

Interfacial Phenomena between Liquid Ga-Based Alloys and Ni Substrate

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In this study, wetting tests for Ni substrate with eutectic Ga-Sn-Zn are carried out using the sessile drop method. The experiments are performed for 1, 10 and 30 days of contact, at temperatures of 100°C, 150°C and 250°C. Selected liquid/substrate couples are cross-sectioned and subjected to scanning electron microscopy with energy dispersive spectroscopy for interfacial microstructure investigation. Phase identification is carried out using synchrotron x-ray diffraction. The growth of the intermetallic Ni-Ga phase layer is studied at the liquid/Ni substrate interface, and the kinetics of the formation and growth of this layer are determined.

Key words: Ni substrate, intermetallic, liquid metal, microstructure, kinetic, wettability

INTRODUCTION

The miniaturisation of electronics which generate a large amount of heat calls for more efficient cooling systems.¹ The potential to use print layouts below 250 μ m^{2,3} means that liquid metals can find applications in soft sensors.⁴ Furthermore, the applications of soft lithography and microfluidics in biology, analytical biochemistry and chemistry have grown as a range of new components and techniques have been developed and implemented for introducing, mixing, pumping, and storing fluids in microfluidic channels.⁵ There is a wide range of potential applications of liquid metals. For example, their use in liquid coolant management systems allows the development of industrial applications using electromagnetic pumps,^{6,7} such as in renew-able energy sources,⁸ electronic devices,⁹ Li-ion and the Liquid Metal Fast Breeder batteries,¹ Reactor.¹¹ Taking into account their thermal conductivity, the proposed liquid metals have attractive characteristics as heat transfer fluids for high power

density devices, which allows increased efficiency and performance of cooling systems.¹² Moreover, the very high bonding temperature of Ga alloys creates the possibility of application in nuclear reactors as a coolant.¹³ The liquid metal most commonly used as a primary coolant in nuclear reactors is sodium, but this has the disadvantage of reacting violently with water and air.¹⁴ With a view to avoiding these dangers, alternative coolant systems such as heavy liquid metals (including pure lead and lead-bismuth eutectic (LBE) alloy) have been extensively studied for their potential application in fast reactors and as spallation targets.^{14,15} However, the reactivity of Ga allovs at higher temperatures causes relatively high corrosion of the structural metals and alloys, due to the dissolution of their various constituents by the liquid gallium.¹⁶ The even application of austenitic stainless steel (AISI 316L) at higher temperature caused the formation of a reaction layer and suggested that improved corrosion resistance with a protective layer such as Al_2O_3 and SiO_2 must be pursued.¹³

The most promising potential application of liquid gallium alloys is in electronic devices. However, liquid Ga alloys react and dissolve the Cu substrate.^{17,18} To reduce the rate of the dissolving

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reaction and minimise the particle internalisation process,¹⁹ the Cu substrate can be coated with Ni.²⁰ This interlayer between liquid metals and Cu substrate causes a reduction in the diffusion of Ga to the Cu substrate by the formation at the interlayer of intermetallic compounds from the Ga-Ni system,²¹ which reduces diffusion of Cu to Ga. The aim of this study is to demonstrate the effect of time and temperature on the kinetics of the formation and growth of the intermetallic layer at the interface of liquid eutectic Ga-Sn-Zn and Ni substrate.

EXPERIMENTAL

Wetting tests²² were performed for eutectic Ga-Sn-Zn alloys²³ 86.3, 90.15 of Ga, 6.64 of Sn and 3.21 of Zn (at.%) corresponding to 86.3, 10.8 and 2.9 (wt.%), on Ni substrate (purity 99.99% and 250 μ m thickness) using the sessile drop method. For this experiment, 15×20 mm sheets of Ni were used, with a drop 2-3 mm in diameter on top of the alloy. Taking into account the possible applications of these materials as cooling liquids in electronics, tests were conducted for three time periods (24 h, 240 h and 720 h) and at three temperatures (100°C, 150°C and 250°C). The obtained samples of cross sections were investigated using scanning electron microscopy (SEM) with energy dispersive spectroscopy (EDS), to observe the microstructure and chemical composition. Phase identification was done by diffraction of high-energy synchrotron radiation ($\lambda = 0.153495$ A) in transmission geometry using the HZG materials science beamline P07B at DESY in Hamburg, Germany. Measurements in transmission geometry yielded diffraction information from representative large sample volumes (beam size $0.7 \times 0.7 \times 0.3$). To ensure better statistical accuracy, the samples were continuously rotated 180° around the ω -axis.^{24,25} Such an approach allows powder-type diffraction to be obtained, excluding the texture effect. The resulting 2D patterns were integrated using the Fit2D software. Rietveld refinement was used to calculate the volume fraction of particular phases, employing the FullProf Suite software. The microstructure spreading areas were calculated using the GetArea application in CorelDraw12. The measurement error was below 2%, so error bars was not marked in the figures. Thermo-Calc was used to calculate the Gibbs free energy of the Ni-Ga and Ni-Zn phases. As the Gibbs free energy of Ni-Sn phases is much lower, and in order to provide a better view of the difference between obtained data, the results for Ni-Sn phases are not presented.

RESULTS AND DISCUSSION

Effect of Time and Temperature on the Microstructure at the Interface after Wetting Tests

Wetting tests using eutectic Ga-Sn-Zn on Ni substrate were carried out. Ni was applied in order to reduce the dissolution of Cu substrates by Ga alloys. The microstructure of the cross section of liquid eutectic Ga-Sn-Zn on Ni substrate after 24 h, 240 h and 720 h at a temperature of 100°C is presented in Fig. 1. After 24 h, the product of reactions between liquid eutectic Ga-Sn-Zn and the Ni substrate, containing higher amounts of Ga, Ni and Zn (Fig. 1a), were formed at the interface. On increasing the time to 240 h, intermetallic layers of Ga₅Ni and very thin Ga₇Ni₃ (according to the Ga-Ni phase diagram²⁶) occurred. However, a difference was observed between the obtained Ga₅Ni and Ga₇Ni₃ phases and the greater thickness of the Ni substrate after 720 h, as shown in Fig. 1c.

Similar microstructure was observed at a temperature of 150°C after 24 h. When the wetting time was increased to 240 h, the Ga₇Ni₃ phase was identified at the interface. After 720 h, the Ga₇Ni₃ and Ga₃Ni₂ phases were observed. Microstructure after 720 h at a temperature of 150°C is presented in Fig. 2, which shows an undulating interface related to the nature of the reaction between the liquid eutectic Ga-Sn-Zn and the Ni substrate. The observed microstructure of the Ga7Ni3 phase is divided by an area of higher Sn content, as shown in EDS analysis in Table I. The main difference between the microstructure obtained at 100°C and 150°C after the wetting process is the formation at the interface, from the Ni substrate, of a very thin Ga₃Ni₂ phase layer.

The greatest changes in microstructure were observed at 250°C, as presented in Fig. 3. After 24 h of aging (Fig. 3a), a thick, continuous layer with the same chemical composition as that of the layer obtained after 720 h at 150°C, (Table I, points 12 and 13), and a very thin Ga_3Ni_2 phase layer, were observed at the interface with the Ni substrate. With increasing time, the thickness of both intermetallic (IMC) layers also increased. After 240 h (Fig. 3b), the Ga_3Ni_2 phase is clearly distinguishable, and this effect persists until 720 h (Fig. 3c). The Ga₇Ni₃ phase has a column structure, which grows with increasing time. The extensive diffusion paths were available in the initial Ni₃Ga₇ layers with loose and columnar structure (Fig. 3d) for molten Ga, which caused a reaction and the formation of the Ga₃Ni₂ phase. This suggests that Ga is the dominant diffusion species in the Ni_3Ga_7 phase, as the newly formed Ni-rich phase formed on the Ni-rich side, instead of on the Ga-rich side. A similar observation was noted in¹⁶ for liquid Ga on Ni substrate at 300°C, where the Ni₃Ga₇ phase was identified at the interface. Over time, this phase transformed to a Ga₃Ni₂ phase, which is connected with the Gibbs free energy of phases and the Ga-Ni phase diagram.²

Kinetic Calculations of IMC Layers

In Fig. 4, the thermodynamic calculation of Gibbs free energy for the Ga-Ni system indicates Ni₂Ga₃



Fig. 1. The microstructure after wetting of eutectic Ga-Sn-Zn alloy on Ni substrate at a temperature of 100°C after (a) 24 h, (b) 240 h and (c) 720 h.



Fig. 2. The microstructure after wetting of eutectic Ga-Sn-Zn alloy on Ni substrate at a temperature of 150°C and 720 h.

as the most stable phase.²⁸ According to,²⁷ there is no information about Ga₇Ni₃, which would allow Gibbs free energy to be calculated. While wetting the Ni substrate with eutectic Ga-Sn-Zn alloy, it became possible that a phase from the Ni-Zn system 22 would occur. Moreover, the Gibbs free energy is much higher compared to the Ga-Ni phase, confirmed by the absence of Ni-Zn precipitates from the microstructure. The phase from the Ni-Zn system was observed in the case of soldering Ni substrate with Sn-Zn alloys,²⁹ and with increasing soldering time. The IMC from the Ni-Zn system has the lowest Gibbs free energy, and the start of the formation of an IMC from the Ni-Sn system is connected to the amount of Zn present. In this study, under different conditions, the most stable IMC is from Ga-Ni, in which Ga is the dominate element in liquid state. However, as shown by EDS analysis, the Zn is dissolved in the area of the IMC layers forming from the Ga-Ni system. As shown in Fig. 1a and b, EDS analysis (points 2, 3 and 4, 5) demonstrates the start of Ni substrate dissolution at 100°C after 24 h, and after 240 h the Zn assists the Ga in interaction with the Ni substrate. This situation is correlated with the low diffusion of Ga in Ni at 100°C and is not observed at higher temperatures of 150°C or 250°C. The IMCs occurring from the Ga-Ni system corresponding with annealing conditions were confirmed by taking synchrotron measurements.

Points	$\mathbf{Sn}^{\mathbf{L}}$	Ni ^K	Zn ^K	Ga ^K
1	7.8	7.4	1.2	83.6
2	0.8	9.3	31.5	57.8
3	0.8	10.7	29.7	58.7
4	9.6	6.2	3.1	81.1
5	0.5	9.2	34.1	56.1
6	1	10.6	33.9	54.5
7	0.9	27.8	5.8	65.4
8		16.5	3.3	80.2
9		27.7		72.3
10	7.8	7.9		84.3
11		28.8	1.1	70
12		29.9		70.1
13		29.2		70.8
14		30		70
15		40.9		59.1
16		29.6		70.4
17		40.1		59.9

Table I. The EDS analysis at the points marked in

The temperature and time dependence of the growth of IMC layers at the interface of liquid eutectic Ga-Sn-Zn on Ni substrate are presented in Fig. 5. The observed microstructure of the cross section (Figs. 1 and 2) shows the greatest thickness of the IMC layers at a temperature of 250° C and 720 h. The thickness of the IMC layer (d) is dependent on the growth rate (k) and growth time (*t*) versus exponential factor $(n), d = k(t)^n$. The character and rate of growth are correlated with parameter n as a coefficient of growth, where n < -0.5 is the grain boundary, n = 0.5 is volume diffusion and n = 1 is the chemical reaction. In this study, the parameter k is 1.30; 7.76; 23.77 $(10^{-6} \text{ m}^2 \text{ h}^{-1})$ and n is 0.55, 0.38 and 0.35 for 100°C, 150°C and 250°C, respectively. The character of growth from volume diffusion at 100°C is changed to grain boundary at 150°C and 250°C. This correlates with the phase diagram,²⁷ which shows that, below 100°C, one phase (Ga₅Ni) occurs, and above this temperature Ga₇Ni₃ causes faster dissolution of the Ni substrate (Fig. 5). Diffusion in the solid is the slowest process among the three steps of the liquid metal corrosion, which can result in a selective IMC corrosion layer



Fig. 3. The microstructure after wetting of eutectic Ga-Sn-Zn alloy on Ni substrate at a temperature of 250°C after (a) 24 h, (b) 240 h and 720 h.



Fig. 4. The calculated Gibbs free energy of the Ni-Ga and Ni-Zn phases using data from. $^{\rm 26,27}$

forming at the surface in contact with the liquid because of the different diffusion coefficients of the Ni substrate constituents in the Ga alloys. This selective diffusion changes the Ni substrate composition in the surface area, which can lead to the formation of IMC layers that create a barrier for the dissolution of the Ni substrate. If the diffusion rate in the solid is very low compared to that at the surface, the diffusion layer grows towards the liquid, not the Ni substrate, as with Cu substrate.¹⁹ In,¹³ the model fitting of the weight loss of nickel in pure gallium, Ga-14Sn-6Zn and Ga-8Sn-6Zn alloys, shows the greatest weight loss compared to Ga–8Sn–6Zn. After 500 h, the weight loss, $D_{\rm wNi},$ is 0.00363 $(g.cm^{-2})$ in pure gallium, 0.00125 $(g.cm^{-2})$ in Ga-14Sn-6Zn and 0.0024 (g.cm⁻²) in Ga-8Sn-6Zn, respectively. However, the IMC layers formed at the interface of the Ni substrate blocked the dissolution of the Ni substrate compared to Cu substrate. During experimentation under the same conditions, the Cu substrate was completely dissolved after 720 h at 250°C. In this study, the opposite was observed; after the formation of IMC layers at the interface, the thickness was 232.9 (μm) , with dissolution of Ni to the level of 35.3 (μm) after 250 h at 250°C. The estimated diffusion coefficients of Ni in liquid gallium at 500°C assume 24.86 $(10^{-5}~{\rm cm^2 s^{-1}})$ compared to in liquid Sn and Zn, which is 11.38 and $3.32 (10^{-5} \text{ cm}^2 \text{s}^{-1})$, respectively.¹³ For the experiment performed at 500°C, it was found that all the diffusivities of metallic species are higher in liquid gallium than in liquid tin and/or zinc when dissolved metallic species diffuse out into the liquid bath.¹³ Therefore, it is generally expected that the diffusivities of metallic species are higher in liquid gallium than in liquid gallium alloys, resulting in greater metal loss and a greater reaction layer thickness in liquid gallium



Fig. 5. The thickness of the IMCs and Ni substrate during wetting test of eutectic Ga-Sn-Zn at temperatures of 100° C, 150° C and 250° C, after 24 h, 240 h and 720 h.

than in liquid gallium alloys.¹³ The growth rate for interface migration is described by an Arrheniustype equation: $k = k_0 \exp\left(-\frac{Q}{RT}\right)$, where: k_0 , Q, R and T represent the migration rate constant, the activation energy, the universal gas constant, and the absolute temperature, respectively. The growth kinetics of IMC layers versus temperature for eutectic Ga-Sn-Zn on Ni and Cu¹⁹ substrates are presented in Fig. 6. The activation energy for Ni substrate is 32.3 (kJ.mol⁻¹), compared to 16.9 (kJ.mol⁻¹) for Cu substrate.¹⁹ Considering pure Ga with Cu substrate, the activation energy can be estimated at 23.8 (kJ mol⁻¹).¹⁷

The Identity and Confirmation of IMC Occurring at the Interface

With regard to the phase diagram of the Ga-Ni system, most studies^{30–34} show only the occurrence of Ga₃Ni₂ and Ga₄Ni, and do not take into account the presence of Ga-richest phases. However, the calorimetry study in³⁵ shows the possible occurrence of a Ga₇Ni₃ phase, as was also presented in Ref. 27 The structure of the Ga₇Ni₃ phase can be classified as Zintl phases, distinctive because of the occurrence of covalently bonded polyanionic substructures formed by the E atoms.³⁶ The structural determination of the compound Ga₇Ni₃ according to analysis of the bonding motifs is Ga compounds in their respective Ga-Ni systems, associated with the Ir₃Ge₇ structure type.³⁶ The Ga-Ni phase diagram, ^{27,37} considering the occurrence the Ga₇Ni₃ (PDF 01-073-4714) and Ga₅Ni (PDF 04-017-2870) phases, agrees with the synchrotron radiation x-ray diffraction (SRXRD) results obtained in this study,



Fig. 6. Growth kinetics of IMC layers versus temperature for eutectic Ga-Sn-Zn on Ni and Cu¹⁹ substrates.

as presented in Fig. 7, using structure to identify phases from.^{27,38} At the low wetting temperature of 100°C for liquid eutectic Ga-Sn-Zn alloy with Ni substrate, the IMC Ga₅Ni occurred at the interface during the chemical reaction, as shown in Fig. 1 and confirmed by chemical EDS analysis. With increasing wetting time, Ga₇Ni₃ also began to be formed at the interface, as confirmed by SRXRD results (Fig. 7a and Fig. 1c), which is correlated with the higher stability of this phase (see Fig. 4). Chemical analysis (Table I) also shows a greater amount of Zn at the beginning, which could be connected with the substitution of Ni atoms for Zn, as was observed in the γ -brass structure in the case of Cu and Al.² With increasing temperature, up to 250°C, the main IMC layer at the interface after 24 h is the Ga₇Ni₃ phase, as shown in the microstructure (Fig. 3a) and confirmed by SRXRD (Fig. 7b). It is shown that increasing time caused the occurrence of the Ga₃Ni₂ phase (PDF 04-001-3663), characterised by the lowest Gibbs free energy (Fig. 4), as confirmed by SRXRD measurements. The volume fraction of two main phases, i.e. Ni and Ga₇Ni₃ for 240 h and 720 h at 250°C was calculated using Rietveld refinement and SRXRD. Quantitative phase analysis showed an increase of Ga7Ni3 to 24.6% after 240 h of annealing (Ni 75.4%). Further annealing increases the volume fraction of Ga₇Ni₃ to 67.1%, resulting in lower Ni content (32.9%) across the entire sample volume. The cross sections of the Ni substrate samples are presented in Fig. 7c, with higher magnification results shown in Fig. 7b. Moreover, as shown by the SRXRD results (Fig. 7c) for wetting at 250°C after 720 h (microstructure Fig. 3c and d), a trace amount of the Ga_5Ni phase



Fig. 7. The SRXRD analysis of samples after wetting tests (a) at 100°C after 720 h, (b) at 250°C and after 24 h and 720 h, (c) higher magnification at 250°C after 720 h.

is also identified. As the volume fraction of this phase was below 1%, it was not considered for volume fraction analysis.

CONCLUSIONS

Taking into account the possible applications of Ga-based liquid metals in the electronics industry, wetting tests were carried out to show the interfacial phenomena between eutectic Ga-Sn-Zn alloy and Ni substrate. The IMC layer from the Ga-Ni system formed at the interface protected against dissolution of the Ni substrate. At the lower temperature, a Ga₅Ni layer was created, and with increasing time and temperature the Ga₇Ni₃ and Ga₃Ni₂ layers occurred. The mechanism of dissolution is controlled by volume diffusion and causes grain boundary dissolution with increasing temperature lowering parameter k. However, in regard to wetting with eutectic Ga-Sn-Zn, the IMC layer

formed at the interface in the Ni substrate is, compared to the Cu substrate, sufficient to block the dissolution of the substrate. Therefore, the thicknesses of the created IMC layers are higher compared to thickness of dissolution of the Ni substrate. This situation is connected to the creation of the Ga₇Ni₃ phase at the interface, which is the phase richest in Ga in the Ga-Ni system and has a growth rate that increases with temperature. The obtained microstructure of the Ga_7Ni_3 layer is characterised by column growth preceded by a Ga₃Ni₂ layer from the Ni substrate. According to thermodynamic calculation, the Ga₃Ni₂ phase has the lowest Gibbs free energy, followed by the Ga₇Ni₃ phase. The applied Ni layer on the substrate could be sufficient to block dissolution in electronics, up to 250°C. However, further experiments to study dynamic flow are needed in order to confirm the possibility of using Ni substrate with the liquid Ga-Sn-Zn alloy in cooling systems.

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REFERENCES

- 1. G. Bo, L. Ren, X. Xu, Y. Du, and S. Dou, Adv. Phys. X 3, 1446359 (2018).
- J. Yang, T. Zhou, L. Zhang, D. Zhu, S. Handschuh-Wang, Z. Liu, T. Kong, Y. Liu, J. Zhang, and X. Zhou, J. Mater. Chem. C 5, 6790–6797 (2017).
- A.D. Valentine, T.A. Busbee, J.W. Boley, J.R. Raney, A. Chortos, A. Kotikian, J.D. Berrigan, M.F. Durstock, and J.A. Lewis, Adv. Mater. 29, 1703817 (2017).
- 4. M.D. Dickey, Adv. Mater. 29, 1606425 (2017).
- D.B. Weibel and G.M. Whitesides, Curr. Opin. Chem. Biol. 10, 584–591 (2006).
- X. Zhou, M. Gao, and L. Gui, *Micromach.-Basel* 8, 365 (2017).
- K.A. Polzin, Liquid-Metal Pump Technologies for Nuclear Surface Power, NASA/TM—2007–21485, Access the NASA STI program home page at: http://www.sti.nasa.gov.
- N. Lorenzin and A. Abanades, Int. J. Hydrogen Energ. 41, 6990–6995 (2016).
- Y.L. Han, H. Liu, C. Ouyang, T.J. Lu, and F. Xu, Sci. Rep. 5, 11488 (2015). https://doi.org/10.1038/srep11488.
- X.-H. Yang, S.-C. Tan, and J. Liu, *Energ. Convers. Manag.* 117, 577–585 (2016).
- T. Sawada, A. Netchaev, H. Ninokata, and H. Endo, Prog. Nucl. Energy. 37, 313–319 (2000).
- A. Miner and U. Ghoshal, Appl. Phys. Lett. 85, 506–508 (2004).
- S.H. Shin, J.J. Kim, J.A. Jung, K.J. Choi, I.C. Bang, and J.H. Kim, J. Nucl. Mater. 422, 92–102 (2012).

- G. Wua, M. Jin, J. Chen, Y. Bai, and Y. Wu, Ann. Nucl. Energy 77, 310–317 (2015).
- 15. F. Barbier and J. Blanc, J. Mater. Res. 14, 737-744 (1999).
- S.-K. Lin, C.-Y. Yeh, and M.-J. Wang, *Mater. Character* 137, 14–23 (2018).
- S.-K. Lin, C.-L. Cho, and H.-M. Chang, J. Electron. Mater. 43, 204–221 (2014).
- J. Tang, X. Zhao, J. Li, Y. Zhou, and J. Liu, Adv. Sci. (Weinh) 4, 1700024 (2017).
- T. Gancarz and K. Berent, Mater. Letter. 227, 116–119 (2018). https://doi.org/10.1016/j.matlet.2018.05.053.
- Z. Wang, L. Fang, I. Cotton, and R. Freer, *Mater. Sci. Eng.* B-Adv. 198, 86–94 (2015).
- W.X. Yuan, Z.Y. Qiao, H. Ipser, and G. Eriksson, J. Phase Equilib. Diffus. 25, 68–74 (2004).
- T. Gancarz, P. Bobrowski, S. Pawlak, N. Schell, R. Chulist, and K. Janik, J. Electron. Mater. 47, 49–60 (2018).
- A. Dobosz, Y. Plevachuk, V. Sklyarchuk, B. Sokoliuk, and T. Gancarz, *Fluid Phase Equilib.* 465, 1–9 (2018). https://doi.org/10.1016/j.fluid.2018.03.001.
- R. Chulist, L. Straka, A. Sozinov, T. Lippmann, and W. Skrotzki, Scr. Mater. 68, 671 (2013).
- E. Pagounis, R. Chulist, T. Lippmann, M. Laufenberg, and W. Skrotzki, Appl. Phys. Lett. 103, 111911 (2013).
- 26. H. Okamoto, JPEDAV 31, 575-576 (2010).
- C. Schmetterer, H. Flandorfer, C.L. Lengauer, J.-P. Bros, and H. Ipser, *Intermetallics* 18, 277–285 (2010).
- 28. W.X. Yuan and Z.Y. Qiao, JPEDAV 25, 68-74 (2004).
- T. Gancarz, P. Bobrowski, S. Pawlak, N. Schell, R. Chulist, and K. Janik, J. Electron. Mater. 47, 49–60 (2018). https://d oi.org/10.1007/s11664-017-5791-3.
- J.N. Pratt and J.M. Bird, J. Phase Equilib. 14, 465–472 (1993).
- J. Gröbner, R. Wenzel, G.G. Fischer, and R. Schmid-Fetzer, J. Phase Equilib. 20, 615–625 (1999).
- H. Okamoto, JPEDAV 29, 296 (2008). https://doi.org/10.10 07/s11669-008-9314-z.
- W.X. Yuan, Z.Y. Qiao, H. Ipser, and G. Eriksson, JPEDAV 25, 68–74 (2004). https://doi.org/10.1361/10549710417696.
- K. Micke, S.L. Markovski, H. Ipser, and F.J.J. van Loo, Ber. Bunsenges. Phys. Chem. 102, 1240–1244 (1998). https://doi. org/10.1002/bbpc.19981020930.
- S. Martosudirjo and J.N. Pratt, *Thermochim. Acta* 17, 183– 194 (1976).
- U. Häussermann, M. Elding-Ponten, C. Svensson, and S. Lidin, Chem. Eur. J. 4, 1007–1015 (1998).
- H. Okamoto, JPEDAV 31, 575–576 (2010). https://doi.org/10. 1007/s11669-010-9785-6.
- 38. E. Hellner, Z. Metallkunde 41, 480 (1950).

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