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On the use of tin–lithium alloys as breeder material for blankets of fusion power plants

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Abstract

Tin–lithium alloys have several attractive thermo-physical properties, in particular high thermal conductivity and heat capacity, that make them potentially interesting candidates for use in liquid metal blankets. This paper presents an evaluation of the advantages and drawbacks caused by the substitution of the currently employed alloy lead–lithium (Pb–17Li) by a suitable tin–lithium alloy: (i) for the European water-cooled Pb–17Li (WCLL) blanket concept with reduced activation ferritic–martensitic steel as the structural material; (ii) for the European self-cooled TAURO blanket with SiC_f/SiC as the structural material. It was found that in none of these blankets Sn–Li alloys would lead to significant advantages, in particular due to the low tritium breeding capability. Only in forced convection cooled divertors with W-alloy structure, Sn–Li alloys would be slightly more favorable. It is concluded that Sn–Li alloys are only advantageous in free surface cooled reactor internals, as this would make maximum use of the principal advantage of Sn–Li, i.e., the low vapor pressure. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Tin–lithium alloys seem to have several attractive thermo-physical properties that would make them potentially interesting candidates for use in liquid metal blankets. Their low vapor pressure [4] appears particularly suitable for wet wall blanket concepts. The thermal conductivity and heat capacity can be assumed to be similar to Sn and thus about twice as high as that for the currently employed alloy Pb–17Li (lead with 17 at.% lithium). These considerations have triggered an assessment to check whether it might be useful to substitute a suitable tin–lithium alloy for the currently employed alloy Pb–17Li in:

- the European water-cooled lithium-lead (WCLL) blanket [1] with reduced activation ferritic-martensitic steel as the structural material,
- the European self-cooled TAURO blanket [2] with SiC_f/SiC as the structural material.

Both blankets are currently being considered for use in a fusion power plant [5]. The nuclear power deposition and tritium breeding ratio (TBR) was calculated for various lithium concentrations and ⁶Li enrichments. The consequences of using tin–lithium alloys in forced convection-cooled divertors are also briefly addressed.

2. Comparison of liquid metal properties

Table 1 compares some thermo-physical properties of Sn and Pb–17Li to those estimated for Sn–20Li (or similar). Thermodynamically, an alloy with about 20 at.% Li has about the same Li activity as Pb–17Li, which justifies the assumption that it may have a similar

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| | Sn | Sn-20Li [4] | Pb-17Li [3] |
|------------------------------|-------|---------------|-------------|
| Melting point (°C) | 232 | ≈330 | 235 |
| Density (g/cm ³) | 6.8 | ≈ 6.2 | 9.5 |
| Li concentration (at.%) | 0 | 20 | 17 |
| Thermal conductivity (W/m K) | 33.5 | | 14.2 |
| Specific heat (J/g K) | 0.318 | | 0.189 |

Table 1 Comparison of some thermo-physical properties of Sn, Sn-20Li and Pb-17Li at 350°C

reactivity with water. This safety consideration led to the choice of Sn–20Li as the reference Sn–Li alloy for this comparison.

A significant difference has been observed in the vapor pressure of Sn–Li alloys compared to Pb–17Li, which would enable the use of Sn–Li as a liquid metal directly exposed to the plasma, e.g., in wet wall blanket concepts for tokamaks or inertial confinement machines.

2.1. Compatibility with structural materials

Materials in contact with liquid metals are subject to corrosion and embrittlement. Present knowledge of materials compatibility with Sn–Li alloys are very limited due to the lack of experimental data. Nevertheless, some information can be drawn from studies carried out in molten Sn.

Ferrous metals (cast iron, low-carbon steel), austenitic and ferritic stainless steels show a poor resistance to attack at 400°C [10,11]. When exposed to liquid Sn, they react to form Fe-Sn compounds, e.g., FeSn₂, the wellknown 'tinning' reaction, so that liquid Sn or alloys containing Sn cannot be used as coolants in ferritic or austenitic systems at any temperatures substantially above the coolant melting point. Most of the metals having an appreciable solubility in Sn (e.g., Ni, Al, Co, Cu etc.) may also be excluded. In particular, Ni-base alloys are not suitable for handling molten Sn. It is reported that Cr could be resistant to attack (at moderate temperatures). Most refractory metals such as W, Mo, Ta, Ti and Nb are compatible with Sn up to rather high temperatures (>800°C). Many ceramics show good resistance to molten Sn [12]. No reaction is observed with oxides (Al₂O₃, SiO₂), nitrides (BN, Si₃N₄) and carbides (TiC, WC). SiC could also be resistant to attack depending on the porosity. It is currently not clear whether protective ceramic coatings on steel could perform well enough to allow use of steels with Sn-Li alloys.

Another critical problem is the embrittlement of materials [13]. Studies have demonstrated the embrittling effect of Sn in a range of austenitic steels [14]. Embrittlement was shown to depend on the presence of a tensile stress and to be associated with intercrystalline penetration. Other work reported no embrittlement of quenched and tempered steel. Conflicting evidence exists regarding the effects of Sn in the embrittlement behavior. It could also depend on temperature. Therefore, no clear conclusion can be drawn, but the phenomenon has to be considered when using steels.

From this broad outline related to molten Sn, it can be deduced that unprotected Fe and Ni-base alloys are not suitable for exposure to liquid Sn–Li in the temperature range 400–500°C. It seems possible to use refractory metals such as W, and SiC could be also compatible. However, the microstructural characteristics of those materials (grain size, impurities, porosity) can significantly influence their behavior in Sn–Li. Therefore, experiments in liquid Sn–Li would be necessary to provide additional information.

2.2. Interaction between Sn-20Li and water

The direct contact between a hot liquid metal and cold water (the temperature of the liquid metal being significantly higher than the saturation temperature of water) can lead to a sharp pressure peak within the interaction zone, adding to pressurization. Hence, a large water leak in a water-cooled liquid metal blanket would lead to pressurization of the blanket module. The pressure evolution directly depends on the ability of the interaction zone to expand and on the exchange area between the liquid metal and the water. Generally speaking, the more confined the energy the higher the pressure peak. The evolution of the exchange area is mainly governed by fragmentation processes within the metal. Only the part of the metal, which has been divided into small fragments (equivalent diameter of about 250 µm) is likely to lead to an explosive interaction.

Some experiments were performed at the University of Wisconsin [15] in order to investigate the interaction between molten tin and water for different initial and boundary conditions (initial mass and temperature of tin and water, external trigger etc.). In these experiments, less than 10% of the tin was finely fragmented. Starting at near atmospheric pressure, the resulting pressure peaks ranged between 3 and 10 MPa depending on the experimental conditions. These values show that a possible interaction between tin and water should be an important concern for a tin–lithium breeder concept.

3. Use of Sn-Li alloys in the WCLL blanket

When substituting the Pb–17Li by a suitable Sn–Li alloy in the WCLL blanket, two major issues must be solved in addition to the Sn–Li/water interaction and material compatibility questions; namely TBR, power deposition and redefinition of the coolant conditions.

3.1. TBR and power deposition

A comparison of the cross-sections for the (n,γ) reaction has shown that the average for Sn–20Li is about two orders of magnitude higher than that for Pb–17Li. On the other hand, the cross-section for the (n,T) breeding reaction is basically the same for both alloys, and depends on the Li concentration. The (n,2n) cross-section of Sn is lower than for Pb. Overall, compared to Pb–17Li we expect a higher neutron absorption in Sn–20Li and, consequently, a lower TBR.

To confirm these considerations, a three-dimensional model for the WCLL blanket for DEMO [6] was used and the data compared between Pb–17Li (90 at.% ⁶Li enrichment) and Sn–Li alloys with different Li concentrations. Fig. 1 compares the results for different Sn–Li alloys to the result obtained for Pb-17Li. In both cases 90 at.% ⁶Li enrichment was assumed.

The TBR of Pb–17Li reaches a value of 1.14 whereas a Sn–17Li alloy would only have a TBR of 0.89. The reference Sn–20Li alloy would improve TBR to 0.93, but more than 33 at.% Li is necessary to fulfill the requirement of TBR > 1.05.

Isotopic tailoring of the Sn was proposed [7] to raise the TBR performance. There are 10 natural Sn isotopes with the abundances listed in Table 2. Additionally, Table 2 ranks the (pure) isotopes with respect to improving TBR. In decreasing order these are Sn-122, Sn-119, Sn-120, Sn-124 and Sn-115 [7]. These isotopes

1.20

1.15 1.10 Sn-Li alloy 1.05 -best fit ▲ Pb-17Li **2** 1.00 0.95 0.90 0.85 0.80 10% 40% 50% 60% 70% 0% 20% 30% 80% Li concentration [at%]

Fig. 1. TBR vs Li concentration in Sn–Li alloys used in the WCLL–DEMO blanket (90 at.% 6 Li enrichment).

| Table 2 | | |
|-------------------|------------|----|
| Isotone abundance | of natural | Sr |

| Isotope | Natural abundance (%) | Ranking for TBR improvement [7] |
|---------|-----------------------|---------------------------------|
| Sn-112 | 0.95 | |
| Sn-114 | 0.65 | |
| Sn-115 | 0.35 | 5 |
| Sn-116 | 14.30 | |
| Sn-117 | 7.61 | |
| Sn-118 | 24.03 | |
| Sn-119 | 8.58 | 2 |
| Sn-120 | 32.85 | 3 |
| Sn-122 | 4.72 | 1 |
| Sn-124 | 5.94 | 4 |

(which have the highest $(n,2n)/(n,\gamma)$ cross-section ratio) have small abundances with the exception of Sn-120. But even at an abundance of almost 33%, Sn-120 is a minority isotope for which isotopic tailoring is expected to be difficult and expensive.

The radial power deposition profiles in the blanket were compared between Pb–17Li and Sn–20Li [8]. It was shown that the use of Sn–20Li has two favorable consequences, namely:

- a reduction of the peak power density in the liquid breeder from approximately 32 W/cm³ (Pb–17Li) to approximately 25 W/cm³ (Sn–20Li),
- a flatter power deposition profile for Sn-20Li.

Both would require a design adaptation leading to a more homogeneous distribution of cooling tubes in the blanket, and possibly to a reduction of the required coolant flow velocities. Owing to the elevated thermal conductivity of Sn–20Li, it can be expected that much more power from the liquid metal pool will be evacuated by the first wall and side walls of the blanket segment, thus reducing the need for a very high cooling tube density right behind the first wall.

3.2. Coolant conditions

The requirement TBR > 1.05 seems to necessitate the use of a Sn–Li alloy with >33 at.% Li concentration at 90% ⁶Li enrichment. Apart from the fact that this increases the Li activity by a factor 3 (which may be acceptable when accidental Sn–33Li interactions with water can be managed), the increase in Li concentration raises the melting point of the alloy from approximately 330°C at 20 at.% to >370°C (cf. Fig. 2). Already a temperature of 330°C is basically incompatible with the use of PWR type cooling water, i.e., $T_{max} = 325°C$ at 15.5 MPa. When considering a margin of 30 K between melting point and coolant inlet temperature, the water pressure would need to be raised to at least 18.7 MPa (for Sn–20Li) to ensure that the breeder remains liquid at all times. The use of the alloy Sn–33Li, which would



Fig. 2. Phase diagram for Sn-Li alloys [4].

ensure sufficient TBR, would clearly require supercritical water conditions. This could become a problem insofar as then the entire blanket box would have to be designed for the increased coolant pressure, requiring more structural steel thus leading to a decrease in TBR. Even if this was considered acceptable from a pure thermal-hydraulic point of view, we should keep in mind that these coolant conditions would significantly raise the temperature level in the structures and notably in the first wall, where temperature limits for the steel (currently 550°C for RAFM steel) are easily exceeded. The use of oxide dispersion strengthened (ODS) steel with higher temperature limits may help to resolve this problem but requires further analysis.

4. Use of Sn-Li alloys in the TAURO blanket

The TAURO blanket is a self-cooled breeding blanket using the alloy Pb–17Li both as coolant and tritium breeder and SiC_f/SiC ceramic matrix composite as the structural material. As a coolant, Sn–Li alloys present several advantages over Pb–17Li because of the higher thermal conductivity and heat capacity. This could result in a more uniform temperature distribution in the blanket together with a reduction of the liquid metal flow velocity and MHD pressure drop. On the other hand, since the volume fraction of Pb–17Li is higher in the TAURO blanket than in the WCLL blanket, the TBR reduction is expected to be higher.

Table 3 TBR and neutron absorption for various Sn-Li alloys compared to Ph-17Li in the TAURO blanket

| Alloy | TBR | Neutron absor source neutron | Neutron absorption (per source neutron) | |
|---------|------|---------------------------------|---|--|
| Pb–17Li | 1.37 | 0.031 (Pb) | 0.137 (SiC) | |
| Sn–25Li | 0.65 | 0.157 (Sn) | 0.160 (SiC) | |
| Sn–30Li | 0.70 | 0.132 (Sn) | 0.155 (SiC) | |
| Sn–50Li | 0.80 | 0.070 (Sn) | 0.137 (SiC) | |

Table 3 reports the results of neutronic calculations performed with the code TRIPOLI 4 on a two-dimensional model of the TAURO blanket assuming 90% ⁶Li enrichment. Note that even for high atomic fractions of Li (up to 50%), tritium self sufficiency is not reached. The reduction in TBR compared to Pb–17Li is mainly due to neutron absorption in Sn which is (considering Sn–25Li) nearly five times higher than in Pb–17Li. Since tritium self-sufficiency cannot be reached, no further investigations have been made.

5. Tin-lithium alloys as divertor coolant

Recent investigations on liquid-metal forced convection cooled divertors yielded initial results [2] indicating that heat fluxes up to 6 MW/m² can be sustained by a castellated square tube concept. This type of divertor is made up of a W-alloy and employs a slotted SiC flow-channel insert as an electrical insulator to reduce MHD pressure drops. Owing to the higher specific heat and thermal conductivity (the values for pure Sn were assumed in [2]), for a given heat flux on the divertor, the substitution of Sn-20Li for Pb-17Li leads to lower liquid flow-rates (lower MHD pressure drop) and higher outlet temperatures (favorable for power conversion). However, as the temperature and stress in the W structure are limiting, the theoretical advantage of Sn-20Li over Pb-17Li cannot be fully exploited. It can be concluded that changing the coolant from Pb-17Li to Sn-20Li in this divertor concept would be advantageous only when combined with a suitable Sn-20Li-cooled blanket, thus allowing the use of a single coolant in the blanket/divertor system.

6. Afterheat and short-term activation

Afterheat and short-term activation of Sn-20Li is higher than for Pb-17Li. The primary isotopes contributing to contact dose from activated Sn are, in order of importance: Sn-117m, Sn-111, Sn-125m, In-113m and In-111. The weight-based radiological hazard of activated tin was found to be similar to that of W. However, similar to Pb-17Li, liquid Sn-Li alloys must

7. Long-term activation

Tin has three long-lived activation products:

- ^{121m}Sn, half life 55 yr,
- ^{108m}Ag, half life 418 yr,
- ¹²⁶Sn, half life 105 yr.

It was estimated [4] that the activation from Sn-126 corresponds to approximately 5% of the total long term activation of the breeder. This Sn-126 activation is by about one order of magnitude lower than the activation due to Bi-208 from Pb–17Li. Sn–Li alloys can, therefore, be considered better than Pb–17Li from a long-term activation point of view.

8. Conclusions

The favorable thermo-physical properties attributed to Sn–Li alloys have motivated us to verify whether it makes sense to use Sn–Li in current blanket and divertor concepts designed for Pb–17Li.

Clearly, the preliminary analyses on the use of Sn–Li alloys in the WCLL and TAURO blanket are unfavorable, the reasons being:

- 1. TAURO and WCLL: unacceptably low TBR,
- 2. WCLL: requirement to increase the water pressure to accommodate the higher melting points, thus causing higher demands for structural integrity and a decrease in TBR,
- 3. WCLL: unsatisfactory compatibility with RAFM steel (although this might be overcome by coatings),

4. WCLL: suspected liquid metal embrittlement.

Concerning the recently investigated forced-convection liquid metal cooled divertor, changing the coolant from Pb–17Li to Sn–20Li would be advantageous only when combined with a suitably performing Sn–20Li cooled blanket.

Since the data base for thermo-physical properties of Sn–Li alloys is still incomplete, the conclusions reached here have some caveats. In particular, the corrosion and embrittlement of RAFM steel, the compatibility of Sn–Li alloys with SiC_f/SiC structures and flow channel inserts as well as technology aspects such as purification, Li-adjustment, tritium extraction and safety would

require many years of R&D to reach a confidence level comparable to Pb–17Li. The tritium solubility and diffusivity in the breeder material must also be measured to determine the tritium inventory and permeation losses into the secondary coolant and to develop suitable countermeasures if required.

It can be concluded that Sn–Li alloys are best suitable for free surface blanket concepts owing to their low vapor pressure. Yet, it cannot be recommended to modify existing Pb–17Li blanket concepts for use with Sn–Li alloys.

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