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COMPOSITES THEORY AND PRACTICE

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THE WAY OF ESTIMATING INTERPHASE TENSION IN THE LIQUID ALUMINUM ALLOY - LIQUID SLAG

In refining treatments (removal of solid inclusions) related to cast aluminum alloys as well as the recycling of cast metal composite material [MMK] on the matrix of aluminum and its alloys, consisting in separation of the components, substances which form another liquid phase (aside from the liquid metal (alloy)) are used. With regard to aluminum alloys, these substances are both refining fluxes and so-called recycling centers. They are composed of molten salt mixtures (mainly chlorides and fluorides). The effectiveness of their action is determined by the value of interphase tensions on the boundary with a liquid metal (alloy), which should be as small as possible. The values of interphase tensions in such system are not very well known, which led to an attempt to specify them in greater detail. A measuring station intended to specify them by means of the so-called ring test was constructed. The principle of conducting such measurements is simple but the method of their performance is difficult due to the specific properties of biphasic systems at significantly elevated temperatures. To complete the test stand (facility), a laboratory resistance crucible furnace was used and a measurement system composed of a probe that cyclically crosses the interphase boundary, the probe drive mechanism and a force gauge that continuously registers the changes in the values of forces acting on the probe. A non-standard massive cast iron crucible with a considerable thermal inertia was placed in the furnace chamber. The metal input (aluminum alloy) and salt mixtures (being refining means or recycling centers) were melted in this crucible. Trials were carried out using fixed values of the probe dimensions, speed of the probe crossing the interphase surface and frequency of registering the force gauge indications. During each series of trials, changes in the temperature values of the tested systems were recorded. It was found that both the method of measuring interphase tensions and the method of its realization performed well, ensuring reliable and reproducible results. The only drawback was the material of the probes used. The stainless steel used to make them proved to have poor resistance to the operating conditions. Despite this, the method and way of determining the interphase tension values in the systems allowed successful realization of the intended research objectives.

Keywords: recycling, refining, interphase tensions, measurements

SPOSÓB OCENY NAPIĘĆ MIĘDZYFAZOWYCH W UKŁADZIE CIEKŁY STOP ALUMINIUM - CIEKŁY ŻUŻEL

W zabiegach rafinacji (usuwania wtrąceń stałych) odlewniczych stopów aluminium oraz recyklingu odlewanych metalowych materiałów kompozytowych (MMK) na osnowie aluminium i jego stopów, polegających na rozdzieleniu składników, stosowane są substancje tworzące drugą obok ciekłego metalu (stopu) fazę ciekłą. W przypadku stopów aluminium substancje te to zarówno topniki rafinujące, jak i tzw. ośrodki recyklingowe. Stanowią je stopione mieszaniny soli (głównie chlorki i fluorki). O skuteczności ich działania decydują wartości napięć międzyfazowych na ich granicy z ciekłym metalem (stopem). Powinny one być jak najmniejsze. Wartości napięć międzyfazowych w takich układach są mało znane, co spowodowało próbę ich dokładnego określenia. Skonstruowano stanowisko pomiarowe mające je określić za pomocą tzw. próby pierścieniowej. Zasada wykonania takich pomiarów jest prosta, lecz sposób jej realizacji jest trudny z racji specyficznych właściwości dwufazowych układów w znacznie podwyższonej temperaturze. Do skompletowania stanowiska badawczego użyto laboratoryjnego oporowego pieca tyglowego oraz układu pomiarowego złożonego z próbnika cyklicznie przekraczającego badane granice międzyfazowe, mechanizm napędu próbnika oraz siłomierza rejestrującego w sposób ciągły zmiany wartości sił działających na próbnik. W komorze pieca umieszczono nietypowy, masywny tygiel żeliwny o dużej bezwładności cieplnej, w którym topiono zarówno wsad metalowy (stop aluminium), jak i mieszaniny soli stanowiące środki rafinujące lub ośrodki recyklingowe. Próby przeprowadzono, stosując stałe wartości wymiarów użytych próbników, prędkości przekraczania przez próbnik powierzchni międzyfazowej oraz częstość rejestrowania wskazań siłomierza. W trakcie każdej z serii prób rejestrowano zmiany wartości temperatury badanych układów. Stwierdzono, że zarówno zastosowana metoda pomiaru napięć międzyfazowych, jak i sposób jej realizowania, zdały egzamin, zapewniając uzyskanie miarodajnych, powtarzalnych wyników. Jedynym mankamentem okazał się materiał stosowanych próbników. Stal nierdzewna służąca do ich wykonania okazała się mało odporna na warunki, w jakich one pracowały. Mimo tego, wykorzystana metoda i sposób określania wartości napięć międzyfazowych w badanych układach pozwoliły na pomyślne zrealizowanie zamierzonych celów badań.

Słowa kluczowe: recykling, rafinacja, napięcia międzyfazowe, pomiary

INTRODUCTION

Interphase tensions constitute a very common physical category present in various disciplines, not only in engineering and technology. They play a crucial role in phenomena occurring among others in metallurgical processes related to refining metals and alloys, the recycling of cast metal composite materials etc. Since these processes and their related phenomena occur at significantly increased temperatures, it is methodically difficult to numerically determine these tensions, and what is more they are performed in environmentally onerous conditions. The need to define the interphase tensions in recycling systems, which is concerned with separating the composite material elements obtained in the aluminum alloy matrix, has led to the development of an effective method of determining them, ensuring reliable results. One of the oldest methods of measuring these tensions was applied, while the tools adapted to perform the task, despite their simplicity as well as the drawbacks that emerged during testing, enabled the authors to achieve the intended goal. A decision was taken to present the proven method of determining interphase tensions on the boundaries of liquid aluminum alloys and liquid slag to a large group of interested persons.

INTERPHASE TENSIONS

Interphase tensions are present on boundaries of two mutually non-soluble liquids. They are the result of an imbalance of forces influencing the atoms or particles of both phases in the boundary zone which separates these phases [1]. Their values are an essential parameter of systems present and used in various disciplines of engineering (chemistry, construction, metallurgy, etc.).

In metallurgical systems are two liquid phases: molten metal (or alloy) and liquid slag. The course of the phenomena, their intensity and thus process effectiveness are determined by values of tensions on the boundaries of these phases. In metallurgy where aluminum alloys are used, refining treatments which eliminate solid inclusions from baths, e.g. oxides, are most frequently applied. In the recycling process of cast metal composite materials based on a matrix of aluminum or its alloys, consisting in separating ingredients, the non-metal liquid phase, also referred to as the recycling center, also needs to be included [2]. This center takes particles from the reinforcing phase from the composite suspension obtained in the process of melting MMK waste (with regard to so-called suspension composites) or replaces the metal matrix in the capillaries of the material reinforcing a composite with saturated reinforcement [3-5]. It is necessary to stress that the mechanisms of refining aluminum alloys aimed at removing solid inclusions and separating phases forming MMK suspensions are identical. The essence is to take the solid particles forming the suspension in the liquid metal (or alloy) and to transform them into a suspension state in refining slag or a recycling center. Analysis of this mechanism proves that the interphase tensions on the boundary of the liquid composite matrix and liquid recycling center or refining slag should have as low a value as possible.

While the chemical compositions of numerous salt mixtures forming refining fluxes intended for aluminum alloys, thus useful in recycling processes of MMK on an alloy matrix, are well-known, the values of interphase tensions in these systems are only roughly estimated [6, 7]. Determining them has been deemed purposeful and useful for both research and application reasons.

TEST STAND

A review of the measurement methods which enable determination of interphase tensions (liquid-liquid) [8, 9] taking into account the nature of the systems: liquid aluminum alloy-molten salt mixtures, led to an attempt to use the ring method [10], which can be considered one of the oldest yet simplest methods. A diagram of the method is presented in Figure 1.

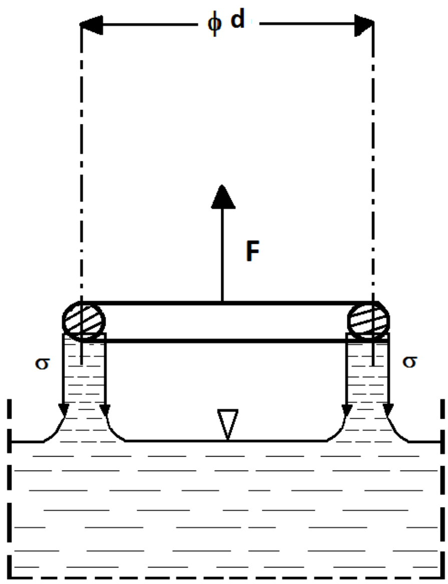


Fig. 1. Ring method diagram

Rys. 1. Schemat metody pierścieniowej

It consists in registering the values of forces influencing the metal probe, whose shape is a horizontally oriented ring, when it transforms from the liquid phase to gas phase. The value of this force at the moment when the probe is removed from the liquid is as follows:

$$F = \sigma_{C\text{-}G} \cdot 2l \quad [\mathrm{N}] \tag{1}$$

where: $\sigma_{C\text{-}G}$ - surface tension on the boundary of liquid and gas phase [N/m]; l - length of contact line of liquid phase with the probe [m].

The digit 2 denotes the two surfaces of the drawn "film" of the liquid phase and since the probe is of a ring-like shape with diameter *d*:

$$F = 2\sigma_{C-G} \cdot \pi d \quad [N] \qquad (2)$$

The value of the registered force F at the moment when the liquid „film" is removed from its horizontal surface is the liquid surface tension and equals as follows:

$$\sigma_{C-G} = \frac{F}{2\pi d} \quad [N/m] \qquad (3)$$

This method is used to determine values of the surface tensions (liquid-gas) and interphase tensions (liquid-liquid) in system of non-dissoluble liquid but at temperatures close to the value of room temperature. The prerequisite for achieving a credible result is excellent wettability of the material used to make the probe by the test liquid.

It was assumed that the trials performed on the boundaries of two liquids would enable determination of the values of interphase tensions on their boundary. During the initial trials carried out at room temperature with the use of transparent mediums conducted with the testing machine INSTRON 4481, the performance of the measuring unit was checked and the speed of the vertical movement of the probe through the boundary of system phases was established and may even be 20 mm/min.

The test stand was created and its diagram is presented in Figure 2. For the purposes of the melting the system (metal-flux) and maintaining its liquid state, the laboratory resistance furnace NABERTHERM equipped with a massive, thick-walled cast iron crucible was used. The crucible ensured considerable thermal inertia of the system in question, which was important because in periods when the measurements were taken the furnace power supply was switched off in order to avoid potential impact of electromotive forces on the force gauge indications.

The force gauge (model FH2 by SAUTER) with a resolution of 0.001 N, maximum load 2 N and precision 0.2% of maximum load, was connected to the servo drive fixed on the bearing arm attached to the mobile hoist frame. Thanks to this solution, it was possible to place the probe in the crucible symmetry axis with ease and the operation of the furnace was not hindered.

The probes were made of stainless steel wire (ϕ 0.8 mm). To shape them, two sections of the wire were wrapped around a rigid steel pipe whose external diameter was a pattern reflecting the internal diameter of the probes. Due to dimensions of these probes, it was possible to adopt values of the denominator in the dependency (3) - equal 0.382 m. The tips of the wires took the form of the stirrup and clamped inside the aluminum tube. The probes intended for the tests are presented in Figure 3.

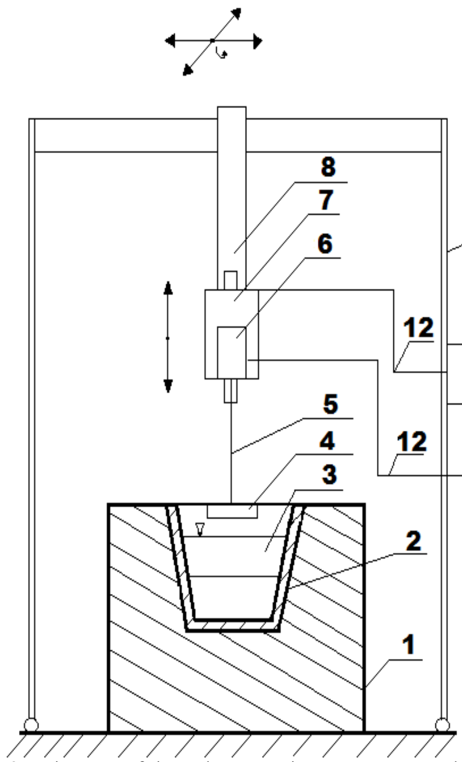


Fig. 2. Diagram of interphase tension measure stand: 1 - resistance furnace, 2 - cast iron crucible, 3 - system in question, 4 - probe, 5 - probe rod, 6 - force gauge, 7 - servo drive, 8 - bearing arm, 9 - hoist frame, 10 - servo drive control, 11 - computer, 12 - electric wiring

Rys. 2. Schemat stanowiska do pomiarów napięć międzyfazowych: 1 - piec oporowy, 2 - tygiel żeliwny 3 - badany układ, 4 - próbnik, 5 - cięgło próbnika, 6 - siłomierz, 7 - serwonapęd 8 - ramię nośne, 9 - rama podnośnika, 10 - sterowanie serwonapędem, 11 - komputer, 12 - przewody elektryczne

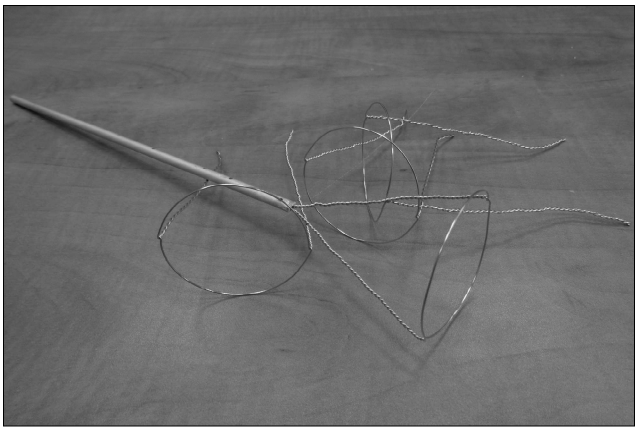


Fig. 3. Probes used in tests

Rys. 3. Próbniki stosowane w badaniach

To connect the probe to a force gauge, a section of the aluminum tube of the probe was used. The internal diameter of the tube allowed sliding movement of the probe. A diagram of this solution is presented in Figure 4.

The test stand was completed with elements listed in Figure 2 as well as the necessary tools and instruments (thermo-elements, scraper buckets, metal moulds serving to pour out the content of the crucible, etc.) Figure 5 shows what the stand in question looks like.

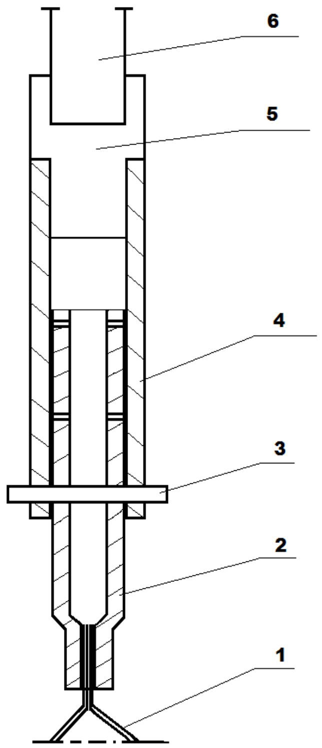


Fig. 4. Probe connected to force gauge: 1 - probe tip, 2 - lower part of rod, 3 - cotter pin, 4 - upper part of rod, 5 - screw nut, 6 - force gauge tip

Rys. 4. Połączenie próbnika z siłomierzem: 1 - końcówki próbnika, 2 - górna część cięgła, 3 - zawleczka, 4 - dolna część cięgła, 5 - nakrętka gwintowana, 6 - końcówka siłomierza

Fig. 5. Test stand view

Rys. 5. Widok stanowiska pomiarowego

COURSE OF TESTS AND EXAMPLE OF RESULTS

The prepared elements of the system in question (aluminum alloy + selected salt mixture) were placed in the crucible heated in order to reach the temperature of ~800°C. After the input had melted, inclusions from the surface of the slag were removed and when the system reached the required temperature, measurements were initiated.

Each measurement consisted in immersing the probe in the metal bath (through the layer of liquid slag), checking the temperature, zeroing the force gauge indications and starting the vertical upwards movement of the probe. Recording of the force values indicated by the force gauge was conducted at intervals of 0.5 s. Breaking the contact of the probe and the boundary of phase division and its total displacement to the slag layer (clearly visible from force gauge indications) cause the probe to stop moving and immerse in the liquid alloy again. The actions specified below (Fig. 6) were repeated cyclically and the system temperature was monitored until salt crystals appeared on the surface of the slag appeared or until the probe was destroyed, which was clearly suggested by the force gauge indications.

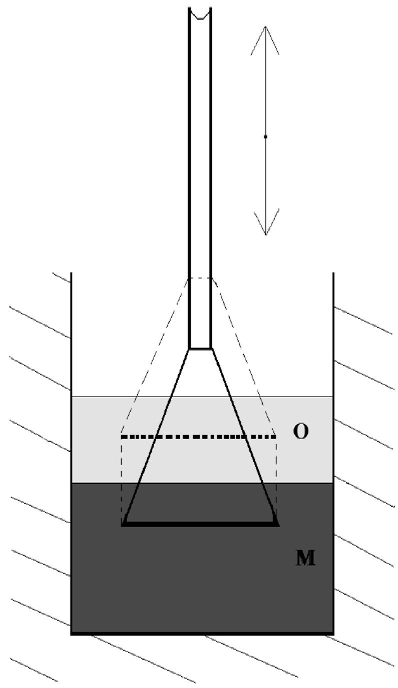


Fig. 6. Probe operation periodicity diagram

Rys. 6. Schemat cykliczności pracy próbnika

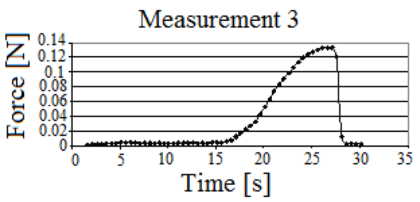
When the early symptoms of slag solidification appeared, the measurements were suspended and the system was heated. When the probe was destroyed, it was then replaced. After the end of the series of trials, the contents of crucible was removed and another series was commenced using fresh inputs of the same composition. The example of changes in the force values registered during a single measurement of one of the tested systems is presented in Table 1. The interphase tension value (in accordance with recommendation 3) in this case was 343 mN/m.

In Figure 7 changes in interphase tension values in one of the tested recycling systems is presented (depending on the temperature). A growth of tension values in relation to an increase in system temperature is observed.

TABLE 1. **Example of changes in force values registered during single measurement of one of the tested systems at temperature of 795°C**

TABELA 1. **Przykład zmian wartości sił zarejestrowanych w trakcie pojedynczego pomiaru jednego z badanych układów w temp. 795°C**

Measurement no. 2 (temp. 795°C)								
Time [s]	Weight	Unit	Time [s]	Weight	Unit	Time [s]	Weight	Unit
1.6	0.001	N	11.6	0.003	N	21.6	0.083	N
2.1	0.001	N	12.1	0.003	N	22.1	0.091	N
2.6	0.002	N	12.6	0.004	N	22.6	0.099	N
3.1	0.002	N	13.1	0.003	N	23.1	0.106	N
3.6	0.002	N	13.6	0.003	N	23.6	0.113	N
4.1	0.003	N	14.1	0.003	N	24.1	0.119	N
4.6	0.003	N	14.6	0.004	N	24.6	0.124	N
5.1	0.004	N	15.1	0.004	N	25.1	0.127	N
5.6	0.004	N	15.6	0.004	N	25.6	0.130	N
6.1	0.004	N	16.1	0.006	N	26.1	0.133	N
6.6	0.004	N	16.6	0.007	N	26.6	0.133	N
7.1	0.003	N	17.1	0.012	N	27.1	0.133	N
7.6	0.003	N	17.6	0.017	N	27.6	0.121	N
8.1	0.003	N	18.1	0.022	N	28.1	0.012	N
8.6	0.003	N	18.6	0.027	N	28.6	0.002	N
9.1	0.003	N	19.1	0.033	N	29.1	0.003	N
9.6	0.003	N	19.6	0.042	N	29.6	0.002	N
10.1	0.003	N	20.1	0.052	N	30.1	0.002	N
10.6	0.003	N	20.6	0.063	N			
11.1	0.003	N	21.1	0.074	N			



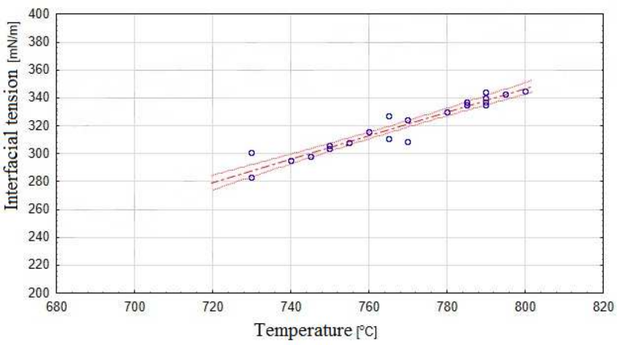


Fig. 7. Changes in interphase tension values depending on temperature in one of the tested recycling systems

Rys. 7. Zmiany wartości napięć międzyfazowych w zależności od temperatury w jednym z badanych układów recyklingowych

CONCLUSIONS

The method of determining interphase tensions in systems composed of liquid aluminum alloys and liquid salt mixtures with the use of the ring method proved to be effective, and the obtained results reliable. Dozens of measurements showed the satisfying repeatability of the results which, as presented in Figure 7, formed a string of values of clearly specified tendency (in each series).

The experiments showed that the dimensions of the probes used as well as the accuracy of changes in the indications of forces registered by the force gauge and the frequency of their registration were found to be sufficient (Table 1).

The drawback of the stand was a limited durability of the probes used. Stainless steel used to make them proved to have poor resistance properties with regard to the operation conditions. One probe provided a dozen measurements at the most, after which, it was destroyed. It is likely that the use of the platinum wire would significantly increase the durability of the probes. The advantage of the stand was the construction which allowed quick replacement of the probes.

What is interesting is the considerably lower values of interphase tensions measured in the first trials with the use of each new probe, which were observed during testing. To explain this, we need to emphasize worse wettability of the probe material (wire) cut from the roll. Once the probe was "etched" during the first contact with the system in question, the values of the measured tensions formed a regular string.

Despite the inconveniences specified above, the method and its implementation performed well while the values of interphase tensions measured in the systems in question proved to be reliable [11].

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