Neutron radiography visualization of solid particles in stirring liquid metal

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Abstract

This paper presents the analysis of the first dynamic neutron radiography experiment that visualized motion of solid particles in liquid metal, which was stirred by a system of four counter-rotating magnets. The paper also contains the quantitative results derived from neutron images: the distribution of particle concentration, number of admixed particles and velocities as functions of the magnet rotation speed.

Keywords: neutron radiography, liquid metal, solid particles, permanent magnet stirrer

1. Introduction

Improvement of melt quality is one of the highest priorities in metallurgical processes. The structure and properties of the final product can be radically changed by adding particles to the melt, e.g., admixture of carbon and chromium to iron prevent movement of dislocations and increase tensile strength of the alloy. Due to the non-transparency of metals it is nearly impossible to observe how the solid inclusions behave within the liquid metal. At the same time numerical models, which generally are powerful tools for studying inclusions in melts, usually

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contain a high number of degrees of freedom due to the complexity of turbulent flows of liquid metal. Therefore, experimental verification of the numerical models is an essential task, which was not satisfactorily solved till nowadays. Furthermore, the development of admixing technology requires better understanding of the structure of the flow, the velocities of the flow and peculiarities of the particle behavior.

This paper presents a new experimental approach for the visualization of solid inclusions in liquid metal – the neutron radiography method. A report on the first experiment, which was recently performed at the Paul Scherrer Institute (PSI), was recently published in [1]. That report contains only raw neutron images and has the main goal to demonstrate to a liquid metal society the general advantages of the experimental method. Now, quantitative results, which are derived from the neutron images, will be presented in this paper.

The aim of the experiment was to visualize the motion of solid particles in electromagnetically induced liquid gallium flow. Such motion is typical for induction furnaces, stirrers and other metallurgical applications. A better view of admixing and homogenization of solid inclusions would enable further improvements of industrial process.

Until now this has been studied only by numerical simulations, as suitable methods for experimental visualization of particle movement in opaque melts have not been available. There are a lot of precise optical methods for investigation of multi-phase flows like the particle image velocimetry or the particle tracking velocimetry with different modifications, but they all are not suitable for non-transparent methods. Moreover, the electromagnetically induced recirculated flows, which are typical for metallurgical equipment, form separate class of turbulent flows due to the specific structure of vortices and their relative oscillations, which cannot be reproduced in any other transparent non- or pure-conductive liquid [2]. Furthermore, the nuclear magnetic resonance investigation is not applicable here due to the strong magnetic field. Finally, the classical X-ray radiography in heavy metals is significantly limited due to attenuation coefficient proportional to an atomic number and is likely impossible for turbulent liquid metal flows. Nevertheless, recently Šeepanskis et al. [2] managed to measure a particle concentration field in a small induction crucible furnace using the original technique. It was possible to acquire results only for the dynamical equilibrium stage – the establishment of the flow and the structure couldn't be observed.

Taking into account all mentioned difficulties, the neutron radiography was chosen as a promising method for the electromagnetically induced liquid metal flows. The first known neutron radiography of a two-phase liquid metal flow was performed by Saito et al. in 2004 [3]. Rising gas bubbles were visualized in the Pb-Bi eutectic. However, the flow was initiated only by the rising bubbles without electromagnetic stirring; therefore, intensity of the flow was low. Thus, the present experiment can be considered as a significant step forward.

A cylindrical vessel stirred by a solenoidal inductor is often used in metallurgy (see, e.g., investigation on the flow of an induction crucible furnace [4,5]). An alternating magnetic field and an induced current in the melt result in the Lorentz force forming counter-rotating vortices. However such devices cannot be used in the neutron radiography since the water-cooled inductor would be in the way of the neutron beam. Similar flow patterns as created by the inductor can be produced with a permanent magnet system. Usage of such systems for stirring and transporting liquid metals in metallurgical applications is slowly increasing because of the fact they significantly improve efficiency [6] and the recent decrease of the cost of such systems due to the reduction of permanent magnet prices. Thereby such permanent magnets system would have two benefits: 1) it would be possible to achieve industrially relevant flow without shadowing a neutron beam with details of the set-up, and 2) enabling investigation of the quality of liquid metal stirring by the permanent magnet system.

2. Experiment

The stirrer is shown in Fig. 1. A rectangular vessel for liquid metal is made out of window glass, which contains silica ($SiO_2$) 72%, sodium oxide ($Na_2O$) 14.2%, lime (CaO) 10.0%, magnesia (MgO) 2.5%, alumina (Al$_2$O$_3$) 0.6%; all these materials have low attenuation coefficients for thermal neutrons (see [7]). The stirrer is constructed in such a way that nothing is situated in the line of the neutron beam, which only has to pass a 2 mm glass two times.
Fig. 1. (a) experimental set-up; (b) cylindrical permanent magnets and the container for liquid metal, the arrows indicate the rotation direction.

The inner size of the vessel is 100 x 100 x 30 mm. The cylindrical magnets are 30 mm in diameter and 50 mm long; the distance between the centres of the closest magnets is 50 mm; the distance between the edge of the magnets and the liquid metal is 9 mm. Due to very strong magnetic interaction between the close magnets they are designed to be counter-rotating like it is shown in Fig. 1 (b). The vessel is centred with respect to the magnets in the direction of the neutron beam. Qualitative images of the flow, which is induced by the system, are shown in Fig. 2. Characteristics of the flow were obtained by numerical simulation.

Fig. 2. Qualitative characteristic of the flow induced by the four counter-rotating magnets: (a) averaged flow velocities; (b) averaged kinetic energy (visualized are especially the regions of intensive flow pulsations).

In order to choose a proper metal for the experiment two properties should be considered: 1) low melting temperature to avoid heating close to a detector and a camera that should be situated as close to the vessel as possible; 2) the attenuation of the neutrons (see [7]). It becomes clear that low temperature eutectics, which contain cadmium (Cd) and indium (In), such as Wood’s metal and Galinstan, cannot be used due to high values of the attenuation coefficients. Bismuth (Bi) is also unwanted because it can be activated in the neutron beam and tiny amounts of polonium (Po) can appear. For these reasons we chose gallium (Ga), which is acceptable for neutron
radiography and has a suitable melting temperature ($30 \, ^\circ\text{C}$).

The material properties of the inclusions used for the neutron radiography experiment have to fulfil various requirements. The attenuation coefficient of the particles cannot be in the same range as that of the liquid metal otherwise they will be invisible. Therefore, the typical SiO$_2$, ZrO$_2$ and Al$_2$O$_3$ oxides are useless here. Boron carbide (B$_4$C), which has a very high attenuation coefficient, was found to be good for the experiment. However, due to its low density (2.52 g/cm$^3$) and the very low size of available particles, it was a significant problem to mix such a powder from the surface into the metal. Therefore, special particles, which consisted of a lead (Pb) kernel, which is a heavy material, and boron carbide powder glued around the kernel, were fabricated. The diameter of the particles was $3.8 \pm 0.5$ mm, and average density was a little bit less than that of gallium (6.1 g/cm$^3$) so that they would remain on the top surface without stirring.

The experiment was carried out on 25$^{\text{th}}$-27$^{\text{th}}$ of July, 2014 at the Swiss Spallation Neutron Source (SINQ) in Paul Scherrer Institute (Villigen, Switzerland) in the thermal neutron transmission radiography facility (NEUTRA). Neutron energy at this facility is about 25 meV; the neutron flux depends on the proton current delivered to SINQ and is about $5 \times 10^6$ neutrons cm$^{-2}$ sec$^{-1}$ mA$^{-1}$ (p-current) at the sample position. An Andor Neo sCMOS pixel detector was used at NEUTRA. For more information about NEUTRA facility we recommend the reference [8] or http://www.psi.ch/sinq/neutra.

The frame rate of the camera should be pointed out additionally. Despite the camera itself allowed high frame rate (up to 32 fps), directly writing individual frames to the computer hard disk limited the real frame rate to approximately 1 fps. This fact was recognized as one of the obstacles to obtain more industrially relevant results during that campaign. Wide ranges of exposure times were available; 0.1 s and 0.01 s were used during the present experiment.

3. Results and discussion

The results of measurements were obtained as neutron shadow images; some of them can be seen in Fig. 3. Two different exposure times were used – 0.01s and 0.1s: shorter exposure makes it possible to see separate particles in the flow and was used to create a particle concentration field, but longer exposure shows the trajectories of particles and, consequently, visualizes the structure of the flow as well as allows to calculate the velocities of the inclusions. As it was already discussed, the camera system supports 1 fps, but the velocity and oscillations of the flow was so high that the frame rate was the limiting factor for using particle image velocimetry (PIV) technique and software for the analysis, i.e. it wasn't possible to unambiguously follow the same particle in two consecutive frames.

Nevertheless, by doing time consuming image processing work it was possible to evaluate admixing of particles in the fluid and the average velocities of the particles in the flow. The image processing program ImageJ was used for the analysis.

To find the concentration every image was overlaid with a reference grid, and every particle was counted in every cell of the grid. By averaging the obtained results it was possible to create a concentration plot, which can be seen in Fig. 4. The maxima of the particle concentration correlate with flow vortices (see Fig. 2(a)). Higher concentration in the upper part of the vessel can be explained taking into account the lower density of the inclusions in comparison to the liquid. From these results it is possible to acquire concentration as a function of magnet revolutions which is shown in Fig. 5 (a). A couple of comments should go with these results. Firstly, the exposure time was low (0.01 s) to avoid blurring of the particles due to their motion lead to significant noise as can be seen in the raw images (Fig. 3 (b)). Noise created an additional challenge for finding all admixed particles in the volume. Secondly, the particles were not equal in their sizes (the average diameter of the particles was $3.8 \pm 0.5$ mm, i.e. 13% error) and shapes, so possibly different behavior for each particle in the flow should be taken into account. The experimental results show a self-evident relation – higher frequencies causes admixing of more particles. However, the noticeable step structure could not be explained without full numerical simulation of the process.
An analytical expression for a magnetic field and an iterative solution for the flow velocity in the limits of a two-dimensional model as well as experimental measurements of the velocity in a liquid metal stirrer based on rotating permanent magnets have been presented by Bojarevics & Beinerts [9]. This analytical method is used in this work for estimation of the flow velocity in the present set-up. The average particle velocity in the liquid metal as a function of magnet rotation speed is shown in Fig. 5 (b). The experimental curve in this figure was obtained by using images with longer exposure time (Fig. 3 (a)), which allows to see particle paths. The velocity of particles was calculated by dividing the length of its path with the exposure time (0.1 s). The results are presented in Fig. 5 (b) as a function of magnet revolution speed. Comparing analytical estimation of the flow with the experimental results, it must be noted that the velocities found analytically are definitely higher than that of particles in the experiment even taking into account the huge errors of the experimental results. This fact seems to be easily explained by the inertial effect due to the big size of the inclusions.
Finally, significant deformations of the free surface of the melt were observed next to the rotating magnets during the experiments with a decreased level of liquid (Fig. 3 (c)). These deformations appear due to high velocity next to the magnets (see Fig. 2 (a)), which throw out the liquid to be compensated by a hydrostatic pressure and a surface tension. This could be considered as different types of flow that could be investigated depending on frequencies, but this task was beyond time limits for this experiment. However, it is still possible to draw some conclusions. Firstly, as it is seen in Fig. 3 (c), the reduced level of fluid decreases the amount of particles that are admixed. The most part of the particles remains stable between the surface deformations (“hills”). Secondly, decreasing the level of the fluid increases the surface deformations. The measured heights of the “hills” for magnet revolutions of 28.2 rps and different fill levels of the vessel, i.e. height of the free surface, are presented in Tab. 1.

Table 1. Velocity of the liquid metal next to the magnets in the cases of reduced fill of the container.

| Fill   | height of the “hill”, cm | Standard deviation, cm |
|--------|--------------------------|------------------------|
| 80%    | 0.63                     | 0.12                   |
| 60%    | 1.63                     | 0.18                   |
| 40%    | 2.08                     | 0.13                   |

4. Conclusions

First of all, we are able to conclude that dynamic neutron radiography is a suitable method for the investigation of flow and behaviour of inclusions in liquid metal, which is of industrial interest. Despite technical difficulties and the low frame rate available in the here reported first measurement campaign, significant quantitative results were obtained additionally to the neutron images: the distribution of particle concentration, number of admixed particles and velocities as function of the magnet rotation speed.
4. Further development

Further plans are aimed to establish the neutron particle image velocimetry (NeuPIV) method, which can be very useful for the metallurgical industry. For this goal, a next experiment is planned for the next year. A high speed camera (up to 32 fps) is available at NEUTRA already. The present team of scientists is going to prepare better particles (smaller and more uniform) for the second experiment as well. Finally, some more advanced filters will be applied during image postprocessing to reduce the magnitude of noise in the images.

The analysis of a full numerical model of the stirring process, which has been developed by the same team of the authors, is currently in progress.

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