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TECHNICAL DIVISION

ENGINEERING RESEARCH SECTION

A DISCUSSION OF LIQUID METALS AS PILE COOLANTS

A. S. KITZES

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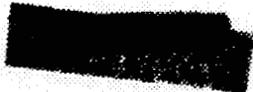
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TECHNICAL DIVISION
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A DISCUSSION OF LIQUID METALS AS PILE COOLANTS

A. S. Kitzes

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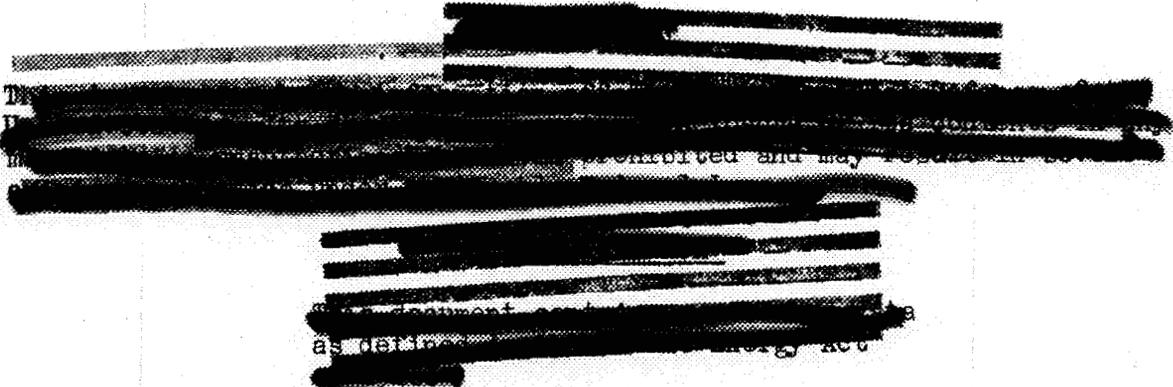


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1.0 STATEMENT OF PROBLEM

Nuclear reactors are being considered for use on ships, planes, submarines, and for other uses where the space allotted for the reactor and its auxiliary cooling equipment is limited. The heat transfer surface is less in enriched piles than in thermal piles which necessitates the use of highly efficient cooling systems and heat transfer agents. A great deal of emphasis, consequently, has been placed on developing more efficient methods and coolants for removing the heat from the reactors. This report is concerned only with the coolants.

Water, both light and heavy, and air are the only fluids which have been used to date to remove the heat from thermal piles. Mercury is being used to a limited extent in the fast reactor at Los Alamos. However, many other possible coolants have been suggested, and of these the liquid metals and helium look the most promising. Many methods have been proposed to evaluate the relative worth of these fluids as heat transfer agents since it is impossible, both from an economic standpoint and the time involved, to determine experimentally the relative merits of these materials.

The purpose of this survey therefore is two-fold:

- 1) To evaluate the various liquid metals as pile coolants.
- 2) To indicate which of these metals should be investigated more thoroughly in the laboratory.

This report is limited to a discussion of the liquid metal coolants since it was felt that the gaseous coolants and water have been covered adequately in the project literature.

2.0 INTRODUCTION

The rate at which energy, as heat can be removed from a nuclear reactor is one of the limiting factors which influences the design of a pile. The heat is removed from the reactor in either one of two ways:

- 1) Recycling of the coolant. The coolant is circulated through the pile and the heat which has been picked up is released in a secondary heat exchanger. The coolant is then recycled.
- 2) Discarding of the coolant. The coolant is circulated through the pile and then discarded at a higher temperature.

A number of different liquids and gases have been proposed as the cooling fluids but very few of these have actually been used. NaK alloys have been suggested because of their low melting points and high fluidity. Bismuth has also been considered because of its extremely low thermal neutron capture cross-section.

Numerous methods for the evaluation of fluids have appeared in the literature. Burton and Davis (4) calculated the "isocaloric cross-section" for a number of materials. The isocaloric cross-section is the cross-section of the molecule for thermal neutrons divided by the molecular weight and the heat capacity, $\frac{\sigma}{Mc}$, and is a criterion for evaluation developed by Wheeler. Bentley, Brown, and Schelegel (2) have evaluated 14 fluids at different temperatures, using as the basis for their comparison the standard heat transfer coefficient equation for a material flowing in a cylindrical pipe. Bismuth, lead, mercury, sodium, sulphur, potassium, NaK alloys, water, helium, and air are some of the substances which were considered for their comparisons. The authors also list the physical properties of the proposed coolants up to that time. Parsons and Gaffney (11) have developed equations relating the film coefficient of heat transfer and

2.0 INTRODUCTION (Con't.)

the energy consumed in circulating the fluid. Water, mercury, air and sodium were some of the fluids evaluated. Of these, mercury and sodium are superior to water as coolants, and water is superior to air.

Some of the liquid metals which are being considered as pile coolants have been evaluated by Lyon (10) who used for his comparisons certain relationships based upon pumping power, absorption cross-section for thermal neutrons, and permissible operating temperature range. Li^7 , proved to be the most favorable coolant according to Lyon's system of evaluation, Bi, Na, NaK (50%) showed up as next best.

Bloom (3) has listed the physical and nuclear properties of twenty-one elements which may be used as coolants. The two criteria which were used for the evaluations were melting point and thermal neutron properties. A number of alloys and salts which can be used as coolants is also included in this report.

Feld (6) has estimated the induced gamma radioactivity for a number of possible liquid metal coolants. Bismuth and lead were found to be the most favorable from the induced activity point of view. Tin, magnesium and sodium were the least favorable.

Everhart (5) has critically reviewed the literature on low melting alloys. Since the author was interested in only those low melting alloys which do not react violently with oxygen or water at high temperatures, the report covers primarily the lead, bismuth, tin, and cadmium combinations. Physical property data, however, are included for the elements and many other alloys. A complete bibliography is also attached to the report.

2.0 INTRODUCTION (Con't.)

Various liquid metals were evaluated by Atkin (1) in his discussion of power conversion equipment for the Project "Genie". The evaluation was based upon heat transfer coefficient, pumping power, chemical reactivity, minimum volume of coolant, shielding and melting point. Sodium appears to be the best choice of coolant with NaK running a close second according to Atkin's evaluation. Gallium, HTS (a mixture of potassium and sodium nitrates), mercury, and Pb-Bi follow in the order listed.

3.0 CRITERIA FOR EVALUATION OF COOLANTS

The choice of a fluid for use as a heat transfer agent in a reactor is dependent upon many factors of which there are three types:

- 1) physical properties of the fluid under consideration
- 2) nuclear properties - both in a thermal and fast pile
- 3) engineering factors such as cost, availability, corrosion effects, health and fire hazards, etc.

These factors can be divided into two separate groups:

- 1) Critical or Screening Factors - factors which determine whether or not a substance can be used as a heat transfer agent.
- 2) Relative Factors - factors which can be used to compare various materials once it has been established that these materials can be used as heat transfer fluids.

These two groups can be sub-divided as follows:

- 1) Critical or Screening Factors
 - a) corrosion
 - b) absorption cross-section (less important in fast piles where moderating may be a critical factor)
 - c) availability

3.0 CRITERIA FOR EVALUATION OF COOLANTS (Con't.)

- d) melting point
- e) stability to continued irradiation

2) Relative Factors

a) Nuclear Properties

- 1) cross-section
- 2) induced activity
- 3) moderating ability

b) Cost

c) Handling Difficulties

- 1) chemical reactivity
- 2) corrosion-container materials
- 3) melting and boiling point
- 4) vapor pressure
- 5) pumps
- 6) Hydraulic properties (viscosity, density, atomic weight)
- 7) health and fire hazards
- 8) electrical conductivity

d) Heat Transfer Properties

- 1) heat capacity
- 2) thermal conductivity
- 3) relative heat transfer coefficient pump power/input

3.1 Critical Factors

These factors are the so called "obvious factors" which can be used to screen out from further consideration those materials which are obviously of no value as heat transfer agents.

3.1.1 Corrosion

'Corrosion effects' is one factor which has been neglected in the various published evaluation schemes. A liquid metal may have excellent thermal properties for heat transfer, acceptable nuclear properties, and it may be available at a reasonable cost - the features of a desirable coolant for a reactor. It has no practical value as a coolant, however, if it reacts with all the metals which might be used to contain it. Gallium, for example, has been considered as an ideal liquid metal coolant. It has average heat transfer properties and acceptable nuclear properties; it is liquid at room temperatures and can be produced at a reasonable cost if there is a sufficient demand for it. A great deal of time, money, and effort have already been expended in trying to develop gallium as a heat transfer agent. If proper consideration, however, had been given to the chemistry of gallium before the development program was initiated, the survey would have revealed that gallium would, probably, not be an acceptable heat transfer fluid because of its high affinity for other metals. It reacts chemically with all the materials which might be used to contain it.

Secondly, if a slight attack of the tube walls must be tolerated, then it becomes necessary to increase the thickness of the tube walls to prevent the frequent replacement of cooling tube sections. The size of the

3.1.1 Corrosion (Con't.)

active section of the pile, consequently, must be increased to maintain the criticality of the pile because of the absorption of neutrons by the added metal.

Thirdly, consideration must be given to the nuclear properties of the material which is to be used to contain the coolant. It is impractical to consider a coolant because of its low absorption cross-section and good heat transfer properties if the material which is necessary to contain it has a high neutron absorption cross-section.

In view of the above facts, any evaluation scheme for liquid metals which does not include corrosion effects must be considered incomplete. Unfortunately, there is very little information in the literature on the corrosive action of molten metals on constructional material. It was, therefore, necessary to omit corrosion from the evaluation scheme which will be presented in this report, even though it was realized that 'corrosion effects' is one of the more important criteria for screening coolants.

3.1.2 Nuclear Properties

Very little has to be said about this criterion. Naturally the liquid metals with low thermal neutron capture cross-sections are more favorable for thermal piles, while those with high neutron scattering cross-sections are more desirable for fast piles. The shielding requirement for the secondary heat exchanger, in both cases, is dependent upon the induced radioactivity of the coolant. The induced activity of the

3.1.2 Nuclear Properties (Con't.)

corrosion products must also be considered if there is any appreciable attack of the container walls by the liquid metal. The question of providing adequate shielding at a reasonable cost for the secondary heat exchanger may become a limiting factor, especially, in those cases where additional space is at a premium.

3.1.3 Availability

It is impractical to consider using a liquid metal as a coolant because of its good heat transfer properties and low power requirements for circulating it when it is available only in limited quantities and the cost/lb. to produce it is high. Consideration should be given first to the availability of the metal and to whether or not it is economically feasible to market it in large amounts.

3.1.4 Melting Point and Stability to Continued Irradiation

The usefulness of these criteria in screening out possible pile coolants is apparent. Elements with high melting points such as iron, nickel, manganese, etc. - elements which would be normally used as containers have obviously no value as coolants. It is difficult to draw the line of demarcation between those elements which have acceptable melting points and those which do not. In this report the upper limit for the melting point was arbitrarily set at 650° C.

The coolant is recirculated continually through the pile to remove the reactor heat in a closed cycle cooling system. It is, therefore, essential that the proposed coolant be stable to repeated irradiations, otherwise, they have no value as a pile coolant.

3.2 Relative Factors

These factors can be considered as the 'comparative factors' or criteria by which the various fluids which have passed the screening test can be compared. There is no single criterion by the use of which a fluid may be selected as the best heat transfer agent in all piles.

The difference between the melting and boiling points determines the operating range of the fluid. It is, of course, desirable that the fluid be liquid at room temperatures so that when the pile is shut down for any length of time, the fluid will not solidify in the cooling tubes. It is reasonable, however, to assume that a cooling system can be designed to use metals or alloys which are not liquid at room temperature. Too much emphasis, therefore, need not be placed on the melting point criterion, except for those metals which melt at extremely high temperatures (above 650° C) - in which case, corrosion and container problems would be the controlling factors in making a selection.

Heat transfer properties and handling difficulties must be included in any evaluation scheme. The design and geometry of a pile for a given power level dictate the amount of space that is available for the cooling tubes. It is, therefore, essential that the coolant remove the heat from the pile efficiently and with a minimum expenditure of the pile energy for circulating the cooling fluid. In other words, a high value of the heat transfer film coefficient for a low energy input in circulating the fluid is the desired condition. Nuclear properties, cost, and corrosion have been discussed under Critical Factors, consequently no further discussion of these factors is necessary.

4.0 EVALUATION SCHEMES FOR LIQUID METAL COOLANTS

This section is devoted to the development of the schemes which are used in this report to evaluate the various liquid metals. Many schemes have been proposed to compare the relative merits of fluids as heat transfer agents. A brief discussion of these schemes was presented in the literature review.

4.1 Heat Transfer Properties

The assumptions which were made in the derivation of the following relationships are:

- 1) A pile of given power, Q
- 2) The dimensions and geometry of the pile are fixed so that the length of the cooling tubes is the same for all coolants.
- 3) The diameter of the cooling tubes is the same for all coolants.
- 4) The pressure drop is proportional to u^2 .
- 5) Absorption cross-section of the channel walls for thermal neutrons, and the moderating effect of coolant and container metal are negligible.

4.1.1 Case I - Temperature rise of cooling fluid is limiting factor.

This situation is usually found in piles where there is considerable surface area available for cooling as compared to the cross sectional area of the cooling tubes.

4.1.1.1 Thermal Piles

$$Q = Wc \Delta T \quad (1)$$

since,

$$W = uS \rho \quad (2)$$

then,

$$Q = uS \rho c \Delta T \quad (3)$$

The macroscopic absorption cross section for thermal neutrons can be expressed by the equation:

$$X = N_0 \sigma = \frac{A}{M} \rho \sigma \quad (4)$$

where

A = Avogadro's Number

M = Atomic weight

or

$$X \propto \frac{\rho \sigma}{M}, \text{ if the constants in Equation (4) are } \quad (5)$$

omitted.

Since the total macroscopic absorption cross-section is proportional to the volume of coolant in the pile, the cross-sectional area, S, of the channels can be expressed in terms of σ by:

$$X' \propto Sl \frac{\rho \sigma}{M} \quad (6)$$

$$\text{or } S \propto \frac{M}{\rho \sigma}, \quad (7)$$

since X' and l are constant for a given pile.

4.1.1.1 Thermal Piles (Con't.)

The pumping power required to circulate a fluid is given by:

$P = \text{volume} \times \text{pressure drop (by definition)}$

$P \propto S \rho u^2 \tag{8}$

or

$P \propto S \rho u^3 \tag{9}$

Equation (3), (7) and (9) are combined so as to eliminate u and S with the result that:

$P_1 \propto \frac{Q^3 (\sigma)^2}{c^3 M^2 (\Delta T)^3} \propto \frac{Q^3}{B} \tag{10}$

where $B = \frac{c^3 M^2 (\Delta T)^3}{\sigma^2} \tag{11}$

and contains only terms representative of the fluid.

4.1.1.2 Fast Piles

The absorption cross-section need not be considered in fast pile calculations. The pumping power, consequently, required to circulate the fluid can be expressed by:

$P_2 \propto \frac{Q^3}{D^4} \left(\frac{1}{\rho^2 c^3 (\Delta T)^3} \right) \tag{10a}$

where $F = \rho^2 c^3 (\Delta T)^3 \tag{11a}$

4.1.2 Case II - Temperature drop from the tube wall to the coolant is the limiting factor. This situation will arise in enriched piles where it is essential that all the heat be transferred across relatively small heat transfer surfaces.

$$Q = ha \Delta T \tag{12}$$

$$a = \pi D L \tag{13}$$

$$S = \frac{\pi}{4} D^2 \tag{7}$$

or,

$$a = \frac{4LS}{D} \tag{14}$$

and since,

$$S \propto \frac{M}{\rho \sigma}$$

$$a \propto \frac{M}{D \rho \sigma} \tag{15}$$

The film coefficient for heat transfer can be expressed by Lyon's approximate equation:

$$\frac{hD}{k} = 7 + .025 (Pe)^{.8} \tag{16}$$

where $Pe = Re \times Pr = \frac{Du c \rho}{k}$

$$\text{or } h = 7 \frac{k}{D} + .025 \left(\frac{k}{D}\right)^{.2} (c \rho)^{.8} u^{.8} \tag{16)a}$$

The following procedure was used to compare the liquid metals:

- a) The film coefficient for heat transfer, h, was evaluated using Equation (16) for various values of D and u.

4.1.2 (Con't.)

- b) The pumping power P_3 , required to circulate the fluid at velocities which might be necessary in a pile was estimated by using Equation (9).
- c) The ratio P_3/h , was then used for the comparisons. The absorption cross-section was not included in these relationships.

In order to take into account the absorption cross-section of the liquid metals, the assumption can be made that for large Peclet numbers, the term, $(Pe)^{0.8}$ is equal to or very much greater than 7. In this case,

$$h \propto \left(\frac{k}{D}\right)^{0.2} (c \rho)^{0.8} u^{0.8} \tag{17}$$

Substituting u from Equation (17) for h in Equation (12) and eliminating u from Equation (9) gives

$$P_3 \propto \frac{Q^{3.75} D^4 \rho^{0.75} \sigma^{3.75}}{M^{2.75} c^3 k^{0.75} (\Delta T)^{3.75}} \propto \frac{Q^{3.75} D^4}{H} \tag{18}$$

since,

$$a \propto \frac{M}{D \rho \sigma} \tag{15}$$

$$\text{and, } H = \frac{M^{2.75} c^3 k^{0.75} (\Delta T)^{3.75}}{\rho^{0.75} \sigma^{3.75}} \tag{18)a}$$

contains only terms representative of the fluid.

4.2 Nuclear Properties

The following assumptions were made in establishing the relationships for estimating the induced activity of the coolant:

- 1) The radioactivity of the coolant material arises from neutron reactions involving the metal itself.
- 2) Pure metal with no impurities is used for the coolant.
- 3) Neutron reactions with the container tube walls are not considered.

The disadvantage factor Z, based on the number of disintegrations per second per cubic centimeter can be determined as follows:

$$N_1 = f(nv)(N_0)(1 - e^{-\lambda t}) \quad (19)$$

where

N_1 = number of disintegrations per second per cc

N_0 = total number of original atoms present per cc

nv = neutron flux (assumed constant)

f = fraction of coolant in the reactor at any one time (assumed constant)

If it is assumed that the reactor is in operation for a long period of time, Equation (19) reduces to

$$N_1 = f(nv)(N_0) \quad (20)$$

since,

$$\lim_{t \rightarrow \infty} 1 - e^{-\lambda t} = 1$$

A atoms weigh M grams and occupy a volume of $\frac{M}{\rho}$ cm³ or,

$$N_0 = \frac{A \rho}{M} \quad (21)$$

A = Avogadro's number

M = Atomic weight

4.2 Nuclear Properties (Con't.)

Equations (20) and (21) are combined so as to eliminate N_0 with the result that:

$$N_1 \propto Z \quad (22)$$

if Z is defined as:

$$Z = \frac{\rho \sigma}{M} \quad (23)$$

since f and the neutron flux were assumed constant Z may be considered therefore, as a disadvantage factor which is proportional to the number of disintegrations per second per cubic centimeter after equilibrium has been established in the pile.

5.0 DISCUSSION OF DATA

5.1 Physical Properties

The physical properties of the liquid metals are listed in Table I. The elements included in this table are those which may be used as a coolant by itself or which may be alloyed with the other elements in the table to form low melting compounds.

More than one value is reported for a physical property in a number of cases. It was impossible to evaluate the data since the original papers from which the data were abstracted are not available in the Oak Ridge National Laboratory libraries; or, in some cases, the source of the data was not listed. Data which were not available and which were needed in appraising the metals were either calculated from known relationships or approximated by using the data from similar metals. Thermal conductivities were estimated from electrical conductivity data by using the Wiedemann-Franz-Lorentz equation:

5.1 Physical Properties (Con't.)

$$k = A \sigma T + B \quad (24)$$

k = thermal conductivity

σ = electrical conductivity

T = absolute temperature, °K

A, B = constants

Little mention has to be made of the data in Table I except to point out the exceptionally high thermal conductivity and heat capacity of Lithium. The high melting point of Lithium may be considered as a disadvantage. The boiling point (280° C) of phosphorous is low which probably precludes its use as a pile coolant.

5.2 Heat Transfer Evaluation

A comparison of the various liquid metals as heat transfer agents is shown in Table II. The pumping power required to circulate the fluid under four different heat transfer conditions has been evaluated for each element. These four cases have been described in a previous section. NaK (47% Na) and Pb-Bi (43.5% Pb) have been included in the evaluation since they are now being tested for use in piles.

The evaluation was based on pumping power, cross-section for thermal neutrons, and operating range. The difference between the melting point and 650° C was arbitrarily chosen as the operating range. Consequently, elements with melting point above 600° C are automatically thrown out. Neutron moderation, corrosion, and handling difficulties have not been included in the evaluation.

TABLE I - PHYSICAL PROPERTIES OF POTENTIAL PILE COOLANTS

ELEMENT	M.P. °C	B.P. °C	SpG		VISCOSITY		C _p		K		VAPOR PRESSURE (atm)					HEAT OF FUSION cal/gm	ELECTRICAL RESISTIVITY		HEAT CONTENT ABOVE 25° C					
			t(°C)	t(°C)	Centi- poises	t(°C)	cal/gm °C	t(°C)	CGS	t(°C)	.0001	.001	.01	.1	1.0		microhms	t(°C)	cal/gm t(°C)	t(°C)	t(°C)	t(°C)	t(°C)	
Al	660	2056	2.4 2.36 2.29	660 800 1000			.25					1110	1263	1461	1713	2056	94.6	20.1	660	291	800	343	1000	
Sb	630.3	1440	6.55 6.45 6.38 6.29	M.P. 700 800 970	1.296 1.113 0.999 0.905	702 801 900 1002	.066	650				759°C	872	1013	1196	1440	38.3	120 117.6 116	860 700 630.5	82	800	94.4	1000	
Bi	271.3	1420	10.04 9.91 9.66 9.40 9.20	300 400 600 802 962	.998 1.28 1.66	600 451 304	.037		.0418 .037 .037 .037 .037	300 400 500 600 700		914	1008	1121	1254	1420	12.5	139.9 128.9 134.2 145.25 153.55	500 300 400 600 750	20.8	300	24.4	400	31.6
Cd	320.9	765	7.56 8.02 7.54 7.52	700 321 720 750	1.63 1.18	600 506	.077 .064	321- 700	.105	350			384	471	594	765	13.2	34	400					
Ce	600.5																5.5							
Cs	28	690	1.84	28			.058						10 ⁻² 153 mm	10 ⁻¹ 207 mm		708	3.8	36.6	28	8.3	100	14.3	200	
Ga	30	2071	6.0947 6.08 6.01 5.91 5.84 5.78 5.72 5.60 5.49 5.445	31 52.9 149 301 402 500 600 806 1010 1100	1.89 1.41 1.03 .88 .81 .77 .65 .59 .58	52.9 149 301 402 500 600 806 1010 1100	.082	119				1178	1329	1515	1751	2071	19.2	28.4 27.2	46 30	26.4	100	36.1	200	
Ge	958	1760															114.3							
In	156.4	1450															6.8	29	156.4					
Pb	327.4	1744	10.69 10.60 10.36 10.14	327 400 600 800	1.38 1.54 1.85 2.32	700 600 500 400	.037 .037 .039	500 400 327	.036 .038	M.P. 400		815	953	1135	1384	1744	6.3	98.3 107.2	400 600	18 31.2	400 800	21.3	500	
Li	186	1372					1.374		1.02			592	707	858	1064	1372	32.8	45	230					
Mg	650	1107	1.58 1.55 1.54 1.53 1.51	666 678 696 710 720			.28							725	886	1107	89							

TABLE I (Con't.)

ELEMENT	M.P. °C	B.P. °C	SpG t(°C)		VISCOSITY		Cp cal/gm t(°C)	K CGS t(°C)		VAPOR PRESSURE (atm)					HEAT OF FUSION cal/gm	ELECTRICAL RESISTIVITY		HEAT CONTENT ABOVE 25°C				
					centi- poises	t(°C)				.0001	.001	.01	.1	1.0		microhms	t(°C)	t(°C)	t(°C)	t(°C)		
Hg	-38.87	357	13.5	50	.90	350	.033		.02	0	77.9	120.8	176.1	251.3	357	2.7	95.8 114	20 200	2.5 9.1	100 300	5.8 200	
			13.35	100	.92	340																
			13.26	140	.95	300																
			13.11	200	1.05	200																
					1.24	100																
		1.69	0																			
P	17	280	1.69- 1.74	44	1.21	80	.2				1.7×10^{-3} 145	3.66×10^{-1} 235	5.45×10^{-1} 250		280	5.0	23					
K	63.7	770	.83	63	.55	64	.20	90-180			261	332	429	565	774	14.5	13 15.3	63 100				
Rb	39	705	1.475	39			.091	50 °C			223	288	377	497	705	6.1	19.6 23.2	40 50				
Se	220	685	3.97 3.95 3.89	225 245 277							282	347	430	540	685	6.6	76.6	860				
Na	97.7	892	.93	98	.73	98	.33	100	.205 .202 .191	100 150 210	349	429	534	679	892	27.5	9.7	100	49.6	100	82.2	200
S	119	444.6	1.81		11.46	119	.22		.000314	115						9.3			48 99.6	200 400	73 300	
					6.72	157.3			.000316	120												
					93100	187.2			.000369	200												
					2100	(max)			.000378	210												
						306																
Te	449.8 [±] .2	1387					.05					509	632	810	1387	7.3						
Tl	300 [±] 3	1457	11.51	300			.04	300- 500	.059	350	692	809	962	1166	1457	7.2	74	300				
			11.0	303.5- 351				.035	300- 500													
Sn	231.9	2270	6.98	232	1.2	500	.064	232	.08	240				1932	2270	14.5	61.2 49.4	750 300	30.8 49.3	300 600	369 400	
			6.93	300	1.3	425		-1100	.08	300												
			6.7	700	1.678	280			.078	400												
			6.5	1000	1.664	296			.076	500												
			6.40	1200	1.421	357																
					1.045	604																
					.905	750																
Zn	419.5	907	6.92 6.81 6.57	420 600 800	2.23	600	.121 .122 .124 .128	420 500 600 800	.14	420		477	579	717	907	24	37.1	500				

5.2 Heat Transfer Evaluation (Con't.)

The data in Table II indicate that Li^7 , and, if the absorption cross-section for thermal neutrons need not be considered, natural lithium is one of the more desirable coolants. Na, K, Mg, Al, and NaK also show up favorably. It must be realized however, that the evaluations are based on single values of the heat transfer properties which were assumed constant over the entire operating range. Also, the reliability of these data was not established. The results, however crude and imperfect do indicate that considerable attention should be given to lithium as a pile coolant.

5.3 Nuclear Properties

The neutron induced radioactivity of the coolant determines the amount of shielding which is necessary for the secondary heat exchanger. A summary of the estimates of the induced radioactivity is shown in Table III. In each case the procedure was to compute the number of disintegrations per second per cubic centimeter in the pure coolant during a constant time of operation of a pile at a constant power level. It was assumed that all of the disintegrations took place in the heat exchanger. Self-shielding and neutron moderation effects were not included in this evaluation. Corrosion, erosion and other reactions with the container walls were also omitted.

It is seen from the data in Table III that Bi, Li^7 , Pb, P, Tl, are ideal for thermal piles since there are no gamma radioactivities resulting from neutron capture. Although Bi appears on top of the list because of its low capture cross-section and low disadvantage factor Z , the use of Bi introduces a serious handling problem. Bi produces Po when it captures a

TABLE II - PUMPING POWER ADVANTAGE FACTORS

	M.W.	M.P. °C	Specific Gravity	Absorption Cross-Section for Thermal Neutrons	Heat Capacity	Thermal Conductivity	Temperature Drop Across Film or Temp. rise in Cooling Fluid	Temperature Drop in Coolant Controlling	Temperature Drop in Coolant Controlling Absorption Cross- Section not Important	Temperature Drop Across Film Controlling						H		
										P ₃ /h Values								
										1 ft/sec	5 ft/sec	10 ft/sec	25 ft/sec	10 ⁻⁶	10 ⁻⁵		10 ⁻⁴	10 ⁻⁴
			ρ	σ	c	k	ΔT 650°C M.P.	B	F									
Li ⁷	7	186	.53*	.033	1.374	1.02	450	1.2x10 ¹³	6.8x10 ⁷	3	4	3.7	4	2.8	3	.04	.25	2.3x10 ²⁰
Na	23	97.7	.93	.45	.32	.20	550	1x10 ¹⁰	5x10 ⁶	26	35	30	30	22	18	30	1.5	2.5x10 ¹³
K	39	63	.83	2.5	.18	.13	600	4.4x10 ⁸	8.9x10 ⁵	40	50	40	400	35	25	50	2.4	1.5x10 ¹¹
Mg	24	650	1.57	.06	.28	.20*	---	---	---	50	60	50	400	40	25	50	2	---
Al	27	660	2.4	.21	.25	.25	---	---	---	50	70	60	400	48	27	57	2.4	---
NaK	30.6	7	.88	1.13	.29	.066	650	5x10 ⁹	5x10 ⁶	70	80	80	500	55	25	70	2.3	5.7x10 ¹²
Rb	85	39	1.47	.56	.09	.085	610	4x10 ⁹	3.9x10 ⁵	100	130	110	1000	84	60	120	5.7	2.4x10 ¹³
Cs	133	28	1.84	26	.058	.044	625	1.3x10 ⁶	1.6x10 ⁵	200	300	260	2000	200	110	250	10	8.1x10 ⁷
Zn	65	420	6.5	.9	.121	.14	230	1.1x10 ⁸	8.4x10 ⁵	300	270	280	1700	200	88	240	7.2	5.9x10 ¹²
Sn	119	232	7.0	.55	.064	.08	420	9.8x10 ⁸	9.5x10 ⁵	400	300	450	1900	300	110	400	8.0	4.9x10 ¹¹
Cd	112	321	7.6	2900	.077	.105	330	24	1x10 ⁵	400	400	400	2800	270	120	350	11	.23
Ga	70	30	6.1	2.2	.082	.044	620	1.3x10 ⁸	4.9x10 ⁶	700	550	600	3000	400	150	450	11	1.7x10 ¹¹
Sb	122	631	6.55	4.5	.066	.042	---	---	---	800	660	800	3400	500	160	500	13	---
S	32	119	1.81	.4	.22	.000316	320**	2.2x10 ⁹	1.2x10 ⁶	4500	1000	1700	3600	800	170	700	14	1.4x10 ¹²
Tl	204	300	11.5	3	.04	.059	350	1.2x10 ⁷	3.6x10 ⁵	1000	900	1000	5000	700	260	800	20	1x10 ¹⁰
Bi	209	271	10	.015	.037	.037	380	5.5x10 ¹¹	2.8x10 ⁵	1400	1200	1300	6000	830	300	1000	25	6.6x10 ¹⁸
Pb	207	327	10.7	.2	.037	.039	320	1.9x10 ⁹	1.9x10 ⁵	1400	1400	1300	6000	1000	300	1000	24	3.7x10 ¹²
Pb-Bi	208	125	10.2	.18	.037	.026	530	1x10 ¹⁰	7.8x10 ⁵	2000	1500	1800	7000	1100	340	1200	27	2.6x10 ¹³
Hg	201	-39	13.5	430	.033	.025	700	2700	2.2x10 ⁶	2600	1700	2000	8000	1400	390	1400	30	2500

* - Estimated values

5.3 Nuclear Properties (Con't.)

neutron which has a long half life and which emits a high energy alpha particle. In addition, Bi cannot be held in an open vessel because of the recoil from the alpha particle emitted from Po. This latter fact makes Bi one of the most powerful physiological poisons known. Feld (6) has estimated the Po activity if Bi is used as a coolant in a thermal or fast pile. The maximum specific activity of Po after one day's operation, according to Feld, is .066 curies/lb for a thermal pile with an average thermal neutron flux of $2.4 \times 10^{14}/\text{cm}^2\text{-sec}$, and 0.56 curies/lbs for a fast pile with an average fast neutron flux of $8 \times 10^{15}/\text{cm}^2\text{-sec}$.

An alpha particle is also emitted when Li^7 captures a neutron. But these alpha particles do not present a serious handling problem because the half-life of the isotope that is formed when Li^7 captures a neutron is less than one second. The isotope, in all probability, will have therefore decayed in the pile before reaching the heat exchanger. A minimum amount of shielding, consequently, is required for the heat exchanger. Another factor should be considered but is not included in the evaluation scheme is the moderating ability of Li^7 in thermal piles. These two factors---short half-life isotope and good moderating ability in thermal piles make Li^7 more preferable than Bi as a pile coolant.

Tl, because of its high disadvantage factor, Z, should appear in the table below Zn and above Ga. It, nevertheless, occurs in fifth place because the isotope that is produced when Tl captures a neutron does not release any gamma rays.

TABLE III

NUCLEAR PROPERTIES

METAL	THERMAL NEUTRON CAPTURE CROSS-SECTION-BARNS σ_a	NEUTRON SCATTERING CROSS-SECTION-BARNS σ_s	NUCLEI PER CC ($\times 10^{-24}$) N	AVERAGE LOG ENERGY LOSS-E	SLOWING DOWN POWER $N \sigma_s$	MODERATING RATIO $(\sigma_s / \sigma_a) E$	Disadvantage Type of Induced Factor - Z Radiation	INDUCED ACTIVITY
Bi	.015	9.2	.0282	.0096	.002	4.2	.0007	Alpha, Beta
Li ⁷	.033	1.5*	.0463*	.2643*	.018*	12*	.0023	Alpha, Beta
Pb	.2	13	.0330	.0097	.003	.39	.010	Beta
P	.23	10	.0455	.0637	.012	.87	.012	Beta
Tl	3	9.7	.0349	.0098	.003	.029	.169	Beta
Mg	.06	3.6	.0431	.0807	.009	.73	.0039	Beta, Gamma
Rb	.56	11.0	.0108	.0234	.003	.47	.0096	Beta, Gamma
Na	.45	3.5	.0254	.0852	.009	.76	.0183	Beta, Gamma
Al	.21	1.5	.0603	.0730	.006	.47	.0185	Beta, Gamma
S	.4	1.5	.0376	.0616	.002	.15	.022	Beta, Gamma
Sn	.55	5.0	.0370	.0169	.003	.11	.0315	Beta, Gamma
K	2.5	1.5	.0134	.0507	.001	.03	.054	Beta, Gamma
Zn	.9	4.2	.0662	.0305	.007	.12	.090	Beta, Gamma
Ga	2.2	18	.0510	.0287	.023	.21	.191	Beta, Gamma
Sb	4.5	4.3	.0031	.0165	.002	.013	.242	Beta, Gamma
Cs	26	9.0	.0085	.0151	.003	.014	.359	Beta, Gamma
Hg	430	15.	.0407	.0100	.006	0	28.9	Beta, Gamma
Cd	2900	5.3	.0463	.0178	.005	0	196	Beta, Gamma
Ce	6	23	.0297	.0143	.010	.055	---	Beta, Gamma
Ge	2.8	6	.0453	.0275	.007	.059	---	Beta, Gamma
In	194	4	.0382	.0175	.001	0	---	Beta, Gamma
Se	15	13	.0343	.0253	.011	.022	---	Beta, Gamma
Te	4	5.0	.0295	.0157	.022	.02	---	Beta, Gamma
NaK	1.13	----	----	----	----	----	.032	Beta, Gamma
Pb-Bi	.18	----	----	----	----	----	.0088	Alpha, Beta

* Natural Lithium

5.3 Nuclear Properties (Con't.)

The other liquid metals listed in Table III are less favorable than Li, Pb, P, Tl, Bi from the radioactivity point of view. Feld (6) has estimated the induced activity of many possible pile coolants and reference should be made to this paper for a more complete evaluation of liquid metals based upon radioactivity.

5.4 Corrosion

The importance of considering corrosion effects in choosing a liquid metal has already been discussed. Unfortunately, pertinent published corrosion data for all the elements are lacking which, of necessity, precluded the inclusion of this important criterion from consideration.

Kelman (9) studied the effect of NaK alloys on various materials at elevated temperatures. He found that uranium, thorium, beryllium and columbium showed good resistance to attack by pure NaK alloy. The presence of oxygen, however, increased the corrosion of these metals. Pure iron and low carbon steels also show good resistance to NaK attack. The presence of chromium or chromium alloys cause these materials to corrode much faster. Nickel and high nickel alloys are apparently not attacked by NaK. Copper and copper alloys are not suitable for use with NaK.

Wilkinson (13) presented a paper at the Symposium on Basic Properties of Liquid Metals held at Argonne National Laboratory, April 5 - 6, 1949 in which he reviewed the reactions of metals in various liquid metals at elevated temperatures. A great deal of reference material was also presented. A summary of Wilkinson's paper is shown in Table IVa. Tidball (12) also presented a paper at the Symposium on Basic Properties of Liquid Metals in

TABLE IVa

A SUMMARY OF THE DATA PRESENTED BY WILKINSON
IN HIS REVIEW OF CORROSION BY LIQUID METALS

Specimen	Liquid Metal	Temp. of Test	Test Period (Static Test)	Reaction and Solubility
Al(2S)	Bi-Pb eutectic	300 - 500° C	1 hour	.003% - .037% Al.
Al(17S-T)	Bi-Pb	500° C	1 hour	.035% Al
Al	Pb	327 - 658° C	-	Nil
Al	Na & K	657° C	-	Nil
Al	Ga	300 - 500° C	-	15 - 45% Al
Be	Bi	1000° C	5 hours	< .01% Be
Be	Bi	700° C	24 hours	Nil
Be	Bi-Pb	500° C	2 1/2	< .01% Be
Be	Na-K	600° C	6 days	-.0015 mg/cm ² /hr
Be	Sn Pb	700° C	24 hours	Nil
Be	Ga	500° C	2 weeks	No change in Wt. or dimensions
Mild Steel	Bi-Pb	1000° C	1 hour	Surface attacked evenly.
Fe	Bi	400° C	-	.0002% Fe
Fe	Bi	1600° C	-	.0004% Fe
Fe	Pb	327 - 1528° C	-	.0002% - .003% Fe
Fe	Ga	600° C	2 days	Formed solid Ga-Fe alloys
Stainless 18-8	Bi-Pb eutectic	500 - 1000° C	1 hour	Stainless attacked
Cr	Bi	268 - 1553° C	-	Nil
Ni	Bi	469° C	-	4% Ni
Ni	Bi	655° C	-	6.5% Ni
Ni & Mn	Bi-Pb eutectic	Molten Temp.	-	Severe attack on Ni and Mn
Cr	Pb	274° C	-	Slight solubility
Cr	Pb	1470° C	-	15% Cr (?)
Ni	Pb	327° C	-	.19% Ni
Ni	Pb	1340° C	-	7% Ni
Ni	Na-K eutectic	600° C	6 days	-.0001 mg/cm ² /hr.
Ni	Ga	600° C	2 weeks	Ni readily alloys with Ga

TABLE IVb

A COMPARISON OF CORROSION DATA

<u>Sample</u>	<u>Bath</u>	<u>Temp.</u> <u>°C</u>	<u>Source</u>	<u>Corrosion Rate</u> <u>mg/cm² mo</u>	
Mild Steel	Na	500	G.E.	-0.72	
Mild Steel	NaK	539	M.S.A.	-0.87	-1.01
Mild Steel	Pi-Pb	500	A.N.L.	-6,950.	
18-8	Bi-Pb	500	A.N.L.	-14,800.	
18-8	NaK	539	M.S.A.	- 1.19	‡0.46
Type 347	Na	500	G.E.	‡ 0.072	‡0.72
Type 347	NaK	539	M.S.A.	‡ 0.50	‡1.17
Be	Na	500	G.E.	- 0.72	‡2.16
Be	NaK	600	A.N.L.	- 1.08	
Be	Bi-Pb	500	A.N.L.	0.01% Be	

G.E. - General Electric Company - Knolls Atomic Power Laboratory

M.S.A. - Mine Safety and Appliances Company

A.N.L. - Argonne National Laboratory

5.4 Corrosion (Con't.)

which he compared his corrosion data with those of Wilkinson, Argonne National Laboratory, and Vandenberg, Knolls Atomic Power Laboratory. These data are also shown in Table IVb.

5.5 Availability and Cost of Metals

Grant (8) has tabulated and discussed the availability and cost of many of the metals which are scarce or available in limited quantities. A part of the data shown in Table V was abstracted from Grant's report. The availability of the scarce elements are shown in Table V. Where not indicated, the element is available in sufficient quantity to be used as a coolant.

It is apparent from the data in Table V that one major draw-back for the use of Li^7 is that there is no Li^7 available. Natural lithium contains 92.5% Li^7 and until Li^6 can be separated economically from natural lithium, Li^7 cannot be considered as a pile coolant.

5.6 Health Hazards

These factors are not too important and are only included in Table V to indicate the precautions that must be taken in handling a specific metal. Toxicity and reactivity data were supplied by Mr. W. H. Baumann of the Medical Division and Mr. D. Gary of the Safety Department at Oak Ridge National Laboratory.

TABLE V

AVAILABILITY AND HEALTH HAZARDS

	Availability		Possible Cost (\$/lb) if produced in quantities	Reactivity with air or water	Health Hazards	Toxicity
	lbs.	Cost \$/lb.				
Al	---	.15	.15			-
Sb	---	.29	.29	†		-
Bi	---	1.80	1.80	†		††
Cd	---	.99	.79	-		†
Ce	---	---	---	-		††
Cs	10	1820	---	-		-
Ga	100	1130	---	-		-
Ge	100	---	2.00	-		-
In	20	27	---	-		-
Pb	---	.12	.12	-		-
Li7	0	---	---	-		††
Mg	---	.21	.21	†		†
Hg	---	1.16	1.16	†		†
P	---	---	---	-		††
k	---	2	.80	†		††
Rb	10	2720	---	†		†
Se	---	---	---	†		†
Na	---	.19	.15	†		††
S	---	---	---	†		†
Te	---	---	---	-		-
Tl	---	12.5	---	-		††
Tn	---	.70	.70	-		††
Zn	---	.15	.15	†		-
						††

† - shows positive reaction
 †† - can cause death
 - - no reaction

6.0 BINARY ALLOYS FOR COOLANTS

Bloom (3) has listed a number of binary metallic alloys which can be used as pile coolants because of their acceptable melting points and capture cross-sections. Of these the following alloys seem to warrant further considerations:

<u>Alloy</u>	<u>Composition</u>	<u>Melting Point °C</u>	<u>Average Neutron Capture Cross-Section Barns</u>
Al-Mg	68% Mg	436°	.11
Bi-Sn	42% Sn	139°	.34
Na-Tl	41% Tl	64°	.65
Mg-Zn	53.5% Zn	340°	.42
Na-Rb	92% Rb	-4.5	.49
K-Rb	91.5% Rb	32.8	1.05
K-Na	47% Na	7	1.13
Bi-Pb	44.5% Pb	124	.18

The bismuth-lead alloy is desirable because of its low melting point, low capture cross-section and negligible gamma radioactivity due to irradiation. The Al-Mg alloy is most desirable from the heat transfer point of view. However, its fairly high melting point and its induced gamma radioactivity are disadvantages which cannot be neglected. Pertinent information for most of the other alloys is lacking. Pb-Bi and NaK alloys have already been discussed.

7.0 RECOMMENDATIONS

7.1 Consideration should be given to Li^7 as a pile coolant because of its excellent heat transfer properties and low absorption cross-section for thermal neutrons. Time and effort, therefore should be expended in developing a process for the separation of Li^7 from natural lithium. A reduction in the Li^6 content of natural lithium from 7.5% to 0.1% would lower the absorption cross-section for thermal neutrons of the Li^7 and Li^6 mixture from 65 barns to approximately 0.9 barns. This amounts to the removal of approximately 99% of the Li^6 .

Corrosion of constructional materials by liquid lithium should also be studied. There is very little information available in the literature on this subject. However it is reasonable to assume that corrosion of metals by liquid lithium should be similar to that by the other liquid alkali metals.

7.2 All corrosion data in liquid metal systems should be issued as separate formal reports by the various governmental nuclear agencies. It is very difficult, at the present time, to locate corrosion data which are contained in monthly, quarterly, or in weekly progress reports. This information should be made readily available to all interested personnel. It is understood that the Committee on Liquid Metals will soon issue a Handbook on Liquid Metals which will contain, as a chapter in the Handbook, a complete summary of the corrosion data collected on the various Atomic Energy Commission and Navy projects.

7.3 Corrosion studies using Na, NaK, Pb-Bi, and Ga should be continued. Corrosion properties of Li should also be included in these studies.

7.4 A study should be made to determine the feasibility of producing the rare elements such as rubidium and thallium in large quantities at a reasonable cost.

7.5 The heat transfer characteristics and the physical properties of the more desirable pile coolants should be ascertained over the complete possible operating range for the coolant. The other desirable coolants besides Li⁷ are according to the evaluation scheme presented in this report:

- Na - good heat transfer characteristics
- NaK - good heat transfer characteristics and low melting point
- Pb - Bi - insignificant gamma radiation--no shielding of heat exchanger is necessary. However, this alloy cannot be used where frequent decontamination or maintenance of the cooling system is required

7.6 No further consideration should be given to gallium as a heat transfer agent until the problem of finding a suitable container for gallium is solved. The work to find a container for gallium should, however, be continued. If and when a suitable container is found, the heat transfer characteristics of gallium should be studied.

8.0 NOMENCLATURE

- a - Total surface area in channels for heat transfer
- A - Avogadro's Number
- B - Advantage factor when temperature rise in coolant controls
- c - Heat capacity
- D - Diameter of channel
- f - Fraction of coolant in pile at any given time (assumed constant)
- F - Advantage factor when temperature rise in coolant controls and thermal neutron absorption cross-section need not be considered.
- h - Heat transfer coefficient
- H - Advantage factor when temperature drop across film controls
- k - Thermal conductivity of coolant
- l - Length of channels (assumed constant)
- M - Atomic weight of metal
- N_0 - Number of atoms per cubic centimeter of coolant
- N_v - Neutron flux
- P - Pumping power
- Pr - Prandtl number, $c_p \mu / k$
- Q - Power output of pile (assumed constant)
- Re - Reynolds number, $\frac{D u \rho}{\mu}$
- S - Total cross-sectional area of channels for coolant through the pile
- t - Time of irradiation of coolants in pile (assumed constant)
- (ΔT) - Permissible temperature rise of coolant in pile or permissible temperature drop from channel wall to bulk of coolant through the film. Temperature change in either case was assumed to be the difference between the melting point of element and 650° C.

8.0 NOMENCLATURE (Con't.)

- T - Half-life of major isotope of coolant that is formed during irradiation
- u - Velocity
- W - Weight of cooling flowing per unit time
- X - Total macroscopic absorption cross-section for thermal neutrons
- Z - Advantage factor based on the number of disintegrations per second per cubic centimeters of coolant.
- μ - Viscosity
- ρ - Density
- σ - Absorption cross-section for thermal neutrons
- λ - Decay constant
- \mathcal{L} - Proportionality factor

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