

A Sensor for Continuous Measurements of the Absolute Thermoelectric Power of Liquid Metal During Turbulent Non-Isothermal Mixing or Segregation of Multi-Component Melts

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Abstract

During mixing or processing binary or multi-component melts, the compositional nonuniformity may be expected. We have developed and tested a simple sensor allowing to monitor continuously the local temperature and the gradient of temperature and, at the same time, to measure the momentous value of the absolute thermoelectric power of the melt. Besides finding the value of the absolute thermoelectric power of a melt, which has been mixed to uniform composition, the sensor may be used to detect the nonuniformity of the melt during either mixing or segregation – the variation of the absolute thermoelectric power at the same temperature would indicate the nonuniformity. The sensor may be used also to detect the composition of a binary melt if the absolute thermoelectric power for the binary system is known as a function of temperature and composition.

Introduction

Thermoelectric phenomena may be of considerable importance during processes which involve liquid metals if quite high heat fluxes through the system are involved. Thermoelectricity may be a cause of considerable electrical current under condition that the melt is non-uniform in composition, as during mixing or segregation of binary or multi-component molten metals. Thermoelectric currents would be present also if a uniform liquid metal has a contact with non-isothermal along such an interface electrically conducting walls. These phenomena became actual during 1960's, when thermo-electromagnetic pumps were developed and tested for liquid metal cooling systems of nuclear energy sources in space applications. Also the pioneering work by J. A. Shercliff in a couple of papers showed the relevance of thermoelectric phenomena in metallurgical or crystal growth processes involving magnetic fields. However up to this time the relevant data on absolute thermoelectric power of the molten liquid metals remain quite scarce. We have attempted to develop a simple probe permitting to measure absolute thermoelectric power of a liquid metal even during a turbulent flow of a non-uniform composition melt.

1. Principle of measurement

The principle of the measurements of absolute thermoelectric power S of a liquid metal requires to measure the electromotive force induced by heat flux in the melt. Traditionally it has been done with the melt contained in a dielectric capillary, ends of which were kept at controlled temperature drop. Two electrodes with known $S_{\text{electrodes}}(T)$, where inserted into melt each at one end. The measured potential difference was sufficient to

determine the mean value of the S over the temperature range equal to the temperature drop in the capillary [1, 2]. The local velocity probe [3] developed to measure in a time dependent non-isothermal liquid metal flows used the potential difference measurements to determine simultaneously both melt velocity components and corresponding transverse components of temperature gradient simultaneously on condition that absolute thermoelectric power of the melt S was known. Actually we have just reversed the principle, measuring the S , while the electrical current density (induced by magnetic field interacting with the flow) was negligible.

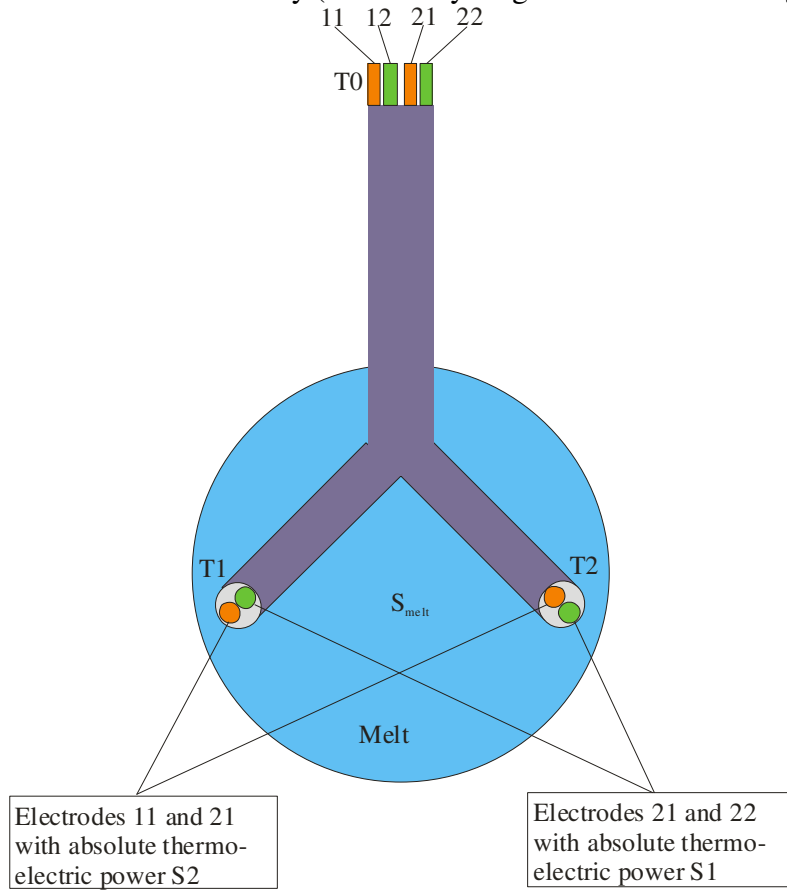


Fig. 1. A schematic of the sensor.

produced by the couple 21-22 represents the temperature drop ($T_2 - T_0$), as for any ordinary thermocouple. Continuous monitoring of voltages U_1 and U_2 at known T_0 is sufficient to obtain the time dependent records of temperature T_1 and T_2 and also the component of a gradient of temperature along the line connecting the tips of both thermocouples.

Simultaneous measurement of a voltage between any of the electrodes including an electrical path between tips at temperature T_1 and T_2 would be sufficient to determine the mean absolute thermoelectric power of the melt $S(T)$ over the temperature range $[T_1, T_2]$. Integrating the Ohm's law, for example, along the path starting and ending at the region with the reference temperature T_0 formed by electrodes and melt 11-melt-21 produces equation:

$$\int_{T_0}^{T_1} S_1(T) dT + S \cdot (T_2 - T_1) + \int_{T_2}^{T_0} S_2(T) dT = U_{11} \quad (1)$$

Assuming that the absolute thermoelectric power of the electrodes has a linear dependence on temperature in the relevant range of temperature

The couples of electrodes 11-12 and 21-22, schematically shown on Fig.1, function as thermocouples without welded joints. Tips of each thermocouple are electrically connected through the liquid metal itself. If the size of those tips is sufficiently small, the temperature there may be considered equal to T_1 and T_2 correspondingly. Therefore the electrical path through the liquid metal, connecting electrodes for each of two couples, does not affect the voltage produced on the electrode connection at reference temperature T_0 ends. There the voltage U_1 produced by the couple 11-21 represents the temperature drop ($T_1 - T_0$) and the voltage U_2

$$S1(T) = S1_0 \cdot (1 + k_1 \cdot T) \quad (2)$$

$$S2(T) = S2_0 \cdot (1 + k_2 \cdot T) \quad (3)$$

the absolute thermoelectric power of the melt averaged over the temperature range [T1, T2] is

$$S(T) = S1_0 \cdot \left[1 + \frac{k_1}{2} \cdot (T1 + T2) \right] + \frac{U11}{T2 - T1} \quad (4)$$

It is obvious that the temperature drop (T2 - T1) should be small, what may be expected if the distance between the electrodes in positions at these temperatures is sufficiently small.

2. Tests of the sensor

The first tests of the sensor were done to determine the absolute thermoelectric power of the binary melt 17Li83Pb, which at this composition forms a eutectic melt. For the electrodes of the sensor we used standard polyimid insulated thermocouple wires Copper and Isotan with diameter 100 μ. Since the producers of the thermocouple wires as a rule give data only about relative thermo-electric power $P = (S1 - S2)$, the handbook data for pure Copper were used to find S_{Isotan} . Fig.2 shows that temperature dependence is not strictly linear and it

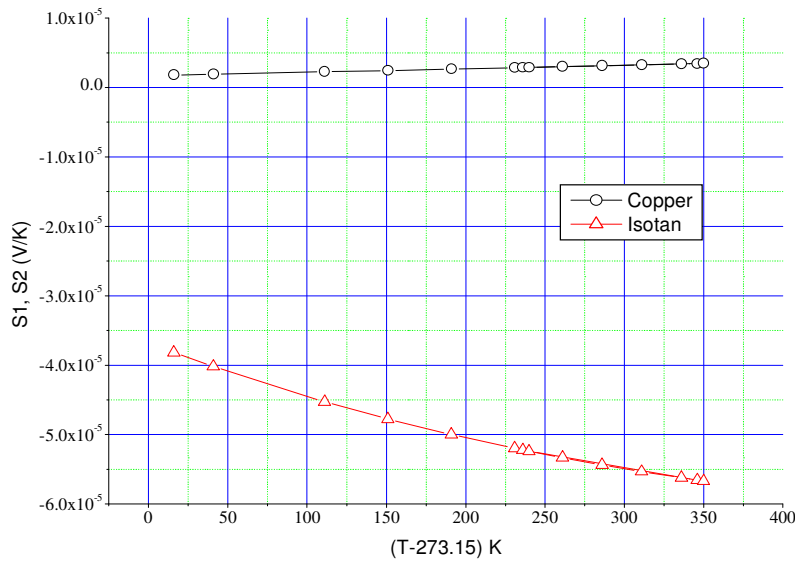


Fig. 2. Absolute thermoelectric power of Copper and Isotan wires as a function of temperature.

would be more suitable to use a polynomial fit instead of linear approximation used to find the expression (4). But, as would be later described, the error analysis shows that other uncertainties affect the accuracy much stronger. A stainless steel capillary with outer diameter 1.2 mm were used as a support, it was coated by a thin layer of electrical insulation to evade the circulation of any thermoelectric currents between the liquid metal and stainless steel. The distance between point T1 and T2 were 12 mm. The relevant potential differences U1, U2 and U11 were measured using thermally stabilized differential amplifiers ensuring zero drift of each not exceeding 0.15 μV over the total period of measurements.

The tests of the sensor were done in a binary melt 17Li83Pb, which at approximately this composition forms a eutectic. A solid block of this alloy was put in an Al₂O₃ crucible with inner diameter 80 mm. The crucible was closed by a lid with a pipe inlet for high purity Argon gas. The outlet of the gas was allowed through a small gap on the perimeter of lid-crucible edge. The air was washed out from the crucible during 1 hour by intensive Argon flow. Then the metal was gradually inductively heated up and melted by 8 kHz magnetic field, produced by a coil surrounding the crucible. When the alloy was completely melted, the quasi-steady state of heat transfer in the setup was ensured cooling the melt surface by intensive Argon flow.

The measurement probe was put into the melt in a position on the axis of the crucible 10 mm beneath the surface of the liquid metal. Since the skin depth of the melt 17Li83Pb at 8 kHz frequency is approximately 6 mm, then the electrodes of the probe were at least 5 skin-depths deep in the melt in radial direction and the induced 8 kHz AC current and AC magnetic field was negligible in the region of the probe vicinity. The time-averaged temperature drop (T₂ - T₁) was zero up to the accuracy of the measurements. Nevertheless the momentous magnitude of the temperature drop reached even up to 2 degrees due to unstable convective

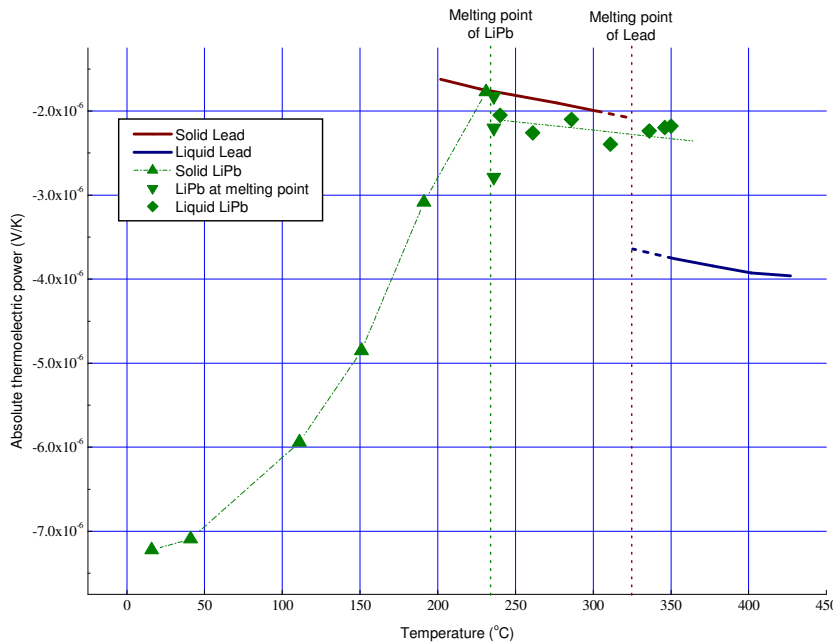


Fig. 3. Measured absolute thermoelectric power of eutectic Li₁₇Pb₈₃. Continuous line shows the experimental data fit for pure Lead [1].

heat transfer produced by AC field. The three relevant voltages were continuously acquired with the sampling rate 30 samples per second, converted to digital format by a separate ADC and saved to a file. The reference temperature was stabilized to (22 ± 0.1) °C. Each experimental point on the Fig. 3 represents the time-averaged value over 5 minutes of measurements, when quasi-steady heat transfer was reached. The measurements were continued also during solidification and melting, and during the cooling of the completely solidified alloy. Solidification was allowed, while mixing action of the AC field was present at reduced coil current and increased cooling by the Argon flow.

The high scatter of the data for absolute thermoelectric power is attributed by us, quite wishfully, to segregation and mixing of Li-Pb melt. Since the Lead is nearly 20 times heavier than Lithium, the production of uniform melt 17Li83Pb requires very specific conditions.

3. Accuracy and limitations of the sensor

Among basic physical limitations of the proposed principle of measurements the most important is the fact that the measurable potential difference U_{11} would inevitably be affected by a presence of the electrical current in the liquid metal and by the presence of gradient of electrical potential induced by the flow interaction with the external DC magnetic field. Electrical current, besides external sources may be caused by thermoelectrical phenomena in nonuniform composition melt, when absolute thermoelectric power $S = S(C,T)$ is a function not only of temperature T , but also of composition of the melt C . During any measurements with the proposed probe the magnitude of both these effects should be carefully estimated.

Other very serious limitation of the sensor follows from thermoelectric phenomena in the electrode wires of the probe itself. Thermoelectric power of a wire at a given temperature is a function not only of the composition of the electrode, but also from mechanical strain and the history of applied tensions, induced mechanically or thermally, in the polycrystalline metal wire. Among those effects only the piezo-thermoelectric effect could be described referring to experimentally determined magnitudes. Piezo-thermoelectric coefficient of Copper wire under longitudinal tension has been measured experimentally – the magnitude was found to be $(3.61 \pm 1.7) \cdot 10^{-17}$ V/(K·Pa) [May]. Threshold of the plastic deformation of unhardened Copper under longitudinal tension is approximately $7 \cdot 10^7$ Pa. For a 100-micron thick Copper wire the threshold of the plastic deformation is reached, if force of 0.55 N is applied. Such magnitude of a force may happen, while producing and handling the probe. Under such magnitude of longitudinal tension the piezo-thermoelectrical effect would produce $2.5 \cdot 10^{-9}$ V/K deviation of the absolute thermoelectric power of the electrode wire (or 0.12 %). Assuming that only one of the electrodes of the velocity probe PMP has been subjected to accidental longitudinal tension of magnitude one tenth of the above mentioned, and the temperature drop between the liquid metal and the cold junction 300 K, the piezo-thermoelectric effect would produce signal 0.75 μ V. The Isotan electrode may not be expected to produce smaller error. If the electrode wires has been subjected to strains leading to plastic deformations, the actual error due to resulting deviation of the thermoelectric power could be detected only experimentally, by testing each electrode, as actually were done during these tests.

Conclusions

The proposed sensor for real time measurements of absolute thermoelectric power in liquid metals was shown to be feasible under real life conditions. However, the conditions under which it may be used should be analyzed for each application, especially, if the temperature of the liquid metal is high.

References

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