Journal of Manufacturing Processes 32 (2018) 77–84

Contents lists available at ScienceDirect

Journal of Manufacturing Processes

journal homepage: www.elsevier.com/locate/manpro

Thermodynamic analysis of the aluminum alloy foaming process by melt route

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ARTICLE INFO

Article history:
Received 14 February 2017
Received in revised form 21 January 2018
Accepted 29 January 2018

Keywords:
Metallic foams
Aluminum alloy
Compounds
Thermodynamic analysis
Foaming agent

ABSTRACT

A thermodynamic analysis was carried out to determine the stability compounds formed by the interaction between the molten alloy (A356 alloy) with a foaming agent (1, 2, 3 wt. % $CaCO_3$) and a thickening agent (1 wt. % Al_2O_3) for the production of closed-cell aluminum alloy foams. Stability phase diagrams were obtained to 973, 1073 and 1173 K and they showed the formation of the compounds $MgAl_2O_4$, $CaAl_4O_7$, Al_4C_3, and Al_4O_4C. Typical closed-cell foams of the A356 aluminum alloy were produced to the same conditions of the thermodynamic analysis. The structure of the foams produced was evaluated by SEM-EDS and Raman techniques. The stability compounds predicted are in good agreement with those determined experimentally. The compounds formed by the interaction between the particles and the melt were increased with the increase of the foaming agent and were located at the cell walls.

1. Introduction

Methods for producing metal foams are already known and classified [1,2]. Molten metal can be processed to a porous material by interacting gas into the melt [3] or by adding a foaming agent into it [4]. Most of the closed-cell aluminum foams are produced by adding a foaming agent to the molten aluminum together with stirring. TiH_2 is a popular foaming agent because of its decomposition temperature, which is close to the melting temperature of aluminum alloys. However, a particular concern is the control of pore size, porosity level, the homogeneity of the foam and high production costs. In order to meet the above requirements, a new easily available agent for metal foaming, calcium carbonate was proposed as a cheaper and safer foaming agent than the conventional agent, titanium hydride [1–6]. Lázaro et al. [7] produced aluminum alloy foams by using an alternative carbonate constituted by a mixture of magnesite and dolomite. The carbonate mixture was added from 0.7 to 5 wt. % and mixing time varied from 1 to 3 min, while stirring at 600 rpm. The foaming agent is trapped and decomposes into a gas and then dissolves into the liquid, which causes the melt to foam. Calcium carbonate gives a finer cell structure than TiH_2 in the melt route. CO_2 gas is readily available from the decomposition of carbonates, which oxidizes and stabilizes the aluminum cell surface. Thus, the aluminum foam produced using carbonates has a finer cell structure than that produced using TiH_2 [8]. In general, CO_2 is the best gas to be trapped in the pores, since it is easily obtainable, has a low thermal conductivity and low toxicity [9]. The foaming process is rather complicated because the foam formation is governed by a complex interplay between cells and the solid-liquid interface so that an unstable liquid film easily leads to bubble collapse and an imperfect foam structure is obtained. To obtain homogeneous pore distribution it is necessary to avoid drainage of aluminum melt, coarsening and rupture of cell walls during solidification, which leads to the generation of coarsened cells. Thickening agents, such as silicon carbide, aluminum oxide or other ceramic particles can be used to carry out the aluminum foaming process [10]. It was found that the foam stability can be increased by using Al_2O_3 instead of SiC, because of increased cell wall thickness [11]. An alternative way of foam generation involves adding calcium to an aluminum alloy and stirring in the presence of air which leads to an in situ creation of solid particles in the melt that increase the viscosity of the melt and suppress the drainage of aluminum melt during solidification [12].

Yang and Nakae [13] added aluminum powder to an aluminum alloy melt and found an increase in viscosity and foamability. Recently, it was reported that magnesium, which oxidizes easily, works as a thickening agent for aluminum alloys containing magnesium in its chemical composition [14]. Liquid metals, especially

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https://doi.org/10.1016/j.jmapro.2018.01.028
1526-6125/

aluminum form a strong, compact and thick oxide layer on the surface in an oxidizing atmosphere. Oxidizing gases and those alloying elements that increase the strength of the oxide film may increase the stability of aluminum foam melt. The accumulation of oxide particles in regions close to the surface of films increases the surface viscosity promoting slowdown drainage. Reactions between the stabilizing particles and the melt could be observed in some cases. A micrograph of an Alporas foam shows by an EDX analysis, the presence of precipitates which contain various mixed oxides of aluminum, calcium, and titanium such as Al_2CaO_4 or $Al_2Ca_3O_6$, or oxide mixes $Al_2O_3 + TiO_2$ or intermetallic compounds such as Al_4Ca, Al_2Ca or Al_3Ti [15,16]. Byakova et al. [17] studied the role of foaming agent and processing route in the formation of by products which contaminate the cell wall and affect the macroscopic mechanical response of closed-cell aluminum foams. For aluminum foams processed with calcium additive, coarse particles of the Al_2CaSi_2 intermetallic compound were observed in the cell wall. These particles reduced the compressive strength to values close or below to those of open-cell foams of the same relative density. It is evident from the literature review [1–17] that the foaming process via melt route involves reactions between the foaming and thickening agents and the melt. The products obtained from these reactions forms clustered structure which can cause faster sedimentation of the particles on the foam surface or in the metallic melts, affecting the cell wall structure and the mechanical properties of the metal foams. It has been reported that Mg in the liquid aluminum can react with Al_2O_3 producing spinel or MgO layer on its surface [18]. Babcsán [19] carried out a thermodynamic analysis to determine the minimum magnesium concentration required to form the compound $MgAl_2O_4$ in Al-10Si alloy at 1000 K. There are few research works [18,19] related to the prediction of compound formation in the manufacturing of aluminum alloy foams by melt route and its effect on the foam structure. In addition, depending on the foaming process parameters, the components can react with the melt to form different compounds that will affect the foam properties. In the present work, a thermodynamic analysis was carried out to determine the compounds formed by the interaction between the A356 aluminum alloy and the thickening (Al_2O_3) and foaming agent ($CaCO_3$) with the thermodynamic software Factsage 7.0 [20]. Stability phase diagrams were obtained at 973, 1073 and 1173 K in order to explain the foaming process and the compounds formation. The predicted compounds were compared with those obtained by the manufacture of A356 aluminum alloy foams by SEM-EDS and Raman analysis.

2. Experimental procedure

2.1. Fabrication of A356 aluminum alloy foams

A master A356 alloy was manufactured by conventional melting in a gas furnace at 1023 K from pure metals. The following chemical composition was obtained by Atomic Emission Spectrometry for the A356 aluminum alloy (92.3 wt% Al, 7.12 wt% Si, 0.38 wt% Mg, 0.2 wt % Cu). 500 g of the master alloy was set in a stainless steel crucible. The alloy was melted and kept at 1023 K in an electric furnace under atmospheric pressure. The heating system was an electrical furnace enabled with control of temperature to within ± 10 K of the set values. The temperature was measured with a K-type thermocouple. The experiments were carried out using a stir-caster system with a stainless steel paddle axle. The viscosity of the melt was modified by adding 1 wt % of Al_2O_3 (0.3 μm, 99%) of the mass charge at a constant stirring speed of 1600 rpm for 2 min. $CaCO_3$ (14 μm, 98.5%) was added as a foaming agent in amounts of 1, 2 and 3 wt % of the mass charge into the melt at a stirring speed of 1600 rpm for 100 s. After the foaming agent addition, the melt was kept in the furnace at the holding temperature of 1023 K for 2 min to allow the foam formation. The cooling procedure was carried out as soon as the expansion process took place. The crucible containing the melt was removed from the furnace and the crucible was cooled by sprayed water.

2.2. Foam characterization

The fabricated A356 aluminum foams were cut on the cross section in order to obtain samples to evaluate density and cell structure. The density and relative density of the aluminum foams were measured by weighing a sample of known volume. The cell structure was observed by optical microscopy and the image analyzer Carnoy. The microstructure examination of Al-foams and a qualitative chemical analysis of the particles formed during foaming were determined with the SEM Jeol 6300 and with the energy dispersive spectra (EDS) analysis. Images were obtained to different magnifications with backscattering electrons with 15 kV and 10 A. Room temperature Raman analysis was performed by employing a Horiba LabRAM HR Evolution spectrometer equipped with a confocal microscope. A He-Ne (632.8 nm) laser was used to excite the samples; the spot size was fixed at 20 micrometers. Spectra were recorded during 20 s with five accumulated scans.

2.3. Thermodynamic analysis

The thermodynamic analysis was carried out with the Equilib module contained in the software Factsage 7.0 [20]. Equilib was used to determine the concentration of the different chemical species once they reach the chemical equilibrium state. The user gives the initial amount of chemical species, the temperature and the pressure of the system (usually 1 atm), then the program calculates the most stable species with the Gibbs free energy minimization method. The thermodynamic analysis was carried out considering the following system: $[Al\text{-}Mg]_{alloy}$-$(aAl_2O_3)_{thickening\ agent}$-$(bCaCO_3)_{foaming\ agent}$. Where a and b values were set in the range from 0 to 1 wt% and 0 to 3 wt%, respectively.

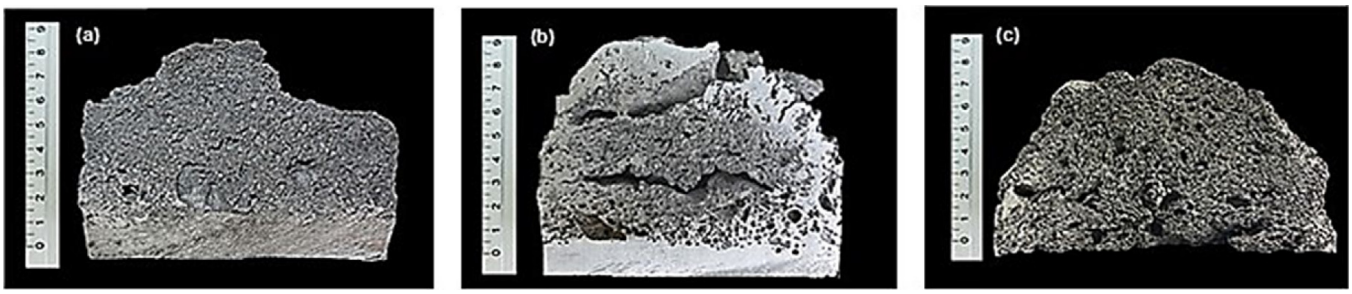

Fig. 1. Structure of the A356 aluminum alloy foams manufactured to the foaming additions of a) 1 wt% $CaCO_3$, b) 2 wt% $CaCO_3$ and c) 3 wt% $CaCO_3$ using Al_2O_3 as a foaming agent.