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Selection of Liquid Metals in Reactor Technology

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Selection of Liquid Metals in Reactor Technology*

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Zusammenfassung

Die Auswahl flüssiger Metalle in der Reaktortechnik

Der Zweck dieser Arbeit ist es, den konsequenten Weg zu der Erkenntnis aufzuzeigen, daß sich von allen Flüssigmetallen Natrium (und seine Legierungen mit Kalium) am besten als metallisches Kühlmittel für Kernreaktoren eignet. Es werden zuerst die Gesichtspunkte für die Auswahl von Flüssigmetallen zusammengestellt und diskutiert. Diese »Auswahlregeln« finden Anwendung auf eine große Anzahl von Metallen und Legierungen mit hinreichend niedrigem Schmelzpunkt. Die Betrachtung der kernphysikalischen Eigenschaften, der Wärme- und Strömungstechnik, des chemischen Verhaltens, der metallurgischen Einflüsse auf das Behältermaterial, nicht zuletzt auch rein wirtschaftliche Fragen wie Preis und Verfügbarkeit führen zu einer sehr engen Auswahl einiger weniger Flüssigmetalle, die für die wirklichkeitsnahe Lösung von Kühlproblemen in der Reaktortechnik in Frage kommen. Nach einer Zusammenstellung aller wichtigen Eigenschaften wird schließlich der Versuch unternommen, die relative »Brauchbarkeit« abzuschätzen und eine entsprechende Rangordnung aufzustellen.

Abstract

The aim of this presentation is to show the consequent way leading to the knowledge that–from all the liquid metals–it is sodium (and its alloys) that best fits the postulates of modern reactor technology. Beginning with the composition and discussion of the points of view for the selection of liquid metals these "selection rules" are applied to a large number of metals and alloys having a sufficiently low melting point. Because of the nuclear properties, the heat transport phenomena, the chemical and corrosion behaviour and the economic situation finally only a few metallic liquids remain which are applicable to real heat transfer problems. After a summary of all the important properties the attempt is made to establish a "range of applicability" for these remaining metals.

EURATOM KEYWORDS:

LIQUID METALS
LIQUID METAL COOLANT
SODIUM
SODIUM ALLOYS
POTASSIUM
POTASSIUM ALLOYS
BISMUTH ALLOYS
LEAD
LEAD ALLOYS
LITHIUM
LITHIUM 7
MERCURY
MERCURY 204
ALLOYS
EUTECTICS
USES
REACTORS
MELTING POINTS
BOILING
HEAT TRANSFER
FLUID FLOW
CORROSION
CHEMICAL REACTIONS
THERMAL CONDUCTIVITY
DENSITY
VISCOSITY
TOXICITY
ACTIVATION
CROSS SECTIONS
ABSORPTION
THERMAL NEUTRONS
FAST NEUTRONS
FERMI AGE
ECONOMICS
TABLES
BIBLIOGRAPHY

1. Introduction

During the past ten or fifteen years a large amount of theoretical considerations and practical experiments were carried out in order to introduce the so-called "liquid metals" into reactor technology. There is a lot of pure metals and a large number of alloys liquid already at relatively low temperatures. But very soon it was recognized that only a few of them could fit the special requirements in the nuclear field. We all know that in this competition it is mainly sodium and its alloys which are preferred.

This paper does not attempt to produce additional suggestions and points of view broadening and influencing the way of development. This would hardly be possible, at least rather useless, for all the possible fundamental considerations and very many selection experiments have already been made, especially in the U.S.A., but also by European experts. The aim of this work is rather to realize the logical evolution–that is to say since the beginning of reactor technology–in the selection of suitable metallic liquids in a consequent manner.

2. Points of view for the selection

The points of view for the critical examination of a metallic cooling medium in relation to reactor technology can be divided into the following items:

2.1 State of liquid and related physical properties

The melting point governs whether the circuit needs an auxiliary heating and–for instance–what installation has to be provided.

The boiling point limits the temperature of the circuit if pressurized systems must be avoided. It also relates to safety considerations.

The linear thermal expansion of a metal solid at normal temperature sometimes is important for the design of the circuit.

The volume change at fusion and the volume expansion of the fluid must be noticed.

The heat of fusion influences the power of the auxiliary heaters.

The vapour pressure at running temperature of the cycle relates to the possible losses of coolant.

2.2 Nuclear properties

The neutron absorption cross section is due to the neutron balance. In a thermal reactor σ_{th} is interesting, in a fast reactor the cross section $\sigma_{100\,keV}$ for 100 keV-neutrons is typical.

The average logarithmic decrement ξ per interaction together with the scattering cross section σ_s describes the moderation of neutrons by elastic scattering (which is desirable in thermal reactors, but not in fast reactors).

The inelastic scattering cross section σ_{inel} is due to moderation of fast neutrons by inelastic interaction.

The (n, γ)-activation, the half life and the γ-energy at decay determine the shielding of the circuit.

2.3 Heat transfer and fluid flow

The judgement of heat transfer is based on the following data:

thermal conductivity λ
specific heat c_p
density ϱ
specific heat per unit volume $\varrho \cdot c_p$
viscosity η

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The wettability of the structural material is of some importance to the heat transfer. It is strongly related to the surface tension of the liquid.

The necessary pumping power is directly influenced by the constants ϱ and η. If electromagnetic pumps are used also the electrical conductivity is discussed.

2.4 Chemical properties and corrosion

Mainly the safety aspect directs our view to the chemical reactions with air and water at the running temperature of the circuit.

The chemical attack on vessels and tubes under defined conditions (purity and velocity of the liquid, purity and composition of the lining material) determines the selection of suitable structural material; at this point also the fabrication technique (welding, cleaning, etc.) has to be kept in mind.

2.5 Economics

The economic considerations primarily ask for price and availability. It has to be emphasized that it often needs some time for a market of new products to be established and rationalized. These circumstances may lead to reasonable prices within some time, also for products which were at first very expensive, e. g. ^{7}Li.

3. Selection by melting point

The "Liquid Metals" comprise by conventional definition all the metals and alloys having a melting point up to that of Al, namely 660 °C. It is useful to arrange all the metallic liquids [1,2] according to their melting point Fp and to divide them into 4 groups:

Group	Fp
1	— 30 °C
2	30 — 150 °C
3	150 — 330 °C
4	330 — 660 °C

The liquids in group 1 are most convenient with regard to the cooling technique. No auxiliary heating is necessary for the start-up of the circuit.

The metals in group 2 need some auxiliary heating, but the expense for it still is relatively low.

The metals of group 3 have a real chance only if all the other properties are very suitable and justify an expensive and difficult heating device.

The metals of group 4 are not reasonable for the present state of reactor development. May be in the future they will come into serious discussion, if the evolution of reactor technology has resulted in some standard-type high temperature power reactors.

Before we arrange all the known metallic liquids into a comprehensive table it is useful and convenient to apply some very simple rules for selection. It needs no discussion that we can omit

- the rare earths
- the noble metals (Ag, Au, Pb, etc.)
- the transition elements (Se, Te, etc.)
- metals with a very high neutron absorption (Cd)

From the binary alloys only those eutectics having an essentially lowered melting point compared to their components (difference at least 10 °C) must be discussed. Also ternary and higher alloys only are important if the melting point lies effectively below that of the simpler alloys.

According to these preliminary rules Table 1 is compiled. No quaternary alloys is contained because not anyone of them fulfills the conditions discussed above (e. g. the Wood-metal, Fp = 60 °C, and the Lipowitz-alloy, Fp = 60 °C, contains Cd!). Although group 4 will not be discussed further at this time the pure metals and some typical binary alloys are adapted in the table for orientation.

Table 1: Low melting metals and alloys

Tab. 1: Niedrigschmelzende Metalle und Legierungen

Group 1 (up to 30 °C)	Fp(°C)	Group 2 (30—150 °C)	Fp(°C)	Group 3 (150—330 °C)	Fp(°C)	Group 4 (330—660 °C)	Fp(°C)
Hg	—38,9	Rb	38,5	In	156,4	Zn	419,5
Cs	28,5	K	63,7	Li	179	Sb	630,5
Ga	29,9	Na	97,8	Sn	231,9	Mg	651
				Bi	271	Al	660,2
				Tl	303		
				Pb	327,4		
Hg–Tl	—59	Na–Tl	63,9	Li–Zn	161	Mg–Zn	343
Hg–Na	—48	Bi–In	72	Ca–Li	165	Al–Zn	382
Cs–Rb	—40	In–Sn	117	Sn–Tl	170	Sb–Zn	414
Cs–K	—37,5	Bi–Pb	125	Pb–Sn	183	Al–Mg	437
Cs–Na	—29	Bi–Sn	139	Bi–Tl	188	Mg–Sb	579
Na–K	—12,3	In–Zn	143,5	Sb–Tl	195		
Na–Rb	— 5			Sn–Zn	198		
Ga–In	—15,7			Mg–Sn	200		
Ga–Sn	20,3			Mg–Tl	203		
				Pb–Sb	252		
				Mg–Pb	253		
				Bi–Zn	255		
		Bi–Sn–In	60,5	Mg–Pb–Sn	166		
		Bi–Pb–Tl	91	Pb–Sb–Zn	239		
		Bi–Pb–Sn	95				
		Bi–Sn–Tl	124				

4. Selection by nuclear properties

The most important properties are the absorption cross section for thermal and fast neutrons, σ_{th} and $\sigma_{100\text{ keV}}$. These data–as to the present knowledge–are compiled for all the elements of group 1, 2 and 3 in Table 2. Considering this table we can introduce a nuclear rule of selection, which explains itself:

$$\sigma_{\text{th}} \leqq 3{,}4 \text{ barn (value of Tl)}$$
$$\sigma_{100\,\text{keV}} \leqq 100 \text{ mb (Ga: 96 mb)}$$

Applying this rule the metals with normal isotope composition Li, Cs, In, Sb and Hg must be dropped. Also their alloys in table 1 do not match the rule, excluding the eutectics Pb-Sb and Pb-Sb-Zn, and are omitted.

Table 2: Neutron absorption of the low melting pure metals

Tab. 2: Neutronenabsorption der niedrigschmelzenden Reinmetalle

	σ_{th} barn	$\sigma_{100\text{ keV}}$ mb		σ_{th} barn	$\sigma_{100\text{ keV}}$ mb
Li	71	~1000	Sn	0,625	45
Na	0,536	1,1	Pb	0,170	4
K	2,07	5,5	Sb	5,7	180
Rb	0,73	50 *)	Bi	0,034	3
Cs	29,0	220 *)			
Mg	0,063	2	Zn	1,10	20
Ca	0,44	1,5 *)	Hg	380	60
Ga	2,80	96			
In	196	380			
Tl	3,4	24			

*) at 400 keV

Now the question arises which of the omitted elements have *suitable isotopes with a low cross section*. The two following ones obey the mentioned rule:

	Abundance %	σ_{th} barn	$\sigma_{100\ keV}$ mb
^{7}Li	92,48	0,033	0,025
^{204}Hg	6,85	0,43	0,1

They are taken up again and it results from the nuclear point of view finally the list of metals and alloys in Table 3.

Table 3: Low melting metals and alloys with $\sigma_{th} \leqq 3{,}4$ barn or $\sigma_{100\ keV} < 100$ mb

Tab. 3: Niedrigschmelzende Metalle und Legierungen mit $\sigma_{th} \leqq 3{,}4$ barn bzw. $\sigma_{100\ keV} < 100$ mb

Group 1 (up to 30 °C)	Fp(°C)	Group 2 (30—150 °C)	Fp(°C)	Group 3 (150—330 °C)	Fp(°C)
Ga	29,9	Rb	38,5	Sn	231,9
		K	63,7	Bi	271
		Na	97,8	Tl	303
				Pb	327,4
Na–K	—12,3	Na–Tl	63,9	Sn–Tl	170
Na–Rb	— 5	Bi–Pb	125	Pb–Sn	183
Ga–Sn	20,3	Bi–Sn	139	Bi–Tl	188
				Sn–Zn	198
				Mg–Sn	200
				Mg–Tl	203
				Pb–Sb	252
				Mg–Pb	253
				Bi–Zn	255
		Bi–Pb–Tl	91	Mg–Pb–Sn	166
		Bi–Pb–Sn	95	Pb–Sb–Zn	239
		Bi–Sn–Tl	124		
^{204}Hg	—38,9			^{7}Li	179
^{204}Hg–Tl	—59			^{7}Li–Zn	161
^{204}Hg–Na	—48			^{7}Li–Ca	165

5. *Suitable structural material*

The practical application of a metallic fluid needs a structural material for vessels and tubes which resists the corrosion attack *at the provided temperature for a sufficiently long time*. At low temperatures (below ca. 300 °C) the commonly used iron-based materials are sufficiently resistant in most cases. At temperatures above 400 °C, however, a very accurate selection according to metallurgical characteristics has to be made.

Especially in the U.S.A. numerous experimental investigations in this field have been carried out. It is feasible to use these results (at first published in [3]) for compiling a table of resistance, *Table* 4. Besides the most fitting materials it also contains the highest temperature permitted (or tested). Moreover the last column brings all those materials which certainly cannot be used because they are dissolved in the *liquid metal to a remarkable extent*.

The container material for a liquid alloy must be also resistant against the components of the alloy. This necessary (but not always sufficient) condition–together with Table 4 –leads to a preliminary judgement also for alloys.

In the list of the liquid metals and alloys in Table 3 there are (besides the liquids taken into account in Table 4) the metals Rb and Ca which for completion must be discussed:

The alkaline metal Rb behaves qualitatively similar to the "*partners*" Na and K. But the chemical reactivity and therefore the corrosion attack is still somewhat higher.

Table 4: Structural material

Tab. 4: Strukturmaterial

Liquid *metal*	Suitable structural *material*	Temp. limit °C	Material certainly not suitable
Na, K	Fe, steels, Co, Ni	900	Si
	Nb, Ta, Mo, W	900	
Li	Fe, Nb, Ta, Mo	900	Mn, Si, Cu
	ferr. Cr-steels	600	
Mg	Fe, Cr-steels (Cr)	650	Ni, Co, Mn, Si, Cu
	Be, Ti (Nb, Ta, Mo, W)	750	
Zn	Mo–Fe-alloy	420	Ni, Co, Mn, Cu
Hg	Cr-steels	650	Ni, Co, Mn, Cu, Zr
	W, Mo	600	
Ga	W	800	Fe, Ni, Mn, V, Cu, Zr
	Ta	450	
Tl	Mn	1000?	Ni, Cu
	steels	600	
Sn	Be, Ti	500	Ni, Co, Cu
Pb	Nb, Ta	1000	Mn, Cu, W
	steels, Be	600	
Sb	no material known		Ni, Co, Cr, Mn, Cu
Bi	Fe, C-steel, Cr, Nb	700	Mn, Cu
	Mo, Be	1000	

The alkaline earth metal Ca is in chemical relation to Mg. The properties are similar. It is supposed to be a little more aggressive than Mg in the liquid or alloyed states.

In the selection of the most fitting structural material also–besides the metallurgical behaviour–the nuclear data are *to be considered particularly for constructions within the* core of a reactor. For the first orientation Table 5 shows the neutron absorption cross sections *of all materials discussed* in Table 4.

6. Further selection by different points of view

The metals and alloys in Table 3 *still contain someones without essential importance:*

Rb has no advantage compared to Na, K and Na-K, but it is more aggressive and expensive.

The alloy Ga-Sn lowers the melting point only by a few degrees, but need a structural material resistant to both components. ^{204}Hg-Tl and ^{204}Hg-Na would additionally complicate the (highly utopian) use of ^{204}Hg because of the corrosion attack, and the lowered melting point would be of no additional value in this temperature range below 0 °C.

Table 5: Neutron absorption of structural materials

Tab. 5· Neutronenabsorption der Strukturmaterialien

Atom-Nr.	Element	σ_{th} barn	$\sigma_{100\ keV}$ mb
4	Be	0,010	0*)
22	Ti	5,8	6
23	V	4,98	9,5
24	Cr	3,1	5,5
25	Mn	13,2	25,6
26	Fe	2,53	6,1
27	Co	37,0	11,5
28	Ni	4,25	12,6
40	Zr	0,180	15,1
41	Nb	1,15	100
42	Mo	2,7	71
73	Ta	21	325
74	W	19,2	178

*) The (n, α)-threshold energy is about 1 MeV

Table 6: Liquid metals and alloys, further selection
Tab. 6: Flüssige Metalle und Legierungen, weitere Auswahl

Group 1 (up to 30 °C)	Fp(°C)	Group 2 (30—150 °C)	Fp(°C)	Group 3 (150—330 °C)	Fp(°C)
Ga	29,9	K	63,7	Sn	231,9
		Na	97,8	Bi	271
				Tl	303
				Pb	327,4
Na–K	—12,3	Bi–Pb	125	Sn–Tl	170
		Bi–Sn	139	Pb–Sn	183
				Bi–Tl	188
				Mg–Sn	200
				Mg–Tl	203
				Mg–Pb	253
		Bi–Pb–Tl	91	Mg–Pb–Sn	166
		Bi–Pb–Sn	95		
		Bi–Sn–Tl	124		
^{204}Hg	—38,9			^{7}Li	179

The alloy Na-Tl does not avoid the difficulties with the alkalines, brings a high cross section because of Tl and has about the same melting point as K.

The Zn-alloys in group 3 are very difficult to handle above 400 °C–because of Zn–and they do not bring any remarkable advantage, e. g. in Fp.

For Sb hardly a structural material sufficiently resistant is known.

Ca only in the ^{7}Li-Ca-alloy is in discussion. But it would not be reasonable to raise the very low cross section of ^{7}Li in order to lower Fp from 186 °C to 165 °C.

All these peculiar metals and alloys in Table 3 can now be dropped. It remains the very diminished compilation in Table 6.

7. Chemical behaviour of the remaining liquids

Above all, one has to look for chemical reactions with air and water, then for useful cover gases. Also any possible toxicity or other particular safety requirement is important.

All the metallic elements contained in Table 6 are now briefly discussed. It is advantageous to arrange them by their chemical relationship:

Lithium

reacts only a little with air and water at room temperature and does not reach the melting point by this reaction;
does not react with dry O_2 below 100 °C;
maybe ignites in air near or above the melting point and then burns with brilliant white flames;
reacts with wet N_2, especially in the fluid state;
in the fluid state dissolves C and Ni from the steels (above ca. 500 °C).

Burning Li reacts with sand, graphite powder is suitable for extinguishing fire. For cover gases only the noble gases are applicable.

Sodium

ignites in air already at slightly elevated temperatures, especially if some humidity is present.

Liquid sodium ignites in a normal atmosphere, reacts with water explosively, neither reacts with pure nitrogen nor with CO_2 at room temperature. As cover gases beyond the noble gases absolutely pure N_2 can be used. No special toxicity is known.

Against Na-fire only cover material like alkaline chlorides, graphite powder and Na_2CO_3 are helpful.

Potassium

has a behaviour similar to Na. It is still somewhat more reactive and maybe ignites in air already at room temperature. As far as the suitable cover gases and safety requirements are concerned it is the same situation as with Na.

Magnesium

in normal atmosphere is covered at once with a protecting oxide film, to a little extent also reacts with N_2, in powder form ignites at some higher temperature.

Molten Mg vigorously reacts with O_2 and H_2O and burns in a humid atmosphere. Additions of Pb and Sn intensify oxidation, Be and Ca reduce it.

Cover gas: Noble gases.

MgO-dust and Mg-ions in higher concentration are somewhat toxic.

Mercury

in a very pure state does not react with air and water save at relatively high temperatures;
reacts in pure O_2 above 350 °C;
vigorously reacts with Na and K.
Cover gas: N_2, noble gases;

Mercury-vapour is said to be very toxic, especially after long-term exposure. The physiological influences are still increased in a humid and CO_2-containing atmosphere.

Gallium

This extremely corrosive metal is rather unreactive in air and water, it wets many non-metals, in a very pure state it can be subcooled up to –20 °C.
Ga is hardly toxic.
Cover gases: Noble gases, N_2, CO_2

Thallium

is oxidized at room temperature rather slowly, above some 100 °C somewhat faster;
does not react with H_2 and N_2;
decomposes water at high temperatures.

Tl is very toxic. Especially Tl_2O which is soluble in water, causes chronic poisoning.
Cover gases: Noble gases, N_2 and possibly H_2.

Tin

is stable in dry air below fusion and is oxidized slowly at fusion;
reacts with water above 650 °C;
is oxidized by CO_2 above 550° C;
ignites above some 1500 °C;
is not toxic.
Cover gases: Noble gases, N_2, H_2, possibly CO_2.

Lead

is very stable in dry air;
is covered by a protecting carbonate film in humid air containing CO_2;
is oxidized above 300 °C, particularly above fusion;
ignites above some 700 °C;
is oxidized by H_2O at very high temperatures;
is slightly toxic in every state, especially as vapour and dust.
Cover gases: Noble gases, N_2, H_2, possibly CO_2.

Bismuth

is oxidized in humid air at room temperature and reacts rather fast above fusion;

ignites at higher temperatures;

reduces H_2O and CO_2 above 600 °C.

Bi itself is supposed to be not toxic, but by neutron activation Po is formed which is α-active.

Cover gases: Noble gases, N_2, H_2.

8. Activation in neutron flux

In neutron flux the metallic fluid is activated mainly by (n, γ)-absorption. At saturation the activity C (Curie/cm³) is given by the formula:

$$C = \frac{1}{3{,}7 \cdot 10^{10}} \cdot a \cdot N \cdot \sigma_{n\gamma} \cdot \Phi$$

where a abundance of the initial isotope in mole-parts
N initial number of atoms per cm³
$\sigma_{n\gamma}$ absorption cross section in cm²
Φ neutron flux in $cm^{-2} \cdot sec^{-1}$

For numerical evaluation we take $\sigma_{n\gamma}$ at 100 keV and $\Phi = 5 \cdot 10^{15}/cm^2 \cdot sec$.

The results for all pure metals of Table 6 are compiled in Table 7. The alloys can be calculated by combining the values of the components according to the composition. As a reasonable temperature in most cases 600 °C was chosen. Only these isotopes of the metallic fluids need to be discussed which are not transferred into a stable isotope by (n, γ)-reaction. From Hg and Li only the isotopes mentioned in Table 6 are considered. As the isotope cross sections for 100 keV-neutrons are exactly known only in a few cases, most of them are estimated.

The following data for the activity after (n, γ)-reaction are fixed in Table 7:

saturation activity C
halflife $t_{1/2}$
(in the case of two isomeric nuclei only that one is cited which is preferentially formed by the (n, γ)-reaction);
type of radiation at decay
gamma quants per decay in %
(as known from literature—see[4]—gammas below 0,05 MeV have been dropped);
gamma energy;
number of gammas per cm³ and sec;
radioactive decay products and their decay properties.

9. Survey of properties

For an evaluation of the so-called "applicability" of the different fluids the physical and technical properties, important for their use in reactor technology, will be compared. As for most of the alloys only a few experimental data are known a lot of values are calculated using the data of the components according to the following formulae:

Density of binary alloys:

$$\varrho = \frac{\varrho_1 \cdot \varrho_2}{g_1 \varrho_2 + g_2 \varrho_1}$$

Table 7: Activation in neutron flux Tab. 7: Aktivierung im Neutronenfluß

	N at 600 °C $10^{22}/cm^3$	Atom- $\sigma_{n\gamma}$ (100 keV) mb	Natural isotope	Abundance a %	Isotope $\sigma_{n\gamma}$ (100 keV) mb	C $\frac{Curie}{cm^3}$	$t_{1/2}$	Activation after (n, γ)-reaction Radiation	$\frac{\gamma}{decay}$ %	γ-energy MeV	$\frac{\gamma}{cm^3 sec}$ in 10^{10}	Radioactive successors
Li	4,11		^7Li	100[4]	0,025	0,14[4]	0,8 s	α, β			0	
Na	2,11	1,1	^{23}Na	100	1,1	3,14	15 h	β, γ	100 100	2,75 1,34	23,2	
Mg	3,90[1]	2	^{26}Mg	11,06	9	5,2	9,5 m	β, γ	30 70	1,02 0,84	19,2	
K	1,08	5,5	^{39}K	93,23	5,5*)	(7,0)[5]	$1{,}3 \cdot 10^9$ a	β, γ	11	1,46	(0,29)[5]	
			^{41}K	6,76	5,5	0,5	12,5 h	β, γ	18	1,53	0,33	
Ga	4,95	96	^{69}Ga	60,1	120	482	21 m	β, γ	~1,2	≦1,04	21,4	
			^{71}Ga	39,9	60	160	14.3 h	β, γ	~200	≦3,35	~1200	
Sn	3,41	45	^{112}Sn	1,02	100*)	4,7	119 d	γ	3 (?)	0,26	18[6]	^{113m}In; 104 m; 100 % γ; 0,39 MeV
			^{116}Sn	14,3	1*)	0,7	14 d	γ	≦200	≦0,32	≦5	
			^{118}Sn	24,1	1*)	1,1	245 d	γ	200	≦0,07	8	
			^{120}Sn	32,5	10*)	15	27,5 h	β			0	
			^{122}Sn	4,8	55	12,2	40 m	β, γ	100	0,15	45,1	
			^{124}Sn	6,1	15*)	4,2	10 m	β, γ	~100	0,33	16	^{125}Sb; 2 a, γ; ≦ 0,64 MeV
Hg	3,86[2]	60	^{204}Hg	100[4]	0,1*)	0,5[4]	5,5 m	β, γ	100 (?)	0,203	~2[4] (?)	
Tl	3,32[1]	24	^{203}Tl	29,5	19	25,7	3,6 a	β			0	
			^{205}Tl	70,5	26	82,1	4,2 m	β			0	
Pb	2,98	4	^{204}Pb	1,37	0,5*)	(0,03)[5]	$5 \cdot 10^7$ a	γ			~0	
			^{206}Pb	25,0	0,02*)	0,02	0,8 s	γ	100 100	1,06 0,57	0,15	
			^{208}Pb	52,4	7,5[3]	15,8	3,3 h	β			0	^{209}Bi
Bi	2,78	3	^{209}Bi	100	3	11,3	5 d	α, β			0	^{210}Po 138 d; 100 % α; 5,3 MeV

[1] at melting point. [2] at 300 °C [3] at 200 keV. [4] only pure isotope considered. [5] saturation never reached. [6] including gammas from ^{113m}In.
*) roughly estimated.

Density of ternary alloys:

$$\varrho = \frac{\varrho_1 \cdot \varrho_2 \cdot \varrho_3}{g_1 \varrho_2 \varrho_3 + g_2 \varrho_1 \varrho_3 + g_3 \varrho_1 \varrho_2}$$

Cross sections of binary (and ternary) alloys:

$$\sigma = n_1 \sigma_1 + n_2 \sigma_2 (+ n_3 \sigma_3)$$

Saturation activity of binary (and ternary) alloys:

$$C = v_1 C_1 + v_2 C_2 (+ v_3 C_3)$$

An analogous formula is due to γ/cm^3 sec.

In these formulae the concentration of the components is given by weight (g), or by mole (n), or by volume (v), always in %/100. The correlations of n, v and g are given

for binary alloys by:

$$n_1 = \frac{g_1 A_2}{g_1 A_2 + g_2 A_1} \qquad n_2 = 1 - n_1$$

$$v_1 = \frac{g_1 \varrho_2}{g_1 \varrho_2 + g_2 \varrho_1} \qquad v_2 = 1 - v_1$$

for ternary alloys by:

$$n_1 = \frac{g_1 A_2 A_3}{g_1 A_2 A_3 + g_2 A_1 A_3 + g_3 A_1 A_2}$$

$$n_2 = \frac{g_2 A_1 A_3}{g_1 A_2 A_3 + g_2 A_1 A_3 + g_3 A_1 A_2}$$

$$n_3 = 1 - n_1 - n_2$$

$$v_1 = \frac{g_1 \varrho_2 \varrho_3}{g_1 \varrho_2 \varrho_3 + g_2 \varrho_1 \varrho_3 + g_3 \varrho_1 \varrho_2}$$

$$v_2 = \frac{g_2 \varrho_1 \varrho_3}{g_1 \varrho_2 \varrho_3 + g_2 \varrho_1 \varrho_3 + g_3 \varrho_1 \varrho_2}$$

$$v_3 = 1 - v_1 - v_2$$

where A_1, A_2, A_3 are the atomic weights of the components. Numerical results of this correlation for all the binary and ternary alloys are given in Table 8.

Table 8: The components of the alloys
Tab. 8: Die Komponenten der Legierungen

	$^1/_{100}$ weight-%			$^1/_{100}$ mole-%			$^1/_{100}$ volume-%		
	g_1	g_2	g_3	n_1	n_2	n_3	v_1	v_2	v_3
Na–K	0,228	0,772		0,334	0,666		0,203	0,797	
Bi–Pb	0,555	0,445		0,552	0,448		0,570	0,430	
Bi–Sn	0,57	0,43		0,429	0,571		0,480	0,520	
Sn–Tl	0,565	0,435		0,691	0,309		0,680	0,320	
Pb–Sn	0,381	0,619		0,261	0,739		0,287	0,713	
Bi–Tl	0,475	0,525		0,470	0,530		0,506	0,494	
Mg–Sn	0,02	0,98		0,091	0,909		0,080	0,920	
Mg–Tl	0,03	0,97		0,207	0,793		0,175	0,825	
Mg–Pb	0,03	0,97		0,208	0,792		0,166	0,834	
Bi–Pb–Tl	0,552	0,333	0,115	0,550	0,334	0,116	0,571	0,324	0,105
Bi–Pb–Sn	0,52	0,32	0,16	0,462	0,287	0,251	0,495	0,286	0,219
Bi–Sn–Tl	0,500	0,357	0,143	0,392	0,493	0,115	0,439	0,450	0,111
Mg–Pb–Sn	0,07	0,20	0,73	0,288	0,095	0,617	0,249	0,114	0,637

Now the survey of properties can be made. The next tables contain all data compiled [3, 5 and 6]. Table 9 brings the physical properties in solid and fluid states, Table 10 heat transfer constants, Table 11 chemical and metallurgical behaviour, and Table 12 nuclear data.

Table 9: Physical properties
Tab. 9: Physikalische Eigenschaften

	Density at 600 °C [g/cm³]	Fp [°C]	Sp [°C]	Volume increase at 600 °C [10^{-6}/°C]	Volume change at melting %	Vapor pressure at 600 °C [Torr]
Ga	5,72	29,9	1983	104	— 3,1	ca. 10^{-8}
K	0,70	63,7	758	335	+ 2,4	128
Na	0,81	97,8	881	247	+ 2,5	26
Sn	6,71	231,9	2270	117	+ 2,6	ca. 10^{-10}
Bi	9,66	271	1477	131	— 3,32	$6 \cdot 10^{-4}$
Tl	10,93*)	303	1457	ca. 100	+ 3,2	$3 \cdot 10^{-2}$
Pb	10,27	327,4	1737	115	+ 3,6	$3 \cdot 10^{-4}$
^{204}Hg	12,20*)	–38,9	357	ca. 180	+ 3,6	22 Atm
^{7}Li	0,47	179	1317	176	+ 1,5	$5 \cdot 10^{-2}$

*) extrapolated

Table 10: Thermal and flow properties at about 600 °C
Tab. 10: Wärme- und stromungstechnische Eigenschaften bei etwa 600 °C

	λ $\left[\frac{\text{cal}}{\text{cm sec °C}}\right]$	c_p $\left[\frac{\text{cal}}{\text{g °C}}\right]$	ϱ [g/cm³]	$\varrho \cdot c_p$ $\left[\frac{\text{cal}}{\text{cm}^3\ \text{°C}}\right]$	η [cPoise]
Ga	0,09	0,08	5,72	0,46	0,7
K	0,084	0,183	0,70	0,13	0,15
Na	0,150	0,300	0,81	0,24	0,21
Sn	0,08	0,07	6,71	0,47	1,0
Bi	0,037	0,038	9,66	0,37	1,0
Pb	0,036	0,038	10,27	0,39	1,5
Tl	0,05	0,035	10,9	0,38	
^{204}Hg	0,02	0,03	12,2	0,37	
^{7}Li	0,07	1,0	0,47	0,47	0,3

Table 11: Chemistry and metallurgy
Tab. 11: Chemie und Metallurgie

	Reaction with air and water at 600 °C	Structural material steel up to °C	other materials	up to °C	Toxicity
Ga	weak	200	W	800	not toxic
K	explosive	900	Nb, Ta, Mo, W	900	fire hazard
Na	explosive	900	Nb, Ta, Mo, W	900	fire hazard
Sn	medium	(200)	Be, Ti	500	not toxic
Bi	strong	700	Mo, Be	1000	Po: α-radiation
Tl	medium	(600)	(Mn)	(1000)	very toxic
Pb	medium	600	Nb, Ta	1000	dust, vapour
^{204}Hg	weak	650	W, Mo	600	vapour
^{7}Li	explosive	800	Nb, Ta, Mo	900	fire hazard

Table 12: Nuclear properties
Tab. 12: Kernphysikalische Eigenschaften

	at 100 keV				Inelast.	Saturation activity	
	σ_{th} barn	$\sigma_{100\,\text{keV}}$ mb	σ_s barn	ξ	threshold MeV	$\frac{\gamma}{\text{cm}^3\ \text{sec}}$	$t_{1/2}$ $\leqq$
Ga	2,80	96	7	0,0283	0,322	∼1220	14,3 h
K	2,07	5,5	3	0,0503	0,028	0,33	12,5 h
Na	0,536	1,1	3,5	0,0847	0,439	23,2	15 h
Sn	0,625	45	6,4	0,0167	0,024	≦ 92	245 d
Bi	0,034	3	9,8	0,0095	0,91	0	5 d
Tl	3,4	24	10	0,0098	0,205	0	3,6 a
Pb	0,170	4	11	0,0096	0,26	∼ 0	3,3 h
^{204}Hg	0,43	0,1		0,0098	0,43	∼ 2	5,5 m
^{7}Li	0,033	0,025	1,1	0,266	0,477	0	0,8 sec

10. Discussion of applicability

It can be tried to discuss the idea of applicability which is not very clearly to define by using a point system. All properties pertaining to construction and operation of a cooling circuit are included in the judgement.

The cooling material Na which is examinated most of all metallic fluids is taken as a standard for comparison. Therefore, Na gets 0 points for all properties. The other metals or alloys receive 1 or 2 positive or negative points, if a certain property is better or worse than in Na. In summing up the most important items, namely melting point, absorption cross section and structural material get a weighting factor of 2.

In detail the following rules have been applied in make-up this judgement:

melting point Fp

Na and the other liquid metals of group 2 get 0 points, group 1 gets +1, group 3 gets −1.

boiling point Sp

700—900 °C: 0
above 1300 °C: +1
Hg (357 °C): −2

volume change at fusion ΔV:

volume increase: 0
volume decrease: −1

absorption cross section σ_{abs}:

^{7}Li : +1
$\sigma_{th} \leqq 0{,}536$ barn (Na) and $\sigma_{100\,keV} \leqq 4$ mb (Pb): 0
$\sigma_{th} \leqq 2{,}07$ barn (K) and $\sigma_{100\,keV} \leqq 30$ mb: −1
$\sigma_{th} > 2{,}07$ barn or $\sigma_{100\,keV} > 30$ mb: −2

logarithmic energy decrement:

^{7}Li : −1
all the others: 0

activation:

more than $50 \cdot 10^{10}$ γ/cm³ sec: −1
5—$50 \cdot 10^{10}$ γ/cm³ sec: 0
below $5 \cdot 10^{10}$ γ/cm³ sec: +1

thermal conductivity λ:

$\lambda \geqq 0{,}07 \dfrac{\text{cal}}{\text{cm sec}^\circ\text{C}}$: 0
$\lambda < 0{,}07$ *and all binary alloys:* −1
ternary alloys: −2

specific heat per unit volume $\varrho \cdot c_p$:

$\geqq 0{,}4$ cal/cm³ °C: +1
0,2—0,4: 0
< 0,2: −1

pumping power:

The pumping power is closely related to the density; therefore:

$\varrho \leqq 1$: 0
$1 < \varrho \leqq 7$: −1
$\varrho > 7$: −2

chemical behaviour:

alkalines: 0
all the rest: +1

structural material:

alkalines: 0
Bi, Tl, Pb, Hg, and Bi-Pb: −1
all the others: −2

toxicity and handling:

not toxic, not caustic: +1
not toxic, but difficult to handle: 0
toxic: −1

price and availability:

below about $10.—/kg, available: 0
above $10.—/kg, available: −1
very expensive and hardly available: −2

Table 13: Judgement of applicability relative to Na
Tab. 13: Beurteilung der Brauchbarkeit relativ zu Na

	Fp	Sp	ΔV	σ_{abs}	ξ	Activation	λ	$\varrho \cdot c_p$	Pumping power	Chemistry	Structural mat.	Toxocity	Price	
Weight	2x	1x	1x	2x	1x	1x	1x	1x	1x	1x	2x	1x	1x	Sum
Ga	+1	+1	−1	−2	0	−1	0	+1	−1	+1	−2	+1	−2	− 7
K	0	0	0	−1	0	+1	0	−1	0	0	0	0	0	− 2
Na	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Sn	−1	+1	0	−2	0	−1	0	+1	−1	+1	−2	+1	0	− 8
Bi	−1	+1	−1	0	0	+1	−1	0	−2	+1	−1	−1	0	− 6
Tl	−1	+1	0	−2	0	+1	−1	0	−2	+1	−1	−1	−1	−10
Pb	−1	+1	0	0	0	+1	−1	0	−2	+1	−1	0	0	− 4
^{204}Hg	+1	−2	0	0	0	+1	−1	0	−2	+1	−1	−1	−2	− 6
^{7}Li	−1	+1	0	+1	−1	+1	0	+1	0	0	0	0	−2	0
Na–K	+1	0	0	−1	0	+1	−1	−1	0	0	0	0	0	− 1
Bi–Pb	0	+1	0	0	0	+1	−1	0	−2	+1	−1	−1	0	− 3
Bi–Sn	0	+1	0	−1	0	0	−1	+1	−2	+1	−2	−1	0	− 7
Sn–Tl	−1	+1	0	−2	0	0	−1	+1	−2	+1	−2	−1	−1	−12
Pb–Sn	−1	+1	0	−2	0	0	−1	+1	−2	+1	−2	0	0	−10
Bi–Tl	−1	+1	0	−1	0	+1	−1	0	−2	+1	−2	−1	−1	−10
Mg–Sn	−1	+1	0	−2	0	−1	−1	+1	−1	+1	−2	+1	0	− 9
Mg–Tl	−1	+1	0	−2	0	+1	−1	0	−2	+1	−2	−1	−1	−12
Mg–Pb	−1	+1	0	0	0	+1	−1	0	−2	+1	−2	0	0	− 6
Bi–Pb–Tl	0	+1	0	−1	0	+1	−2	0	−2	+1	−2	−1	0	− 8
Bi–Pb–Sn	0	+1	0	−1	0	0	−2	0	−2	+1	−2	−1	0	− 9
Bi–Sn–Tl	0	+1	0	−1	0	0	−2	0	−2	+1	−2	−1	0	− 9
Mg–Pb–Sn	−1	+1	0	−1	0	−1	−2	+1	−1	+1	−2	0	0	− 9

Table 14: Final selection
Tab. 14: Letzte Auswahl

	Applicability	Fp °C	Structural material	Highest temperature permitted [°C]
Na	0	97,8	steel, Nb, Ta, Mo	900
^{7}Li	0	179	steel, Nb, Ta, Mo	800—900
Na-K	− 1	−12,3	steel, Nb, Ta, Mo	900
K	− 2	63,7	steel, Nb, Ta, Mo	900
Bi-Pb	− 3	125	steel, Be, Ti	600—700
Pb	− 4	327,4	steel, Nb, Ta	600—1000
^{204}Hg	− 6	−38,9		
Mg-Pb	− 6	253		
Bi	− 6	271		
Ga	− 7	29,9		
Bi-Sn	− 7	139		

In Table 13 now all these points for the individual properties are summarized. Summing up the last column then gives a relative value of applicability. Using this judgement on the basis of Table 6 final selection is made. The liquid metals are arranged by their values of applicability. The last table, 14, contains all fluids up to a value of −7. The upper group (up to −4) includes the metals which indeed have been under discussion and examination for a long time. The lower group is supposed not to be in practical consideration.

(Eingegangen am 24. 1. 1964)

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