided mixed powders of uranium and beryllium to 1,100°C. Further advantages of this fuel addition emerge when considering metallurgical aspects of the sodium-uranium-beryllium system.

METALLURGICAL PROBLEMS OF THE SODIUM-BASED FUEL/COOLANT

Mention has been made of the influence of oxygen impurity on corrosion and mass transfer of metals in flowing sodium, and the effect is greatest for metals of high affinity for oxygen. In systems using sodium as a coolant only, several methods have been developed to maintain a low oxygen concentration in the liquid metal.

One such system is "cold trapping," consisting of passing the sodium stream, or a portion of it, through a region at temperatures just above the melting point. Because the solubility of oxygen in sodium falls with decreasing temperature, precipitation of sodium oxide occurs in the cold trap, and filtration or settling in a region of low velocity can retain this oxide. By this means oxygen content can be minimised to less than 100 parts per million. This system is not applicable to circuits containing metals with oxides more stable than sodium oxide.

A second system is to "hot trap" the sodium (Siegel, 1955) by passing it through a region containing material with a higher affinity for oxygen than any other material in the circuit. Results to date indicate that it is necessary to run hot traps higher in temperature than the material to be protected, particularly for reactive materials such as beryllium or zirconium. Attempts have been made to protect beryllium in sodium circuits by hot-trapping with thorium (Bett, 1958). Theoretically this should be effective if both metals are at the same temperature, because thorium oxide is more stable than beryllium oxide. However, the results indicate that considerable oxidation of beryllium occurs despite the hot trap, indicating that an important factor is the rate of oxidation. The kinetics of oxidation of reactive metals in sodium will be studied by chemists and metallurgists as part of the Australian project.

In the case of a liquid metal fuel containing fine particles, neither of these systems can be adopted, because both depend for removal of oxygen on a low-velocity reaction vessel, which would allow settling of the fuel. A third alternative is the use of a soluble "getter" in the sodium, of which calcium is the most promising. Preliminary work indicates that corrosion of beryllium metal in sodium is reduced considerably by the presence of calcium (Bett, 1958). However, calcium and nickel interact, and the effect of calcium additions on nickel-bearing stainless steels is being studied. Initial results indicate that the quantity of calcium required for "gettering" has negligible effects on 18/8/1 stainless steel (Bett, 1958).

The use of UBe_{13} as fuel addition may provide another method of oxygen control. Fission of uranium atoms in the compound particles will release beryllium atoms, and it is hoped that the higher chemical activity of this beryllium compared with the moderator material will result in continuous "gettering" of oxygen without appreciable corrosion of the moderator. This hypothesis will be evaluated by irradiation experiments in HIFAR.

In addition to the problems of oxygen impurity, fission products released within the sodium may cause corrosion and mass transfer of moderator and circuit materials. The system is so complex that experimental work, including irradiation tests, will provide the only real guide. It may be seen in general that a considerable amount of metallurgical research is required before the fuel-coolant, moderator, and constructional materials for a reactor can be defined.

SUMMARY

The reasons for the choice of a sodium-based liquid metal fuel system as a high temperature reactor study in Australia may be summarised as follows:—

(i) A liquid metal fuel system has potential advantages in high fuel rating, high burn-up, and high temperature operation, the latter leading to high thermal efficiency.

(ii) Only two liquid metal fuels are attractive; these are a solution of uranium in liquid bismuth, and a dispersion of uranium or a compound in liquid sodium. Bismuth solution systems are being studied in detail in at least two other countries, whereas sodium fuels are not receiving much attention.

(iii) Sodium presents less problems of compatibility with container materials than does bismuth. It is in this field that the bismuth system has encountered its most critical technological barriers.

Not all of the many possible combinations of fuel, moderator, and container materials have been discussed, but the aim has been to indicate the main reasoning behind the current research programme, and the nature of the problems involved.

Sufficient design calculations have been done to indicate that the sodium-based system is a promising one if the technological problems can be solved. Design of a liquid metal fuel reactor experiment must await satisfactory completion of compatibility and fluid flow studies, which involve considerable metallurgical, chemical, and engineering effort.

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