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Stabilisation of aluminium foams and films by the joint action of dispersed particles and oxide films

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ABSTRACT

Aluminium alloy foams are created by injecting gas containing different levels of oxygen (from ≪1 ppm to 21%) into melts stabilised with SiC or TiB_2 particles. Individual liquid aluminium alloy films meant to represent the films in a foam are produced of the same materials. For foams and films, the oxygen concentration of the atmosphere is controlled. Synchrotron X-ray radioscopy on liquid films is applied to track the movements of the particles within and to observe how they flow, pile up and form clusters. Experiments on aluminium foams show that only when the injected gas and the surrounding atmosphere contain oxygen foams can be expanded continuously. In contrast, if foaming is carried out by injecting argon into the melt and the Ar atmosphere is free of oxygen no stable foams can be created, even if the melt contains 20 vol.% SiC particles. Both film and foam surfaces are analysed ex-situ by energy-filtered TEM and SEM. It is found that oxide layers form, cover the particles and push them into the metal. A high oxygen content in combination with Mg in the alloy promotes this process. It is concluded that not only particles are required to allow for foaming, but also the formation of an oxide skin is necessary and the combination of both are the basis of film and foam stabilisation.

1. Introduction

The stabilisation of liquid metal foams is still only poorly understood for any of the different types of foaming processes based either on metal powders or on molten alloys [1–4]. Liquid metal foams are transient and prone to drainage, coarsening and coalescence. An investigation of the stabilising mechanisms in such liquid foams is necessary to be able to improve foaming processes and the resulting solid foam properties such as the pore size distribution and uniformity of density, and finally their mechanical parameters.

Up to now, most of the efforts to understand foam stability are based on the study of entire foams, more specifically how they expand, what the reasons for destabilisation are and how the decay of foams can be prevented [3,5,6]. The present authors have suggested investigating liquid foam stability by studying the simplified model system of single metal films instead [7,8]. In aqueous systems, the comparability of the model system 'single film' and real foams has been substantiated [9,10]. This allows one to apply a wide range of experimental methods to investigate specific effects more precisely as it would be possible for entire foams, e.g. particle movements in the liquid or a defined oxygen concentration and oxidation time.

For aluminium alloys, a certain amount of solid particles (for example in AlSi9Mg0.6 either 20 vol.% SiC or 6 vol.% TiB_2) is required to stabilise a liquid film [11], which agrees with what is known for foams created by gas injection, where particles have to be present as well [12,13]. It comes as a new finding that also a minimum amount of oxygen, namely >200 ppm is required [8]. This suggests that oxygen is not only important for the creation of metal foams as already known but essential.

Hitherto, the stability of aluminium foams has been investigated by analysing solidified foams by microscopy and studying the morphology and the arrangement of solid particles inside their cell walls [14,15]. In addition, liquid foams have also been studied in situ by means of X-ray radioscopy with an emphasis on foam expansion and decay (characterised by the coalescence rate) [16,17]. In the present work, we combine these two approaches and demonstrate for the first time how particles behave and arrange inside an individual liquid aluminium film by using in situ synchrotron X-ray radioscopy [18]. We foam different alloys

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by injecting gas containing various oxygen levels and select different surrounding atmospheres. The impact of oxygen on film stability is also investigated using energy-filtered transmission electron microscopy (EFTEM) on samples taken from the solidified inner bubble surface.

2. Experimental

2.1. Materials

Aluminium alloys containing various elements and stabilising particles are used for studying liquid film and foam stability. Three particle-free alloys are prepared: pure Al, AlSi9 and AlSi9Mg0.6. The latter is made by adding 0.6 wt.% Mg to an as-received commercial AlSi9 alloy. Three composites, each containing 6 vol.% TiB_2, are made via flux-assisted synthesis by an in situ reaction, yielding particles of 0.5–3 μm mean diameter [19]. Moreover, a composite of AlSi9Mg0.6 containing 20 vol.% SiC particles with a mean particle size of 10 μm (F3S20S made by Alcan, Montréal, Canada) is used. Particle-free AlSi9Mg0.6 melt is used for diluting this composite to the second desired concentration of 10 vol.% SiC particles. Hence, in total 5 different composites are investigated.

2.2. Films

Single metallic films (SF) are produced in a controlled atmosphere by pulling a circular frame made of molybdenum wire out of a melt and solidifying the film created within 10 s. The oxygen content of the surrounding atmosphere is adjusted in the range from $\ll$1 ppm to 21% O_2 to study its influence on film stability. The setup is sketched in Fig. 1(a). A description of the film pulling process can be found elsewhere [8]. The shape of the wire frame and the structure of the corresponding film are further modified to more complex quadruple parallel frames (QPF). These allow us to produce films that resemble the Plateau borders of foams and to keep films liquid for up to 120 s [11]. The pulling velocity of 10 mm/s and the melt temperature of 680 °C are kept constant for both model systems.

2.3. Foams

In order to evaluate whether metal films represent the phenomena occurring in foams, corresponding metal foams are produced via gas injection. The gas injection furnace used contains two vertically arranged insulated heating plates, each with 1200 W power, enough to heat up the melt to 680 °C, see Fig. 1(b). The temperature is recorded by a thermocouple and maintained constant using a power controller. Perpendicular to the heating system, two X-ray transparent boron nitride plates close the furnace. This allows us to study foaming in situ via a microfocus X-ray source and a flat panel detector [20]. The X-ray spot size on the target is 5 μm.

In the past, mostly static cannulas have been used to inject gas into melts and to convert them into foam. However, such cannulas may be disadvantageous. For example, an overpressure is required to break the oxide skin at the beginning of injection and the oxide filaments blown off might remain in the liquid. Therefore, a moveable cannula system powered by a stepper motor is used that allows one to insert a cannula from the bottom into the melt and to avoid coverage with an oxide film. The gas injection pressure is adjusted by a needle valve and recorded by a sensor. To investigate the influence of the oxygen level of the surrounding atmosphere the furnace is kept inside a gas-tight stainless steel chamber. The oxygen contents of both the injected gas and the atmosphere in the chamber are controlled individually by an "Oxygen Service Controller II" of Prozess-Informatik GmbH, Germany. Before starting the foaming experiments, the chamber is evacuated and purged twice with Ar at a flow rate of 100 l/h. Thereby moisture is removed and the same low oxygen content as for the model system ($\ll$1 ppm O_2) is achieved. All experiments are performed at 680 °C at 200–300 mbar gas injection over-pressure and using a conical injection cannula [21]. Varied composites, different oxygen contents and injected gases are applied. Foam expansion as well as the coalescence of bubbles are recorded by the software "HiPic 7.1" and analysed with the software "ImageJ" [22].

2.4. Microstructural analysis

For a microstructural analysis of the particle arrangement at the solid–gas interface of films and foams, a Zeiss CrossBeam 1540 EsB© workstation is used. It comprises a scanning electron microscope (SEM) with an ultra-high-resolution GEMINI© field emission column and two secondary electron detectors, namely an in-lens detector perpendicular to the surface and a lateral assembly SE2 detector (both detect secondary electrons). Moreover, it contains a focused ion beam (FIB) unit, which is used to cut fine lamellae out of the sample surface for the characterisation of the oxide layer by transmission electron microscopy (TEM). The SE2 detector is used for investigating the surface topology, the in-lens detector to observe the milling process and for higher Z-contrast imaging. A 1-μm thick carbon layer is deposited on the surface to protect the oxide layer during milling. Milling is performed with a 30 keV gallium ion beam with a current decreasing in steps from 10 nA and 2 nA to 200 pA to prepare a 10-μm long lamella and to reduce its thickness to <1 μm. After this, the lamella is bonded to a copper grid to further reduce its thickness to <100 nm employing a current of 50 pA and 20 pA. Eventually, the lamella becomes electron transparent, which allows for investigating the oxide layer using energy-filtered TEM (EFTEM) in a Zeiss LIBRA 200 microscope. The TEM operates at 200 kV and is equipped with a field emission gun and a high-resolution in-column energy filter. In the electron energy loss spectroscopy (EELS) mode this system yields an energy resolution of <0.7 eV. To visualise the oxides at the solid–gas interface, two images are acquired at 510 and 530 eV, which is below the oxygen peak edge at 542 eV. From this image another one taken at 552 eV is subtracted using the imaging software "DigitalMicrograph" [23]. To achieve a high-contrast oxygen map, 15 images with each 10 s exposure time are summed up. For a better evaluation, a further bright-field image of the same area is recorded and overlaid with the intense oxygen image using the software "ImageJ".

2.5. Synchrotron radioscopy and tomography

In situ visualisation of particles in liquid films is carried out by hard X-ray radioscopy using high-brilliance synchrotron radiation at beamline ID 19 of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France [24]. Imaging with polychromatic radiation is performed in the phase contrast mode with a sample-detector-distance of 1 m and an mean X-ray energy of around 19 keV, thus allowing for the separation of materials of similar electron density, in our case the aluminium alloy and the ceramic particles. The X-ray beam is converted to visible light using a LuAG:Ce single crystal scintillating screen (Ce-doped $Lu_3Al_5O_{12}$). The image is projected onto a "pco.dimax S" CMOS camera through a folded optical system to avoid any damage of the detector. The camera has 2.7 μm pixel size and can cope with frame rates up to 12 fps at full resolution [25]. For our purpose, the gap of the U17.6 single-harmonic undulator is set to 14.5 mm. The field of view is covered by 2016 × 2015 pixels.