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INTERACTION OF SOLID NICKEL WITH LIQUID MIXTURE OF ALUMINUM AND NICKEL AND FORMATION OF INTERMETALLIC PHASES

Blagoj Rizov*, Jon Magdeski

Ss. Cyril and Methodius University in Skopje
Faculty of Technology and Metallurgy, 1000 Skopje, Republic of Macedonia

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Abstract

In this study the interaction between solid nickel and liquid aluminum was investigated from three main aspects: mechanisms of creation of intermetallic phases, determination of the type of intermetallic phases and the sequence of their formation and growing. The experiments were performed in a laboratory medium-frequent induction furnace in temperature range between 750–950 ^{o}C and interaction time up to 1200 s in reductive atmosphere. The intermetallides formed at the solid-liquid interface were studied by scanning electron microscopy, microprobe analysis and X-ray diffraction analysis.

Key words: solid nickel, liquid aluminum, intermetallides, solid-liquid interaction

Introduction

The interaction of solid metal and liquid metal sample as a heterogenic metallurgical reaction is virtually present in many processes such as: application protection cases over metal surfaces, soldering and welding, production of super-conductible materials, preparation of composite materials etc. Simultaneously, the production of casting alloys and refractory alloys is based on interaction of solid and liquid metal hence it arises the importance of researching and examining the basic laws of this process. Typical illustration of interaction solid-liquid is the process of alloying the smelted metal when preparing aluminum alloys and aluminum refractory alloys. Alloying is usually made by dissolving the solid metal in liquid aluminum where two cases are possible. In the first case, the solid metal has a lower melting temperature then the melted metal and the metal being dissolved revolves into liquid condition and its further distribution in the melted metal is connected with the diffusion and convective

* Corresponding author: Blagoj Rizov, rizov@tmf.ukim.edu.mk

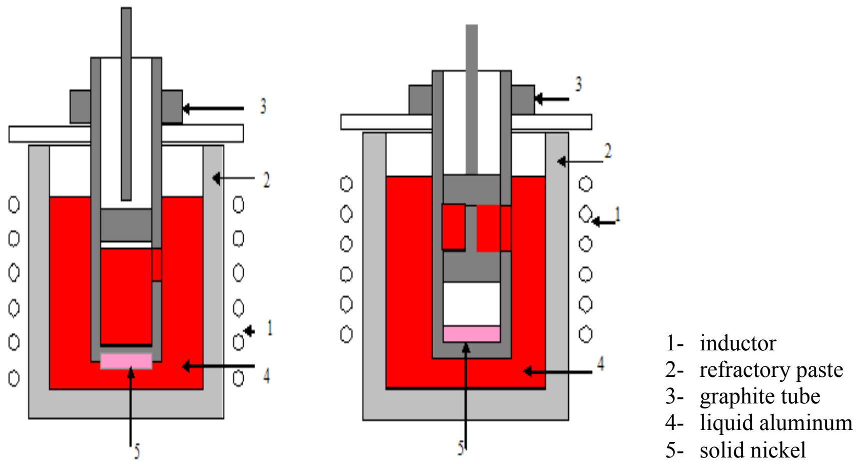
transfer of mass. In the second case, the metal being dissolved possesses higher melting temperature then the liquid metal. In such conditions, dissolution of solid metal in the melted metal may be accomplished via direct diffusion of the solid metal atoms in the melted metal or via temporary formation of a layer of intermetallic phases or compounds at the solid-liquid interface [1]. In the later case, the examination of interaction of solid and liquid metal is impeded because of the involvement of numerous factors influencing the process. Hence, the problem of interaction of solid and melted metal by formation of intermetallic phases and compounds is usually referred to by considering only separate aspects. The examinations on interaction of low melting metals from the group of refractory elements (Cr, Ni, Mn, Fe etc.) and liquid aluminum is interesting from both theoretical and practical aspect. Thus, it is important to examine the formation of intermetallic layers on the surface of solid phase and their impact on the further dissolution of the solid metal in the liquid aluminum. Current review of bibliography data [2] reveals that in case of interaction in metal-metal systems, there is one or more intermetallic phases or compounds formed at the interface, depending on the thickness of the diffusion couples, temperature, interaction time etc. Beside the proposed mechanisms by different authors still there is a large discrepancy in terms of which mechanism would be generally more acceptable for formation of intermetallic phases during solid-liquid interaction. Literature data are fairly poor on this issue with exception of some systems.

Based on the above mentioned, this paper encompasses realization of series of experimental procedures that contain the following examinations: metallographic analysis of obtained intermetallic phases and compounds during interaction of massive peaces of solid nickel and liquid aluminum, determination of the type of intermetallic phases, the sequence of their formation and growing with the time and temperature.

Experimental

Technically pure aluminum (99.7% Al) and nickel with purity of 99.9% Ni for laboratory examinations were used. The nickel was added in form of balls with granulation (-12 +0.0) mm and in form of discs with diameter of 15-16 mm. The height of the nickel disks was in the range of 5-7 mm. The material was fed in the melted metal at a room temperature or previously heated up at temperature between 100°C and 500°C.

In order to establish contact between the solid metal and the liquid aluminum, special graphite test-tube has been constructed according to the author's design, Figure 1. The graphite test-tube is consisted of the tube body and mobile piston. By moving the piston to the utmost lower position, isolation is conducted of a certain working area from the atmosphere where contact between the two reactants is established. The isolation of the working area from the atmosphere inside the test-tube is carried out through submerging the test-tube into the melted aluminum. Thus, there is a certain quantity of oxygen from the trapped air in the working area which is surrounded with constant source of carbon from the graphite. At the prescribed experimental conditions, it is assumed that in the working area reductive atmosphere will occur (formation of CO_g), which shall execute reduction of the oxide layer at the surface of the solid metal. To define the starting conditions of the contact between the solid metal and the smelted metal, graphite test-tube with an open piston has been used whereas the time frame in which the temperature of the solid metal equalizes with the temperature of the melted metal is determined. The temperature of the solid metal surface was measured with thermocouple type K, placed at the opening of the piston. The time of equalization of the temperatures of both reactants was 5 minutes.



a) *b)*

Figure 1. Outline of the contact establishment process between the reacting components: a) equalization of the reactants temperature; b) contact establishment

Figure 1b shows the outline of the contact establishment process between the solid nickel and melted aluminum. Firstly, the sample of solid nickel is placed at the bottom of the working area in the graphite tube and then the working area is closed with the piston. The necessary quantity of aluminum is smelted and overheated at experimental temperature in the pot of the medium-frequent induction furnace. After the experimental temperature (750 or 950 ^{o}C) is achieved, the graphite test-tube together with the solid metal sample is sunk into the solution. Five minutes later, the solid metal reaches the temperature of the melted metal and the piston is lifted (opened) 10 minutes later. Thus, through the opening of the test-tube, which is placed deeply in the melted metal, pure aluminum enters and creates a contact with the solid metal. It is assumed that during this process, the oxide layer is largely reduced and there is contact realized between pure metals.

After preparation of the samples with grinding and polishing, the examinations were directed towards determination of the microstructure of the intermediate zone between the two reacting components. The microstructural examinations should provide information about the nature and morphology as well as the thickness of the formed intermetallic phases (layers) at the interface. For these experiments the scanning electron microscope of type JEOL SM-35 was used. Simultaneously, SEM-microanalysis (EDAX-quantitative and qualitative) for identification of the type of intermetallic compounds and phases formed during the interaction of the solid nickel with liquid aluminum was conducted.

Also, to confirm the presence of the intermetallic phases and compounds at the interface, beside the scanning-electron microscopy with microanalysis, the method of X-ray diffraction was used.

These examinations are conducted in a two ways. The first one was reaction of solid nickel with a pure aluminum at temperatures of 750 ^oC and 950 ^oC and the times of interaction from 1 to 1200 s. The second was interaction of solid nickel with a mixture of liquid aluminum and different contents (5, 10 and 30 mass %) of dissolved nickel at the same temperatures and times.

Application of massive peaces suggests a constant source of the reactants for diffusion and reaction. Different geometries have been used for the solid-liquid contact trough alteration of the form of the solid metal (balls and discs).

Results and Discussion

During interaction of the solid nickel and liquid aluminum, a layer of intermetallic phases and compounds is formed and dissolved simultaneously at the solid-liquid interface. The type and the sequence of formation of intermetallic phases at the interface depend on the temperature and the interaction time.

The results of the examinations of the reaction zone between the solid nickel and liquid aluminum at temperature of 750 ^oC and interaction time of 1, 6 and 15 s, shows that there is not formation of intermetallic phases on the interface. The occurrence of intermetallic layer is noticed after interaction time of 60 s. The microstructure of the reaction zone between the nickel and aluminum after interaction time of 1200 s at temperature of 750 ^oC is shown on Figure 2.

The results of the SEM microanalysis of the formed intermetallic layer, in all obtained samples, when interaction between the solid nickel and the liquid aluminum takes place at temperature of 750 ^oC and interaction time of 60-1200 s, exhibit that the layer is mono-phased and contains from 58.97 to 60.42 mass % Al and 39.58 to 42.57 mass% Ni. This composition matches the intermetallic compound Al_3Ni, meaning that the interface solid-liquid under these conditions consist of only one intermetallic compound formed.

During the interaction between solid nickel and liquid aluminum at temperature of 950^oC and time from 1 to 6 s, the formation of intermetallic phase Al_3Ni on the interface was noticed but only at certain locations. The morphology of this phase indicates formation with crystallization of melted material when cooling the sample. In case of longer times (15, 30, 60, 600 and 1200s), in the same experimental conditions, two intermetallic layers were formed. The first layer was immediately next to the nickel and has approximately uniformed thickness while the second one was branched and borders with the aluminum, Figure 3.

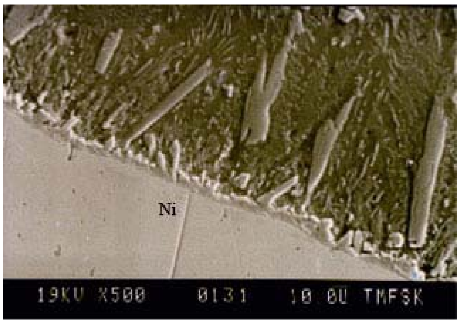


Figure 2. SEM micrograph of a sample obtained during interaction of solid nickel and liquid aluminum at 750 °C, 1200 s.

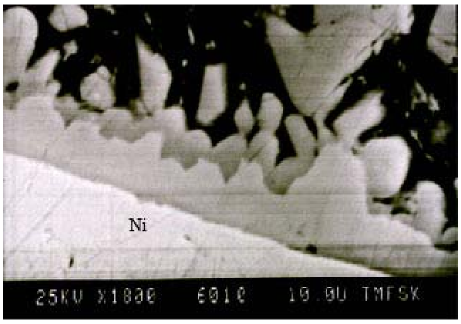


Figure 3. SEM micrograph of a sample obtained during interaction of solid nickel and liquid aluminum at 950 °C, 30 s.

The results of the SEM microanalysis confirm that the first layer presents intermetallic phase Al_3Ni_2 and the second one is Al_3Ni. When the solid nickel and the liquid aluminum interact at temperature of 950°C in set time intervals of interaction, a layer of Al_3Ni_2 is formed with approximately equal thickness of around 7-10 μm. Thickness measurement of the formed Al_3Ni_2 layer has been conducted for all set time intervals. The obtained values are graphically shown on Figure 4 where the variation of the Al_3Ni_2 layer thickness can be seen in relation to the time.

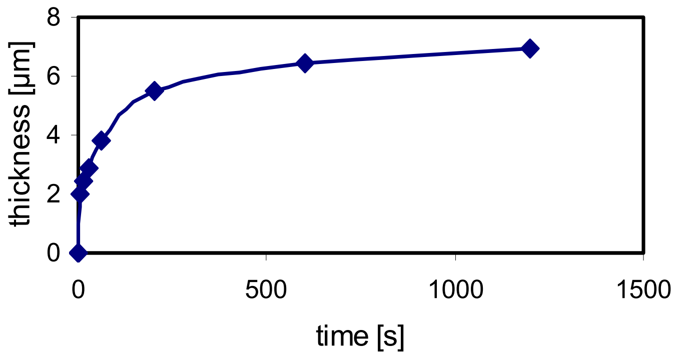


Figure 4. Dependence of the thickness of Al_3Ni_2 intermetallic layer with time, obtained during interaction of solid nickel and liquid aluminum at 950 °C.

In order to confirm and identify more precisely the intermetallic phases and compounds, X-ray analysis was conducted on specially prepared samples. The sample for analysis was obtained with interaction of solid nickel and liquid aluminum in graphite test-tube at temperature of 950 °C and time period of 1200 s. The X-ray diagram, Figure 5, quite clearly identify the intermetallic phase Al_3Ni_2 marked with number 1 and the compound Al_3Ni (2), but with much lower quantitative participation.

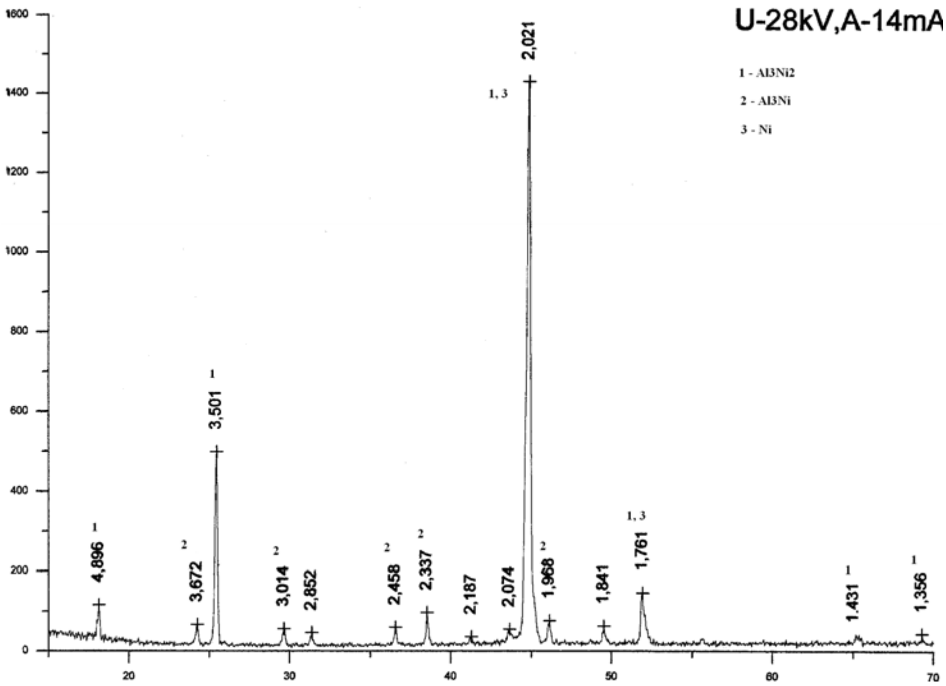


Figure 5. XRD of a sample obtained during interaction of solid nickel and liquid aluminum at 950 ^oC after 1200 s.

The microstructural examinations over the reaction zone during interaction of solid nickel and liquid aluminum with 5 mass % Ni at temperature of 950 ^oC in interaction time of 6, 60 and 600 s illustrate that in such conditions at the solid-liquid interface, again there is formation of two intermetallic layers. The layer with equal thickness in immediate vicinity of the nickel is Al_3Ni_2, and the second layer which is branch-like is Al_3Ni, Figure 6. The formation of these two phases is confirmed by the values of the chemical content of the layers obtained via SEM microanalysis of the mentioned samples. It is important to emphasize that in such conditions of interaction when the aluminum already contains a certain quantity of nickel, the share of intermetallic phases increases emerging with crystallization when cooling the melted metal. Special attention attracts the example of interaction of the solid nickel with liquid aluminum at temperature of 750 ^oC when there are 10 mass % Ni dissolved in the solution. Figure 7, illustrates the microstructure of the reaction zone between the nickel and the aluminum solution with 10 mass % Ni at T=750 ^oC and interaction time of 600 s. The results of the microanalysis of the intermetallic layers demonstrate that in such conditions, the phase Al_3Ni_2 is formed next to the nickel and the compound Al_3Ni is formed next to the aluminum. Also, it should be emphasized that these intermetallic layers were formed in very short time (6 s) unlike the example of interaction of solid nickel with pure aluminum where in the same conditions (times of 15-1200 s), formation of only Al_3Ni intermetallic layer is noticed. In the later, the intermetallic layer (Al_3Ni) was formed after interaction time of 15 s. Thus, it can be concluded that in case of aluminum solution with 10 mass % Ni and at temperature of 750 ^oC, first the Al_3Ni is formed first and in very short time interval (up to 6 s), the critical thickness of the layer is attained, which is a basic condition for the formation of the Al_3Ni_2 layer.

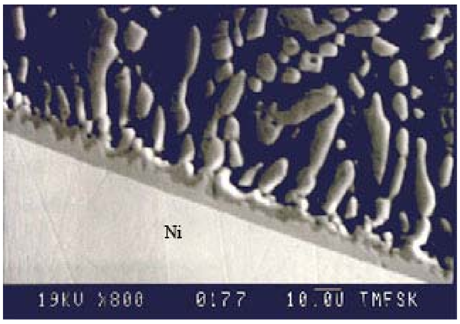


Figure 6. SEM micrograph of a sample obtained during interaction of solid nickel and solution of aluminum with 5 mass % Ni at 950 ^oC after 60 s.

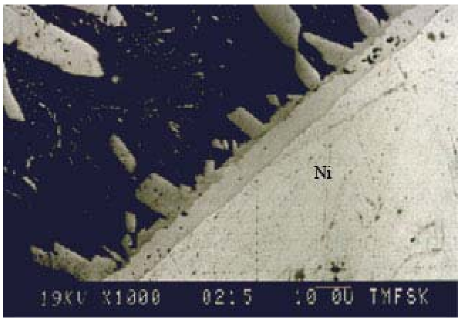


Figure 7. SEM micrograph of a sample obtained during interaction of solid nickel and solution of aluminum with 10 mass % Ni at 750 ^oC after 600 s.

The obtained results for the influence of nickel concentration in the smelted aluminum on the thickness of the formed Al_3Ni_2 layer, Figure 8, exhibit that the thickness increases within the increase of the interaction time. A conclusion can be drawn that the thickness of the layer is mostly formed in the starting period of interaction between the components (up to 60 s) and later the layer grows at a much slower rate. It is evident that the thickness of the layer is bigger at temperature of 750 ^oC when the aluminum contains 10 mass % Ni in comparison with the example when the aluminum contains 5 mass % Ni and the temperature level is 950 ^oC.

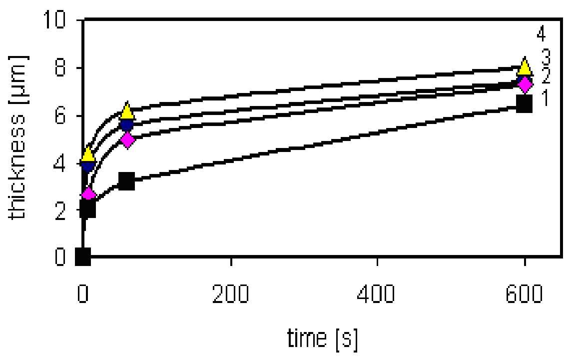
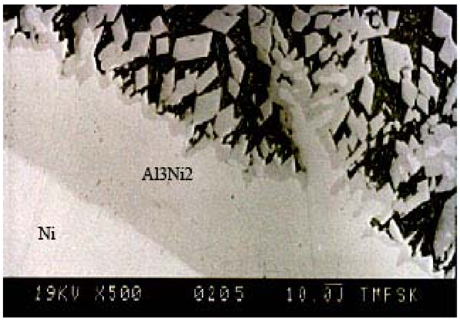


Figure 8. Dependence of the thickness of Al_3Ni_2 intermetallic layer with time, during interaction of solid nickel and liquid aluminum (pure and solution). 1 - Pure aluminum (99.7 mass % Al), at T=950 ^oC.; 2 - Solution of aluminum with 5 mass % Ni, at T=950 ^oC.; 3 - Solution of aluminum with 10 mass % Ni, at T=750 ^oC.; 4 - Solution of aluminum with 10 mass % Ni, at T=950 ^oC.

Such occurrence leads to the fact that the concentration of nickel in the smelted aluminum has higher influence on the thickness of the formed intermetallic layer then the temperature of interaction between the components. Summing up, the conclusion is that the thickness of the intermetallic layer increases with the increasing of the temperature and the increasing of nickel concentration in the solution.

The highest thickness of the Al_3Ni_2 intermetallic layer (~ 50 μm) is achieved by interaction of solid nickel and aluminum solution with 30 mass % Ni content at temperature of 950 ^oC after time period of 600 s. The microstructure of obtained layers is shown on Figure 9 and the dependence of the layer thickness with time is shown on Figure 10.



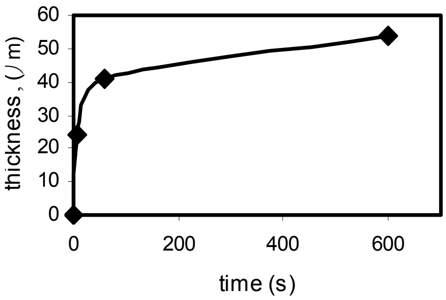


Figure 9. SEM micrograph of a sample obtained with interaction between the solid nickel and aluminum solution with 30 mass % Ni at T=950^{o}C and time period of 600s.

Figure 10. Dependence of the thickness of Al_3Ni_2 intermetallic layer with time, obtained during interaction of solid nickel and aluminum solution with 30 mass % Ni at T=950^{o}C and time period of 600 s.

This occurrence is reasonable considering that this nickel concentration approaches the concentration of saturation of the solution and with this the dissolving velocity of the intermetallic layer is brought down to minimum.

Conclusions

The interaction of solid nickel with pure aluminum (for the both temperatures) shows that the thickness of the Al_3Ni_2 layer increases in a parabolic way within time which indicates to a diffusion controlled process. At a particular temperature, the first formed intermetallic phases at the solid-liquid interface are the phases which are in equilibrium with the melted metal in compliance with the equilibrium binary diagram.

The change of nickel concentration in the smelted aluminum causes variation of the concentration gradient in the boundary Nernst's diffusion layer which is the major driving force for diffusion and conversion of the solid into liquid metal. The increase of nickel concentration in the melted metal reduces the concentration gradient thus also decreasing the velocity of dissolution of the formed intermetallic layers on the interface. In such conditions, where the velocity of formation becomes higher in comparison to the velocity of dissolution, it would be expected increasing of the formed intermetallic layers thickness.

The influence of nickel concentration in the smelted aluminum on the thickness of the formed Al_3Ni_2 layer exhibits that the thickness increases with the increase of the interaction time.

The highest thickness of the Al_3Ni_2 intermetallic layer (~ 50 μm) is achieved by interaction of solid nickel and aluminum solution with 30 mass % Ni content at temperature of 950 °C after time period of 600 s.

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