Liquid Sn-20Li alloy as a heat transfer fluid

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Abstract. The purpose of present study was to obtain information on the solubility of components of structural materials in liquid Sn-20Li alloy in the temperature range of practical interest. To solve this problem, a thermodynamic simulation method was used, which included the following steps: (1) selection of models for the Gibbs energy functions; (2) selection and evaluation of input data; (3) optimization of model parameters; (4) calculations and comparisons. Using information on the excess Gibbs energies for the liquid phase in the form of Redlich-Kister polynomial expansions, for the corresponding binary systems, the temperature dependencies of solubility of nickel, iron, chromium, molybdenum and tungsten in the liquid Sn-20Li are calculated. The results of the calculations are in satisfactory agreement with the experimental data known from the literature.

1. Introduction

In the overall development strategy of renewable energy sources, one of the main roles is given to Concentrated Solar Power (CSP) systems. Central receiver systems (CRS) as the most promising direction of development of solar thermal power are characterized by higher operating temperatures and larger heat flux densities [1]. An important step in creating an economical CRS is the selection of an efficient heat transfer fluid (HTF). One of the potential classes of such liquids are liquid metals [2]. One of the features of liquid tin-lithium alloys is that conventional structural materials in contact with liquid Sn-Li are subject to intense corrosion, because tin is one of the most aggressive liquid metals. It is known [3, 4] that tin reacts with steels with the formation of FeSn2 and FeSn intermetallic compounds; as a result, liquid tin or liquid alloys containing Sn cannot be used as a coolant in circuits made of ferritic or austenitic steel at temperatures substantially exceeding the melting point of the liquid alloy. According to preliminary estimates [3,5], materials based on Mo, W and Re should have satisfactory corrosion resistance in liquid tin over a wider temperature range.

One of the main parameters determining the kinetics and dynamics of mass transfer in liquid metal circuit is the solubility of the components of the structural material in a low-melting metal or alloy. The lack of reliable data on the equilibrium solubility of components of structural materials in a liquid metal makes it difficult to determine the temperature range of availability of these materials.

If unsystematized data on the solubility of iron, chromium, nickel and molybdenum in liquid tin can be found in the literature, there is no such information for the solubility of the above metals, as well as tungsten in liquid Li-Sn alloys. Experimental studies that would contain reliable information on the solubility of tungsten in liquid lithium and liquid tin are also unknown, which is explained by the extremely low solubility of this metal in almost all liquid metals [6].
Among the possible candidates for use as HTFs for central receiving systems are Sn-Li alloys, in which the lithium content varies from 20 to 30 mol % [7]. In the present work, a liquid alloy with a component content of 20 mol % Li and 80 mol % Sn, for which the \( \text{Li}_{20}\text{Sn}_{80} \) designation will be used in what follows.

In the present work, using thermodynamic modeling, calculations have been made to find the solubility parameters of the components of structural materials in liquid \( \text{Li}_{20}\text{Sn}_{80} \) within a temperature range of practical interest. The following metals were considered as such components: Fe, Ni, Cr, Mo and W. The problem was reduced to consideration of phase equilibria involving the liquid phase in certain concentration ranges in the following three-component systems: Li-Sn-Fe, Li-Sn-Ni, Li-Sn-Cr, Li-Sn-Mo and Li-Sn-W.

2. Materials and methods

The state of a pure element in the solid state in its stable form at \( T_0 = 298.15 \) K was chosen as the reference state in the system under consideration. The Gibbs energy \( \delta G(T) - H^\text{SER}(T) \) for element \( i \) (\( i = \text{Li}, \text{Sn}, \text{Fe}, \text{Ni}, \text{Cr}, \text{Mo}, \text{W} \)) in phase \( \phi \) is described by the following equation

\[
\delta G(T) - H^\text{SER}(T_0) = a + bT + c \ln(T) + \sum_{n} d_n T^n,
\]

where \( T \) is the absolute temperature, \( H^\text{SER}(T_0) \) is the molar enthalpy of element \( i \) at 298.15 K and a pressure of \( 10^5 \) Pa in its SER (standard element reference) state. The SGTE database (Scientific Group Thermodata Europe) [8] was chosen as the source of thermodynamic data necessary for pure elements calculations.

Liquid metal solutions, as well as solid solutions, were considered as substitution solutions. The molar Gibbs energy of the solution (phase \( \phi \)) can be written as follows

\[
G^\phi_m - H^\text{SER} = \text{ref} \ G + \text{id} G^\phi + \text{xs} G^\phi,
\]

\[
\text{ref} \ G = \sum_i \left( \delta G_i(T) - H_{i}^{\text{SER}}(T_0) \right) \cdot x_i
\]

\[
\text{id} G^\phi = RT \sum_i x_i \ln(x_i)
\]

\[
\text{xs} G^\phi = \sum_{i \neq j} x_i x_j \left[ L_{ij}^0 (x_i - x_j) + L_{ij}^1 (x_i - x_j)^2 + ... \right],
\]

where \( i, j = \text{Li}, \text{Sn}, \text{Fe}, \text{Ni}, \text{Cr}, \text{Mo}, \text{W} \); \( \text{ref} \ G \) is the Gibbs energy of the phase \( \phi \), consisting of pure components, or, as it is also called, in a reference state; \( \text{id} G^\phi \) is the mixing energy in the case of ideal solutions; \( \text{xs} G^\phi \) is the excess Gibbs energy of mixing, which is presented in the form of the Redlich–Kister polynomial equation [9]; \( L_{ij}^0, L_{ij}^1, L_{ij}^2 \) are the pair interaction parameters for a given binary system, which are also functions of temperature; \( x_i \) and \( x_j \) are the molar fractions of components of system. The methods of critical estimation and optimization of thermodynamic parameters for binary systems are described in detail in the monograph [10].

If only terms with \( L_{ij}^0 \) and \( L_{ij}^1 \) coefficients are left in the polynomial expansion Eq. (2c) and at the same time the temperature dependence of these coefficients is preserved, then such a solution model is called pseudo-subregular model [11]. In this case, Eq. (2c) is rewritten as follows:
\[ x_i \varphi = \sum_{i \neq j} x_i x_j \left[ \Omega_{ij}^i (T) \cdot x_i + \Omega_{ij}^j (T) \cdot x_j \right], \]  

(3)

where \( \Omega_{ij}^i (T) = (\Omega_{ij}^i + \Omega_{ij}^j \cdot T) \) and \( \Omega_{ij}^j (T) = (\Omega_{ij}^i + \Omega_{ij}^j \cdot T) \) are the pair interaction parameters for the binary system \( A_i-A_j \).

**Table 1.** Pair interaction parameters for the liquid phase in binary systems.

| System   | Pair interaction parameter (J / mol) | Concentration range | References |
|----------|-------------------------------------|---------------------|------------|
| Li-Sn    | \( \varphi_{LiSn}^i (T) = -1.4252 \times 10^5 + 22.0141 \cdot T \) | \( 0 \leq x_{Li} \leq 0.5 \) | [12]       |
|          | \( \varphi_{LiSn}^j (T) = -4.936 \times 10^4 + 0.28945 \cdot T \) |                       |            |
| Li-Fe    | \( \varphi_{LiFe}^i = \Omega_{LiFe}^i = 58059.7 + 44.950 \cdot T \) | \( 0 \leq x_{Fe} \leq 0.1 \) | [13]       |
|          | \( \varphi_{SnFe}^j = 28659 - 1.7881 \cdot T \) | \( 0 \leq x_{Fe} \leq 0.25 \) | [14]       |
| Sn-Fe    | \( \varphi_{SnFe}^i = 20054 - 1.7881 \cdot T \) |                       |            |
| Li-Ni    | \( \varphi_{LiNi}^i = \Omega_{LiNi}^i = 60732.8 - 0.8386 \cdot T \) | \( 0 \leq x_{Ni} \leq 0.1 \) | [15]       |
| Sn-Ni    | \( \varphi_{SnNi}^i = -87255 + 280.03 \cdot T - 31.934 \cdot T \cdot \ln(T) \) | \( 0 \leq x_{Ni} \leq 0.5 \) | [14]       |
|          | \( \varphi_{SnNi}^j = -205955 + 528.66 \cdot T - 63.8687 \cdot T \cdot \ln(T) \) |                       |            |
| Li-Cr    | \( \varphi_{LiCr}^i = \Omega_{LiCr}^i = 170966.6 - 52.341 \cdot T \) | \( 0 \leq x_{Cr} \leq 0.1 \) | [15]       |
| Sn-Cr    | \( \varphi_{SnCr}^i = 37105 + 8.566 \cdot T \) | \( 0 \leq x_{Cr} \leq 0.5 \) | [14]       |
|          | \( \varphi_{SnCr}^j = 21257 + 1.196 \cdot T \) |                       |            |
| Li-Mo*   | \( \varphi_{LiMo}^i = \Omega_{LiMo}^i = 2.5085 \cdot 10^5 - 55.4900 \cdot T \) | \( 0 \leq x_{Mo} \leq 0.1 \) | [16]       |
| Sn-Mo    | \( \varphi_{SnMo}^i = \Omega_{SnMo}^i = 69000 + 11.72 \cdot T \) | \( 0 \leq x_{Mo} \leq 0.01 \) | [17]       |
| Li-W*    | \( \varphi_{LiW}^i = \Omega_{LiW}^i = 219001.7 - 61.187 \cdot T \) | \( 0 \leq x_{W} \leq 0.1 \) | [16]       |
| Sn-W*    | \( \varphi_{SnW}^i = \Omega_{SnW}^i = 244970.5 - 62.367 \cdot T \) | \( 0 \leq x_{W} \leq 0.1 \) | [16]       |

* Theoretical assessment using the model of A. Miedema [16].

Some advantage of Eq. (3) is that its use provides the possibility of obtaining analytical dependences of the coefficients of the thermodynamic activity coefficients of the metal solution components as a function of temperature and composition in the case of a three-component system \( (i,j=1,2,3; i \neq j) \). In particular, for the coefficient of thermodynamic activity of the third component A3 in the Li-Sn-A3 system, the following relation is valid:

\[ R T \ln \gamma_3 = x_1 Q_{31} + x_2 Q_{32} - \Delta H_{mix}, \]

(4)

where

\[ \Delta H_{mix} = \sum_{i \neq j} \left[ x_i x_j \left[ \sum_{i \neq j} \left( \frac{x_j}{x_i + x_j} \right) \right] \right], \]

(4a)
\[ Q_{31} = \Omega_{43}^{1} (T) \cdot \left( \frac{x_1}{x_1 + x_3} \right) + \Omega_{43}^{3} (T) \cdot \left( \frac{x_3}{x_1 + x_3} \right) + \left( \frac{x_3}{x_1 + x_3} \right) \cdot \left[ \Omega_{13}^{3} (T) - \Omega_{13}^{1} (T) \right], \quad (4b) \]

\[ Q_{32} = \Omega_{23}^{2} (T) \cdot \left( \frac{x_2}{x_2 + x_3} \right) + \Omega_{23}^{3} (T) \cdot \left( \frac{x_3}{x_2 + x_3} \right) + \left( \frac{x_3}{x_2 + x_3} \right) \cdot \left[ \Omega_{23}^{3} (T) - \Omega_{23}^{2} (T) \right]. \quad (4c) \]

In our case, \( A_3 = \text{Fe, Ni, Cr, Mo, and W}. \)

Since there are no experimental data in the literature on the thermodynamic activity coefficients of elements such as Li, Sn, Fe, Ni, Cr, Mo and W in liquid metal alloys Li-Sn-Fe, Li-Sn-Ni, Li-Sn-Cr, Li-Sn-Mo and Li-Sn-W, to find the necessary thermodynamic parameters for the above three-component systems, the available experimental data for the corresponding binary systems were used.

The values of the pair interaction parameters for the liquid phase in binary systems, which were used to calculate according to the equations of the pseudo-subregular model, are given in Table 1.

The temperature dependences of pair interaction parameters are the result of applying the regression model equations to an array of experimental data related to a specific binary system, including information obtained from the corresponding phase diagrams [6]. The optimization method developed by the authors of [18] was used to obtain polynomial expressions for the excess Gibbs energy as a function of composition and temperature for all binary systems considered here.

An analysis of the diagrams of Fe – Sn, Ni – Sn, Cr – Sn, Mo – Sn, and Sn – W binary systems shows that only the last two of these systems are characterized by the absence of solubility regions of a more refractory metal in liquid tin. The Sn-W system belongs to the class of systems with almost complete absence of interaction between the components in the solid and liquid state [19]. This especially distinguishes this system from other binary systems considered here, in which, in addition to the regions of mutual solubility, the formation of intermediate phases is also recorded [6].

Also, the temperature dependences of the metal solubility in liquid lithium and liquid tin obtained from analysis of experimental data were used as the initial data for calculations in the present study (Table 2).

### Table 2. Temperature dependence of the solubility of metals (mole fractions) in liquid lithium and liquid tin at 673–1173 K.

| Metal | Solvent | References |
|-------|---------|------------|
| Fe    | Li      | \( x_{\text{Fe}(\text{Li})}^{\text{sat}} = \exp(5.409 - 6.987 \cdot 10^3 / T) \) | [13, 20] |
|       | Sn      | \( x_{\text{Fe}(\text{Sn})}^{\text{sat}} = \exp(3.974 - 8.001 \cdot 10^3 / T) \) | |
| Cr    | Li      | \( x_{\text{Cr}(\text{Li})}^{\text{sat}} = \exp(6.299 - 20.57 \cdot 10^3 / T) \) | [15, 21] |
|       | Sn      | \( x_{\text{Cr}(\text{Sn})}^{\text{sat}} = \exp(4.053 - 8.474 \cdot 10^3 / T) \) | |
| Ni    | Li      | \( x_{\text{Ni}(\text{Li})}^{\text{sat}} = \exp(0.101 - 7.308 \cdot 10^3 / T) \) | [15, 22] |
|       | Sn      | \( x_{\text{Ni}(\text{Sn})}^{\text{sat}} = \exp(2.335 - 5.523 \cdot 10^3 / T) \) | |
| Mo    | Li      | \( x_{\text{Mo}(\text{Li})}^{\text{sat}} = \exp(6.678 - 30.19 \cdot 10^3 / T) \) | [16]*, [17] |
|       | Sn      | \( x_{\text{Mo}(\text{Sn})}^{\text{sat}} = \exp(1.203 - 12.89 \cdot 10^3 / T) \) | |
| W     | Li      | \( x_{\text{W}(\text{Li})}^{\text{sat}} = \exp(7.363 - 26.35 \cdot 10^3 / T) \) | [16]* |
|       | Sn      | \( x_{\text{W}(\text{Sn})}^{\text{sat}} = \exp(7.505 - 29.48 \cdot 10^3 / T) \) | |

* Theoretical assessment using the model of A. Miedema [16].

### 3. Results

As a result of thermodynamic modeling, the obtained dependences of the limiting solubilities of Fe, Ni, Cr, Mo and W in liquid Li\(_{20}\)Sn\(_{80}\) in the temperature range 623–1173 K are given below:
where $x_{A}^\text{sat}$ values are expressed in molar fractions.

As can be seen (Figs. 1 and 2), of the five transition metals discussed here, nickel has the highest solubility in the temperature range of 673 to 1173 K. The results of thermodynamic modeling show that of the metals considered, only Mo and W are characterized by acceptable solubility levels in $\text{Li}_{20}\text{Sn}_{80}$ at temperatures above 600 °C.

![Graph showing solubilities of metals in liquid tin and liquid Sn-20Li alloy plotted as function of the reciprocal temperature.](image)

**Figure 1.** Solubilities of metals in liquid tin and liquid Sn-20Li alloy plotted as function of the reciprocal temperature.
Figure 2. Solubilities of molybdenum and tungsten in liquid tin and in liquid Sn-20Li alloy plotted as a function of the reciprocal temperature.

4. Discussion

It is possible to evaluate how the deviation from the ideal behavior of the liquid phase in any of the three-component systems affected the solubility of the structural materials component in the liquid metal solution. For example, let’s turn to the Li-Sn-Fe system and select as such a dilute solution of iron in the Li-Sn liquid phase, in which the mole fractions of the components satisfy the ratio $x_{Li} : x_{Sn} = 20 : 80$. Assuming that the lithium in the tin solvent is an ideal solution, according to [23], the solubility of Fe in the liquid phase can be described by the following equation:

$$\ln x_{Fe(Li-Sn)}^{sat} = x_{Li} \ln x_{Fe(Li)}^{sat} + x_{Sn} \ln x_{Fe(Sn)}^{sat},$$  (10)

where $x_{Li}=0.2$ and $x_{Sn}=0.8$ are molar fractions of lithium and tin in binary liquid Li-Sn alloy.

As shown in Fig. 1, the calculation by Eq. (10) results in overestimated iron solubility values in the $Li_{20}Sn_{80}$ alloy compared to the data obtained from the rigorous thermodynamic simulation (Eq. (5)). This seems understandable when considering that liquid alloys of Li-Sn system belong to the class of melts with strong chemical interaction of components, which are characterized by significant negative deviations from ideal behavior [12]. It should be noted that the solubility of Fe in $Li_{20}Sn_{80}$ even at 1000 °C does not exceed 2 mol %.
Considering the three-component Li-Sn-Fe solution from the position of the surrounded atom model [24-26], the above-noted feature of the Li-Sn system \( \gamma_{Li}[1] \) clearly indicates that there is a tendency for short-range ordering\(^1\) in the liquid metal solution when lithium atoms are located predominantly in the first coordination sphere around the Sn atom. This should be expressed in a decrease in the fraction of atomic configurations in which Fe atoms are located in the first coordination sphere around the Sn atom. Thus, in a real Li-Sn-Fe metal solution, \( \chi_{Fe(Li-Sn)}^{sat} \) decreases compared to the level of solubility that could have place, if the binary Sn-Li liquid solution could be considered as an ideal in its properties.

Current knowledge about the compatibility of materials with Sn-Li alloys cannot be considered satisfactory due to the lack of experimental data. The high solubility of Fe in the liquid \( \text{Li}_{20}\text{Sn}_{80} \) alloy is reported by the authors of publication [28], in which the corrosion of ferritic-martensitic steel JLF-1 (Fe-9Cr-2W-0.1C) at 600 °C was studied. The data of the above-mentioned work are in good agreement with the numerical estimates of this work, from which it follows that in such a parameter as the solubility of the main components of structural materials, the liquid \( \text{Li}_{20}\text{Sn}_{80} \) alloy is close to liquid tin.

### 5. Conclusion

Based on the Gibbs energy data of the components in various phases of the binary systems, as well as using the thermodynamic database for pure elements, the temperature dependences of the solubility of nickel, iron, chromium, molybdenum, and tungsten in the Sn-20Li liquid alloy were calculated by thermodynamic modeling. Among the transition metals considered, only Mo and W are characterized by acceptable solubility levels in \( \text{Li}_{20}\text{Sn}_{80} \) at temperatures above 800°C. The results of the calculations show reasonable agreement with the data of corrosion tests of ferritic-martensitic steel JLF-1 (Fe-9Cr-2W-0.1C) in a liquid Sn-20Li alloy known from literature.

Thus, present work allows us to significantly limit the range of materials that can be used as containment materials for liquid tin-lithium alloy as a heat transfer fluid for high temperature concentrated solar installations.

Since, in addition to the low solubility in HTF, the material must also be resistant to other corrosion factors, many types of liquid metal corrosion tests must be conducted to determine the most satisfactory container material for a particular set of environmental conditions. In this context, oxide ceramic materials are also considered promising in addition to metal materials.

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\(^1\) In the liquid solution atoms are in constant motion, so it makes sense to talk about the average for some time \( t \) the configuration of atoms. For such configurations, a reasonable estimate of the averaging time falls into the range \( \tau_0 < t < \tau_D \) ( \( \tau_0 \) is a period of oscillations in the equilibrium position; \( \tau_D \) is time of the “settled life” of an atom). According to [27], the following estimates are used in this paper: \( \tau_0 \approx 10^{-13} \text{ s} \) and \( \tau_D \approx 10^{-11} \text{ s} \).
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