THE EFFECTS OF DISSOLUTION OF THE SOLID NICKEL IN LIQUID ALUMINIUM

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Abstract

The preparation of a large number of materials trough the propagating exothermic reactions has been the objective of numerous investigation. Self-propagating reactions are typically associated with high temperatures and extremely steep thermal gradients and thus offer the opportunity to investigate the formation of intermediate and metastable phases. Such reactions are also of interest from a practical point of view since, as has been described in numerous literature accounts, they provide the opportunity to prepare materials with unique properties.

In this study the thermal effects of dissolution on the solid nickel in liquid aluminium and formation intermetallic phases was studied by simple experimental procedure. The thermal effects generated by exothermic solid-liquid metal interaction was judged by the rise of temperature in the system. Depending on experimental conditions, two different regimes of behavior were found: solid-liquid interaction leading to the considerable rise of the temperature in the system, and characterized by continuos decrease of the temperature in the system with time. The results of the experiments on the heat effects during the interaction of solid nickel with different particle size and liquid aluminium, with and without external mixing, are presented. It has been shown that, depending on experimental conditions, the evolved heat may influence the structure of a zone near to the solid-liquid interface. The experimental procedure correlates with the practical conditions of semi-industrial production of aluminium-nickel master alloys with relatively high percentage of nickel.

Keywords: aluminium-nickel master alloys; solid nickel; mechanical alloying

1 Introduction

It has be known for many years that exothermic reactions between metals, when at least one of them is liquid, leads to considerable increase of temperature in the system [1-5]. The reactions between metals are generally less exothermic than those between metals and non-metals. It is obvious from thermodynamical data, that formation of nickel aluminides is accompauning by the largest amaunt of heat released. The formation of nickel-aluminium intermetallic phases are a subject of special interest, because of the exothermic nature of their formation, which suggests high stability and because of their attractiveness as promising new materials [6-9]. The nickel-aluminium phase diagram [10-12], shawn that the equiatomic phase AlNi has the highest value of negative heat of formation (58.9 kJ/g-atom), but Al_3Ni_2 has a value (56.8 kJ/g-atom), close to

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it. It may be concluded that formation of AlNi or Al_3Ni_2 will give high degree of heat generation, which will have in turn, serious effects on the process of melting of aluminium master alloys [13-16]. In this paper the results of an investigation on exothermic effects accompauning solid nickel-liquid aluminium interaction are presented. Experimental conditions related to the semi-industrial practice of aluminium-nickel master alloy production have been used. The process was used to study solid nickel-liquid aluminium interaction under different experimental by changing mixing conditions, nickel particle size, temperature of liquid aluminium, temperature of solid nickel, charging procedure. In the paper, we report the results on the influence of mixing, nickel particle size and temperature of solid nickel. These results may be of interest in the production of aluminium master alloys.

2 Experimental

The thermal effects from solid nickel-liquid aluminium interaction were investigated using simple device consisted of shell molded cup and chart recorder (**Fig. 1**).

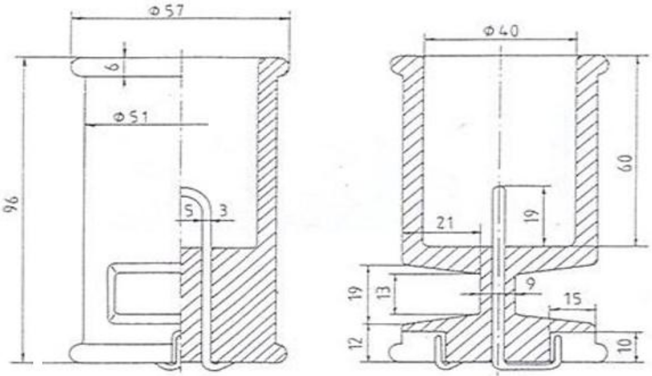

Fig. 1 Diagram of the shell molded sand cup (Tectip thetmocouple, Leeds&Northrup) used for investigation of the change of temperature during solid nickel-liquid aluminium interaction

The temperatures are measured by thermocouple type S, placed in the glass tube at the bottom of the cup. This ensure different time-temperature profiles to be observed during solid-liquid metal interaction. This type experimental procedure is frequently used to measure the changes in the temperature of molten metals. Spherical nickel particles with 99,99 wt. % were used as a solid metal reactant. Nickel particles were classified in size fraction : -4,0+3,0; -3,0+2,4; -2,4+1,8; -1,4+0,5; and -0,5+0,0 mm. 2 kg aluminium (99,97 wt. %Al) was carried out in normal atmosphere. The temperature of molten aluminium was continously recorded by immersed thermocouple type K. The ratio of Al/Ni quantities was designed to produce 20 wt. % aluminium-nickel master alloy. The metals were brought in to contact by pouring liquid aluminium directly from induction furnace into shell molded cup containing nickel particles placed at its bottom. The temperature of molten aluminium in the furnace was measured to be 1050^oC. Nickel parcticle classes were preheated in the graphite crucible to the temperature between $750-800^oC$. Mechanical mixing or vibration were applied approximately 10 seconds after solid-liquid metal contact was established. Different time-temperature curves were obtain upon solid nickel-liquid aluminium interaction as a function of mixing conditions (mechanical mixing, no mechanical mixing, vibration mixing), nickel particle size fractions, and temperature of the solid nickel particles (room temperature and 750-800°C).

3 Results and disscusion

The temperature-time response of nickel-liquid aluminium interaction in the presence of mechanical mixing, for five different nickel particle size fractions are shown in **Fig. 2**. With exception of curve 1, three characteristic points are observed on the curves: the initial temperature (*Ti*) of the system after liquid aluminium was poured in the sand cup; the lowest temperature of the system (*Tl*) registered 10 seconds after the cup was filled up with liquid aluminium; maximum temperature (*Tm*) reached after mechanical mixing was applied. Although, these three points may be detected on the curves, it is obvious that the courses of the curves, as well as the values of the characteristic temperatures are different.

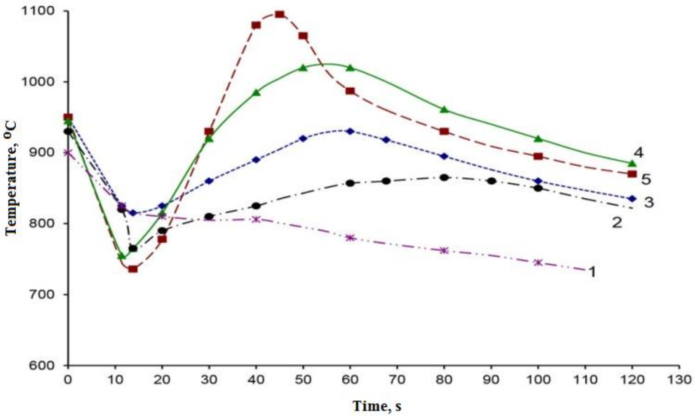

Fig. 2 The effect of nickel particle size on the temperature changes during solid nickel-liquid aluminium interaction. Nickel particles size: -4,0+3,0 mm, curve 1; -3,0+2,4 mm, curve 2; -2,4+1,8 mm, curve 3; -1,4+0,5 mm, curve 4 and -0,5+0,0 mm, curve 5

The results show that the change in the course of the curves depends on the nickel particle size. The most extreme changes in temperature is depicted for the sample containing smallest particle size (curve 5). On the other hand, the course of the curves are more flattened going to the samples with coarser nickel particle fractions (curves 2 and 3). In the case where sample with the most coarse particles was used, no rise in temperature is observed in spite of the presence of mechanical mixing. In contrast, continous decrease in temperature with time is registered. The differences in the values of *Ti* may be attributed to the experimental errors and the experimental procedure itself, but as a general picture it can be concluded that the values of *Ti* are grouping mainly between 950-970°C. Similar grouping of the values are observed for *Tl*, in spite of certainty of solid nickel-liquid aluminium interaction and heat release after solid-liquid metal contact was established. It is clear that the heat release during 10 seconds of contact without mixing does not result in observable increase in temperature of the system. The major differences are seen for the values of *Tm*, 30-50 seconds after mechanical mixing was introduced. In the case of the samples with -1,4+0,5 mm fraction, and especially in the samples with -0,5+0,0 mm, nickel particle size fraction, the temperature rise is well above *Ti*. The rise in temperature following mechanical mixing is a result of solid nickel-liquid aluminium interaction. This interaction is accompanied by strong exothermic effects. Since the heat of formation is closely related to the type of bonding, it is obvious that the large exothermic heat of formation is a result of changes in the nature of atomic bonding. Microprobe and x-ray analysis have shown that under the given experimental conditions mainly Al_3Ni_2 intermetallic phase is formed [10], which is in agreement with the results of other autors [11].

Sample considerations indicate that the amount of intermetallic being formed, is proportional to the solid-liquid metal contact. It is clear that more contact is expected when small nickel particle fraction (high surface area) are used enhancing interaction and heat generation. No rise temperature is observed (curve 1), when coarse nickel particles (-4,0+3,0 mm) are reacting with liquid aluminium, although some heat release must take place. This relatively small amount of release heat may be the cause of the plateau seen on time-temperature curve. The effect of mixing on the temperature for samples with different grain size is shown in **Fig. 3**.

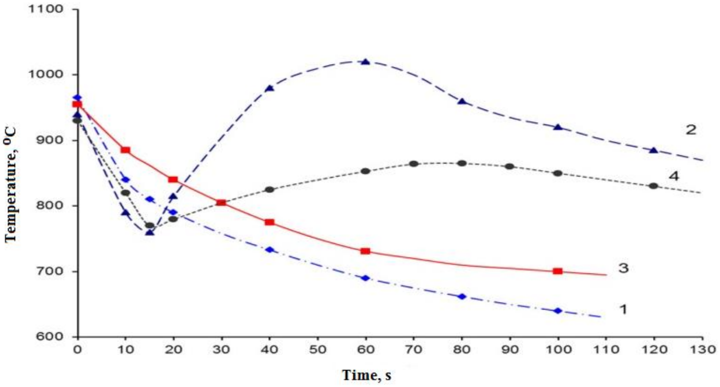

Fig. 3 The effect of mixing on temperature changes during solid nickel-liquid aluminium interaction. Curves 1 and 3, for interaction without mechanical mixing, curves 2 and 4, in the presence of mechanical mixing. Grain size fraction of nickel particles: -1,4+0,5 mm (curves 1 and 2), and -2,4+1,8 mm, (curves 3 and 4)

Two modes of change in temperature with time are observed. Curves 1 and 3, follow continous decrease in temperature with time irrespective of nickel grain size in the sample. In both cases solid nickel-liquid aluminium interaction proceeds without mechanical mixing. As can be expected, when mechanical mixing was applied the temperature of the systems starts to rise. The effect of mechanical mixing is more pronounced in the sample with smaller particle size, (curve 2). Although the picture on the influence of mixing is rather qualitative, since no data are available on the intensity of mixing, the presented experimental evidence emphasizes the importance of mixing to achieve sustainable rate of reaction if rise of temperature in the system is necessary to accomplished. Our results have shown that the mode of mixing has strong influence on the release of heat. The mode of mixing (electromagnetic in induction furnaces, vibration, mechanical mixing) makes the difference in amount of heat released. Highest increase in temperature was reached when mechanical hand mixing was applied.

The influence of temperature of solid nickel particles on temperature change during solid-liquid interaction was investigated using two different temperatures (**Fig. 4**). Curve 3 in **Fig. 4** was obtained by solid nickel-liquid aluminium interaction when nickel spheres at room temperature were brought in contact with molten aluminium in the presence of mechanical mixing. The difference between *Tl* and *Tm* in curve 3, is shown to be about 100°C. With nickel particles preheated at 750°C, solid nickel-liquid aluminium interaction gives the difference of about 150°C (curve 4). Since there is not abrupt changes in the course of two curves representing time-temperature profiles when no mechanical mixing is involved (curves 1 and 2), it can be concluded that the temperature of the solid metal (up to 750°C) is not the major factor causing extensive heat release. Slower decrease in temperature during cooling seen in curve 2 is expected because of the preheated solid metal, but slight change of the shape of the curve is also noticed in 20-30 seconds after mixing was applied, caused probably by limited extent of solid-liquid interaction.

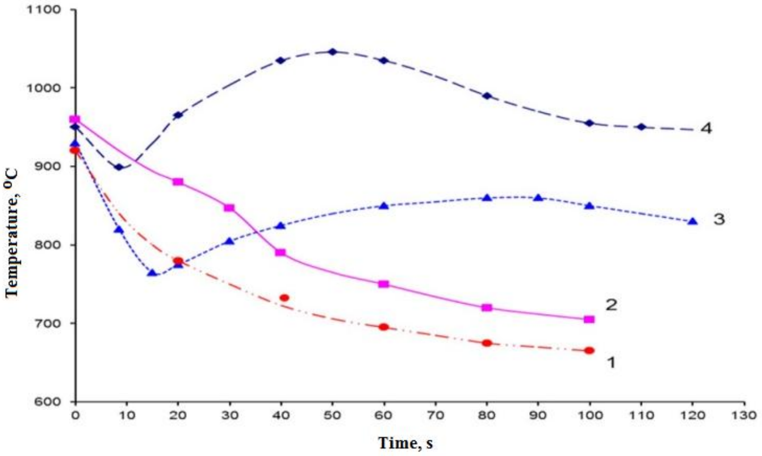

Fig. 4 The influence of the temperature of solid nickel particles on the temperature change in the system during solin nickel-liquid aluminium interaction with (curves 3 and 4) and without (curves 1 and 2) mechanical mixing. Nickel particles of -2,4+1,8 mm grain size are used at room temperature (curves 1 and 3), and preheated at 750°C (curves 2 and 4)

The samples that were gained when experimental procedures were executed, they were examined by SEM microscopy and microanalysis. The aim of these examinations is to obtain information on interactive zone between the components in accordance with the set experimental conditions. The main conclusion form the obtained results is that on the interphase surface solid-liquid layers of the intermetallic phases are formed. Their thickness and type phase depend on the achieved system temperature, when interaction of the reactants occurs and it depends on the quantity of the dissolved nickel within the solution. Samples which did not achieve contact between the solid nickel and liquid aluminium are obtained or no interaction among reactants exists. This is due to the chillness of the aluminium for the account of the nickel which is at room temperature and absence of the mechanical mixing of the solution. However, on the surface layers of intermetallic phases are formed as a result of the contact between reactants and non-mechanical mixing. The results from the SEM microanalysis (**Fig. 5**) of the sample obtained during solid nickel with particle size -2,4 + 1,8 mm, and liquid aluminium interaction (**Fig. 2**, curve 3), they show that the layer to the nickel contains 39.90 wt. %Al and 59.89 wt. %Ni, and the layer to the aluminium contains 58.53 wt. %Al and 41.42 wt. %Ni. These chemical composition of the layers confirm the formation of Al_3Ni_2 and Al_3Ni, intermetallic phases. It should be emphasized that the already formed intermetallic phase Al_3Ni_2, in the conditions related to the obtained sample is the first equilibrium solid phase with the solution, which is in accordance with the binary diagram Al-Ni [17]. Some of the samples which were obtained during the realization of the experimental procedures, in which nickel with small particle size was used (-0,5 + 0,0 mm), and the use of the mechanical mixing of the solution was applied, show different area of intermetallic layers formation. When these kind of conditions, the formed layer to the nickel contains 30.42 wt. %Al and 68.69 wt. %Ni, (**Fig. 6**), which matches to the content of the high temperature phase AlNi, and the second layer contains 47.35 wt. %Al and 53.56 wt. %Ni, which confirms Al_3Ni_2 phase formation [18, 19]. The occurrence of the high temperature phase AlNi, is due to the enhance of the system temperature (over 1135^{o}C) as a result of the released heat when the components interact. Temperatures over 1135^{o}C are not registered during the experiment realization, but it is assumed that they are achieved locally on the interphase surface solid-liquid [20, 21].

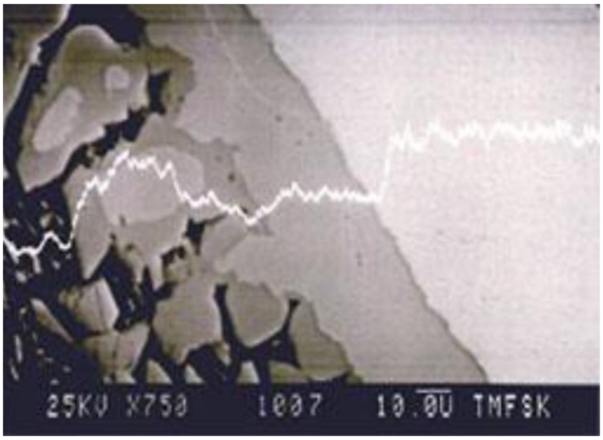

Fig. 5 SEM microphotography of a sample obtained during solid nickel and liquid aluminium interaction (**Fig. 2**, curve 3), with microanalysis of nickel

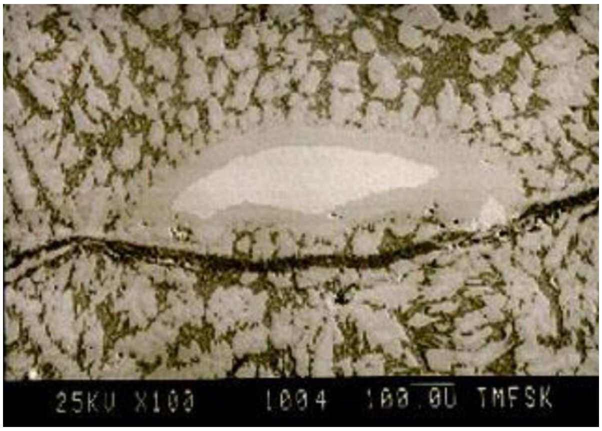

Fig. 6 SEM microphotography of a sample obtained during solid nickel and liquid aluminium interaction (**Fig. 2**, curve 5)

The presented results of the thermo-chemical effects examinations could not show if the released heat is a result of the intermetallic phases composition or it is due to the dissolution of the solid phase into liquid one. Due to practical application of the results from the examinations, it is the fact that by the mechanical mixing and appropriate granulation application, significant amount of heat is released, which enhances the system temperature thus making accelerated transfer of the atoms of the solid phase in the solution and achieving the required concentration of alloying element within the alloy for shorter period. The discrepancies of the amount of the released heat that occur, are result of the discrepancies of the system mixing intensity and used granulation of the solid phase, i.e. the level of the contact solid-liquid. Although the following of the curve in further chilling, can give information of the phase changes within the system, they are of minor significance for the subject which shall be examined related to this work. Beside the evidences for intermetallic layers formation and the presence of the solid phase dissolved into liquid, the scanning electronic microscopy with microanalysis enables the composition of the formed phases to be determined. The examinations show that the intermetallic phase that often occurs on the interphase line directly on the solid non-dissolved nickel is Al_3Ni_2. Between this phase and the solid dissolved of nickel in aluminium, Al_3Ni phase richer with aluminium emerges.

4 Conclusions

The intention of this work was to investigate thermal effects during solid-nickel-liquid aluminium interaction under experimental conditions related to semi-industrial production of aluminium-nickel master alloy and formation of intermetallic phases at the interface. In spite of difficulties to determine and control all variables affected aluminium master alloy production on laboratory scale, the results of this investigation reveal the major factors influencing solid nickel-liquid aluminium interaction. Solid nickel-liquid aluminium interaction may result in considerable release of heat. The released heat is large enough to rise the temperature (*Tm*), of the system well above the initial temperature (*Ti*), of liquid aluminium. Main factors influencing heat release accompanying solid nickel-liquid aluminium interaction are mixing conditions and nickel particle size. When mechanical mixing is applied the temperature of the system increases with decreasing nickel particle size. No rise in temperature is observed when samples with large particles (-4,0+3,0 mm) are used. There is no indication of temperature rise in the absence of mixing for all fractions of nickel grain size. Only simultaneous cooperative action of both factors enhanced solid nickel-liquid aluminium interaction, giving rise to the temperatures of the system. Preheating of solid nickel particles up to 750^{o}C is not a major factor causing extensive heat release during solid nickel-liquid aluminium interaction. Thermal effects accompanying solid nickel-liquid aluminium interaction can be used to intensify the process of aluminium-nickel master alloy production. During dissolution of the solid nickel in liquid aluminium, a layer of intermetallic phases and compounds is formed and dissolved simultaneously at the solid-liquid interface. The type and the sequence of intermetallic phases formation at the interface depends on the temperature, the interaction time and the solid metal concentration in the liquid aluminium.

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