Evaluation of High-temperature Physico-chemical Interactions Between the H282 Alloy Melt and Ceramic Material of the Crucible

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Abstract
Nickel alloys belong to the group of most resistant materials when used under the extreme operating conditions, including chemically aggressive environment, high temperature, and high loads applied over a long period of time. Although in the global technology market one can find several standard cast nickel alloys, the vast majority of components operating in machines and equipment are made from alloys processed by the costly metalworking operations. Analysis of the available literature and own studies have shown that the use of casting technology in the manufacture of components from nickel alloys poses a lot of difficulty. This is due to the adverse technological properties of these alloys, like poor fluidity, high casting shrinkage, and above all, high reactivity of liquid metal with the atmospheric air over the bath and with the ceramic material of both the crucible and foundry mold. The scale of these problems increases with the expected growth of performance properties which these alloys should offer to the user.

This article presents the results of studies of physico-chemical interactions that occur between the H282 alloy melt and selected refractory ceramic materials commonly used in foundry. Own methodology for conducting micro-melts on a laboratory scale was elaborated and discussed. The results obtained have revealed that the alumina-based ceramics exhibits greater reactivity in contact with the H282 alloy melt than the materials based on zirconium compounds. In the conducted experiments, the ceramic materials based on zirconium silicate have proved to be a much better choice than the zirconia-silica mixture. Regardless of the type of the ceramic materials used, the time and temperature of their contact with the nickel alloy melt should always be limited to an absolutely necessary minimum required by the technological regime.

Keywords: Nickel alloys, Superalloys, Melting, Reactivity, Metal/ceramic reaction, Refractory materials

1. Introduction
Nickel alloys form a group of materials that in liquid state strongly react with the ceramics of which both the crucible in a melting furnace and casting mold are made. This applies in particular to superalloys, including H282, which contain a significant amount of reactive alloying elements such as aluminum and titanium. In H282 the content of Al exceeds 1wt% and that of Ti -2wt% [1-3]. These elements at high temperature are very intensively combined with oxygen.
Therefore, during melting, it is very important to reduce to minimum the amount of oxygen dissolved in liquid alloy\[4,5\].

H282 is an alloy of the latest generation and is regarded as a strategic material. For this reason, the available reference sources lack any reliable data on the thermo-physical and technological aspects of melting and solidification of this alloy in the high range of temperature values, necessary for proper development of the casting process and heat treatment of this alloy.

One of the many problems faced during melting of these alloys is the selection of proper refractory materials. This issue has been the subject of numerous studies undertaken by various research centers dealing with the melting and casting of nickel alloys. A number of research works in this particular field of knowledge have been carried out also by the Foundry Research Institute in Cracow\[6-8\], focusing attention on the development of ceramic materials characterized by a low reactivity with liquid nickel alloys.

2. Test methodology

To examine the melt/ceramic interaction, tests were carried out which consisted in melting the H282 alloy in a vacuum medium frequency induction furnace in a specially designed “insert” placed in the furnace heating inductor. Due to the use of this element it was possible to simultaneously melt under the same conditions several small batches of metal (up to 2kg), placed in specially designed small ceramic crucibles (“inserts”). Tests were performed by melting the H282 alloy in 3 crucibles made of different ceramic materials, i.e. alundum-crucible no. 1, zirconia+silica-crucible no. 2, zirconium silicate-crucible no. 3.

Figure 1 shows the test crucibles filled with a charge (H282 alloy), the location of control thermocouples and the process of metal melting.

Fig. 1. Photographs showing the laboratory melting system for nickel alloys based on a KOPP vacuum induction furnace: at the top- loading of charge, at the bottom- melting in one of the three small crucibles

The metal after melting was superheated to a temperature of 1450°C. At this temperature, it was held for 30 minutes and then the power to inductor was switched off, allowing the ingots to cool down together with furnace to ambient temperature.

3. Results

Tests showed different degree of the crucible damage (Figure 2) and of the metal/ceramic reaction products penetration into the ingot surface layer (Figure 3).

Fig. 2. The appearance of the inner surface of respective crucibles after melting the H282 alloy

1. Crucible no. 1 – alundum, crucible no. 2 – zirconia + silica, crucible no. 3 – zirconium silicate

Fig. 3. The appearance of the surface of ingots made from the H282 alloy melted in different crucibles

1. Crucible no. 1 – alundum, crucible no. 2 – zirconia + silica, crucible no. 3 – zirconium silicate
The ingots cast in individual crucibles were next subjected to metallographic examinations. Using an optical microscope, the outer layers of metal directly contacting the ceramic material were examined. The microstructure of these layers is shown in Figure 4. The observations of microstructure showed that the metal/ceramic reaction was progressing most intensively in crucible no.1 (large pits penetrating from the surface into the ingot interior). The metal from the other two crucibles behaved in a similar manner. The only difference was that the metal from crucible no. 2 contained under the outer ingot layer numerous colonies of titanium nitride precipitates.

![Fig. 4. Microstructure in the outer layer of ingots crucible no. 1 – alundum, crucible no. 2 - zirconia + silica, crucible no. 3 – zirconium silicate](image)

The aim of further metallographic studies was to carry out a chemical analysis in the edge layer of the cast ingots. Based on the results obtained, an attempt was made to identify the phases present in this layer. The study was conducted using an EDS LINK ISIS X-ray microanalyzer made by Oxford Instruments.

In the case of alundum crucible (crucible no.1), the edge layer of the ingot was found to be enriched in titanium, aluminum and oxygen, which means that oxides of those metals were present there. Figure 5 shows this effect.

![Fig. 5. Local analysis of the edge layer of an H282 alloy ingot melted in alundum crucible (arrows mark the examined points)](image)
In the case of zirconia + silica crucible (crucible no. 2), the edge layer of the ingot was found to be totally free from the titanium compounds, as illustrated in the graphs in Figure 6. On the other hand, the presence of aluminum oxides (point 3) and of the compounds of aluminum and silicon with a low content of chromium and nickel (points 1 and 4) was detected. Under this layer there were fine single precipitates of the complex compounds (point 2), and still deeper the precipitates of titanium nitrides (point 5).

Fig. 6. Local analysis of the edge layer of an H282 alloy ingot melted in zirconia + silica crucible (arrows mark the examined points)
When the crucible made of zirconium silicate is used (crucible no. 3), in the edge layer of the ingot, as shown in Figure 7, there are occasionally near the surface single complex precipitates of the size of about 1 µm, containing mainly chromium and silicon (point 1). In these precipitates, other elements also occur, including zirconium originating from the ceramic crucible. In the surface layer of the ingot, aluminum and silicon oxides appear (points 2 and 3), but the whole layer is relatively thin and less coherent than the same layers formed when crucibles made of zirconia + silica or of alundum (especially the latter ones) are used. Under the surface of this layer there are numerous precipitates with varied chemical composition (points 4 and 5).

The distribution maps of elements analyzed in the surface layer of the individual ingots, shown in Figures 8, 9 and 10, confirm that when alundum crucible is used, the layers of aluminum and titanium oxides are formed at the metal/ceramic interface. A weak interaction with this layer (dissolution) shows elements such as nickel and chromium, iron contained in the liquid H282 alloy and cobalt. No effect of molybdenum has been observed in this case (Figure 8).
Fig. 8. Distribution of elements in the metal/alundum crucible interface

Fig. 9. Distribution of elements in the metal/zirconia+silica crucible interface
In the case of zirconia + silica crucible, in the outer layer of an ingot, as shown in Figure 9, there is a zone of the large precipitates of chromium compounds. A small amount of oxygen present in these areas and a high content of silicon indicate that, in accordance with the Cr-Si phase equilibrium diagram, these can be the intermetallic phases, such as Cr$_3$Si, Cr$_2$Si, Cr$_5$Si$_3$,Cr$_5$Si or Cr$_3$Si$_2$. In these precipitates, certain amount of titanium is dissolved; other alloying elements were not found.

Under the layer of the precipitates of the chromium-silicon-titanium phases, there is a layer of aluminum oxides, as indicated by a large amount of both oxygen and aluminum.

Absence of chromium and aluminum in the material of the ceramic crucible, on the one hand, and high content of these elements in the H282 alloy, on the other, indicate that both the chromium-containing layer as well as the layer of aluminum oxides are formed by a reaction of these elements with the crucible material at the liquid metal/ceramic interface.

When the crucible made of zirconium silicate was used, in the outer layer of an ingot there were some areas with the precipitates similar to those observed in the case of the zirconia + silica crucible. This is shown in Figure 10.

The observed, relatively small, amount of chromium, aluminum and oxygen in the outer layers of an ingot may indicate a lower reaction rate at the liquid metal/ceramic crucible interface, where the H282 alloy melt is in contact with the sole zirconium silicate and not with the whole mixture of zirconia and silica. This may result from a smaller amount of oxygen bounding the ceramics containing no silica.

4. Summary and conclusions

The developed methodology of studies of an interaction taking place between the liquid metals or their alloys and ceramic materials used for the refractory lining of induction furnaces enables carrying out tests in a laboratory scale to assess this interaction for the three different ceramic materials in contact with the same metal and under the same time and temperature conditions of the experiment.

By using an indirect charge heating technique, i.e. through a graphite insert placed in the furnace inductor, the designed and constructed test device allows evaluating the intensity of reaction taking place between the molten metal and a ceramic crucible in the case of different cast alloys (alloys of iron, nickel, cobalt, aluminum and copper) and different refractory ceramic materials from which the test crucibles are made.

By placing the designed laboratory device in a vacuum induction furnace it becomes possible to use a protective atmosphere (vacuum, nitrogen, argon) during the test melting, thereby limiting access of atmospheric oxygen to the melted charge, first, and to the liquid metal, next.

In the conducted experiment, the intensity of reaction of the H282 alloy with the three selected, commonly used in foundry processes, ceramic materials, such as alumina, zirconia+ silica and zirconium silicate was evaluated.

The H282 alloy contains a large amount of highly reactive alloying elements (aluminum, titanium), and therefore its melting or remelting causes serious problem related with the proper choice of foundry ceramic materials which will contact the liquid metal.

The observations made in the course of the experiments revealed that, as regards the intensity of an interaction with the tested ceramic materials, the contact of molten H282 alloy with the alundum ceramics is less favorable than with the ceramics
based on zirconium compounds. In the latter case, the use of zirconium silicate gives better results than the use of a zirconia + silica mixture, since the former material is less harmful as regards the effect of an interaction at the liquid metal /ceramic material interface (surface pits, reaction products). In spite of this, it is recommended to reduce to an indispensable minimum any contact of the molten H282 alloy with any of the ceramic materials (the time of melting as short as possible and the temperature of alloy overheating as low as possible).

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