Density of the System Al_2O_3-ZrO_2 in a Liquid Phase

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***Abstract*— Physical properties of materials should be studied in order to describe their behavior for design of new devices or modeling of physical processes. However, it is difficult to determine them for materials at high-temperatures. In this study, a method suitable for determination of density in a liquid phase at high temperature is proposed and tested on Al_2O_3-ZrO_2 system. The material was melted in an induction furnace with a cold crucible. The measurement was based on evaluation of the volume of the melt at different temperatures. The density of material was calculated from the mass and volume of the melt. The densities of material in the solid-state and skull-layer were measured using pycnometer. A temperature dependence of density was established, and the results were compared with literature. The difference between existing data and experimental one was about 10%. Thus, proposed methodology provides reliable density values in extreme conditions. And further it will be used as a fundamental support to increase the amount of experimental data in material properties databases.**

Keywords—density, induction melting, cold crucible, Al_2O_3, ZrO_2

I. INTRODUCTION

The material properties are important in many branches of science and engineering. Nowadays, special methods for the determination of physical properties of materials based on calculation exist [1]. However, it is necessary to realize that material parameters should be determined experimentally, or at least to verify the data obtained using computational methods. Our research aims to develop and verify the method which can be used for the determination of the density of oxides and their mixtures in a liquid phase at high temperature and establishment of its temperature dependence. Several researches studies focused on this issue are available [2] but many materials and mixtures are not described in sufficient range of temperatures or at all. Several mixtures were chosen for verification of the suggested method. The experiment for verification of the method was performed in the wider temperature range than the one in the comparative data. After verification of the method, it will be used for the density determination of materials with unknown dependence of density on temperature.

The density of a material is defined by its mass per unit volume. Thus, the suggested technique has to allow the measurement of the volume of the material and its weight. Investigation of density has been performed in both solid-state at room temperature and liquid phase.

Density measurements were performed in the range of temperatures from 2100 °C to 2400 °C. The lower level of the temperature range was chosen with respect to the liquidus temperature of the mixture [3]. Our aim was to measure the density in liquid phase. Therefore, the melt temperature had to be above the liquidus temperature. The brand-new equipment was developed for the density measurement. It was decided to use lower parameters for the first experiment in this system configuration, thus, 2400 °C was chosen as the upper level of the range.

II. EXPERIMENT AND METHODS

The experiment was designed to measure the density of the mixture 50 % Al_2O_3 and 50 % ZrO_2 in the liquid phase. In order to obtain the liquid phase of the oxide mixture, the induction skull melting (ISM) was used. The principle of ISM consists in melting by a time-varying electromagnetic field in a water-cooled crucible, in a so-called cold crucible. A thin protective layer between the cold crucible and the melt, called skull, is created. It prevents contamination of the melt and reduces thermal stress of the crucible. The density was measured with a temperature step of 100 °C, and its value was calculated from the measurement of the melt volume and its weight. Two quenched samples were taken for X-ray powder diffraction (XRD) to define the exact composition of the melt. The melt was assumed to be homogeneous due to intensive stirring. The temperature of the melt surface was measured using chromatic pyrometer.

The volume was calculated from the height of the melt, which was measured by a special stainless-steel sample. For each measured temperature, 3 samples were taken to calculate the average height of the melt. The sample was immersed into the melt for a short period of time (2 s) and it was colored due to the oxidation processes taking place on the surface. The diameter of the melt was measured by two-step measurements after cooling the ingot. The first step was the measurement of the ingot volume. Then, after the ingot was cut into halves the thickness of the skull layer and diameter of the melted part in different vertical positions were determined. The measurement was made using self-developed software with calibrated scale developed for this measurement. The same measurement was performed using an optical microscope and the results were compared to detect potential errors. However, it was shown that the results are in very good agreement.

The weight of the ingot was further measured. Then, the samples of the skull layer and ingot were taken and their densities in the solid phase were measured. The measurements were done by pycnometry in water [4]. Five measurements for each of 3 samples of the skull layer and 3 samples of the material in the solid-state were carried out, and the average values were used for the calculations. The mass of the skull

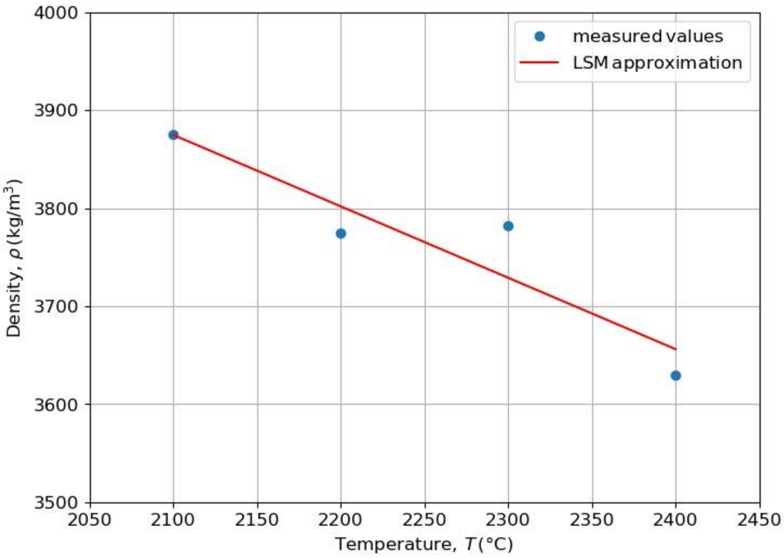

Fig. 1. Temperature dependence of density of 50% Al_2O_3 + 50% ZrO_2

was calculated using the density of the skull and its volume. It was subtracted from the mass of ingot to obtain the mass of the melt.

From the known mass and volume of the melt the density in the liquid phase for each measured temperature was calculated.

The material was melted using an electric current with a frequency of 1.7 MHz in a 4-turns inductor with an inner diameter of 110 mm. The inner diameter of the used cold crucible was 57 mm and the height of the melt was 35 mm. The power transferred into the melt was 8 kW at 2100 °C and 15 kW at 2400 °C.

III. Results

The weights, volumes and calculated values of densities are listed in Table I. The values of the density were approximated using the least-squares method (LSM). The density (kg/m^3) dependence on temperature (°C) of 50% Al_2O_3 + 50% ZrO_2 was approximated by a linear function:

$$\rho = -0.729 \cdot T + 5405.28 \quad (1)$$

The measured results are shown in Fig. 1 together with the dependence of density on temperature according to the linear regression.

IV. Discussion and Conclusion

The result for the temperature of 2100 °C was compared with published value [5]. It was not possible to compare more data because the published research was focused on the densities in the area of the lower temperatures. Our experiment was carried out at 2100 °C to verify the measurement function. For further experiments, a temperature closer to the liquidus temperature will be considered.

The density is equal to 3523.0 kg/m^3 according to [5], the value obtained by our experiment is 3875.17 kg/m^3. The difference 352.17 kg/m^3 is about 10%. Thus, this result is in a good accordance with [5]. In principle, the density should decrease with increasing temperature, so the slope of (1) and the slope presented in [5] were compared.

TABLE I. Weight, volume and density of melt depending on the temperature

T (°C)	m (g)	V (cm^3)	ρ (kg/m^3)
2100	264.97	68.38	3875.17
2200	264.97	70.20	3774.61
2300	264.97	70.06	3782.16
2400	264.97	73.00	3629.71

The difference between them is caused by different temperature ranges. The measurements published in [5] were carried out also in the area of the solidus and liquidus temperatures. In this area, the dependence of density should be less steep than in the liquid phase and this assumption is fulfilled. The density comparison at the same temperature has a higher value for verification of the method. From this point of view, the methodology appears to be reliable. Currently, an experiment to validate and refine the data obtained from the presented one is being prepared.

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