UNCONVENTIONAL HEATING TECHNIQUE FOR PREPARING CLOSED-PORE CELLULAR ALUMINUM

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***ABSTRACT**: Closed-pore cellular aluminum was experimentally made using recycled commercial post-consumer aluminum containers as an extremely fine powder (below 32 µm) and calcium carbonate (CaCO3) as a foaming agent, replacing the much more expensive titanium hydride (TiH2) frequently used. The aluminum powder was processed by the own original method of microwave melting and atomizing by contact with nitrogen jets. The mixture foaming was performed by irradiation heating in microwave field at 750 ºC. The main characteristics of aluminum foam specimens (apparent density in the range of 0.88-1.10 g·cm-3, heat conductivity between 3.30-5.65 W·m-1·K-1, compression strength within the limits 3.9-7.4 MPa, and homogeneous microstructure with cell size below 0.45 mm) were almost similar in terms of quality with products made with TiH2.*

***KEYWORDS**: closed-pore cellular aluminum, recycled aluminum waste, calcium carbonate, microwave heating, energy efficiency.*

1. INTRODUCTION

Aluminum foam is an isotropic material with cellular structure, whose physical and mechanical characteristics are excellent under conditions of very low density values compared to the compact metal. It is very rigid, insulating, and fireproof, much more stable than wood or plastic as organic materials, and it is completely recyclable. The cellular aluminum is effective in terms of electromagnetic shielding and structural damping (Closed cell, 2022).

There is a fundamental difference between the production technique of aluminum foam with open cells and that with closed cells. Open-cell foam is based on the use of a foamed precursor (usually a polymer) as raw material. Through an electrochemical process, the metal is deposited on the polymer foam precursor. Open-cell foam is characterized by the complete communication between the cells. Thus, the material is a very good acoustic insulator, impact absorbent, catalyst support, gas or metal filter, architectural applications. Closed-cell foam involves the existence of a foaming agent, which releases a bubble-forming gas inside the powder mixture of metal and agent. The material structure contains completely closed-cells separated from each other by metal walls, ensuring a superior mechanical resistance (Farhadi et al., 2020).

The worldwide interest for applying especially the closed-cell aluminum foam in aerospace and car industries is growing. Aluminum foam is attractive in these domains where absorption of impact energy and vibration as well as decreasing the weight are essentially (Garai, 2020; Banhart, 2000; Mahadev et al., 2018; Tatt et al., 2021).

Aluminum foam was produced for the first time more than five decades ago, but its promotion and application are still at a relatively low level. According to (Aluminum foam, 2021), the main reason is the poor reproducibility of the structure and properties of the foams.

Aluminum foam can be produced by several techniques such as powder metallurgy, sintering, addition of a gas into molten metal, the use of expanding agent in raw material, casting method, etc. (Mahadev et al., 2018; Karuppasamy & Barik, 2021). Gas streams or blowing agents can be used to form the foam. The method is based on the thermal decomposition of the agent, sometimes in the presence of a suitable additive, which releases gases. This method is recommended for the production of aluminum foam sandwiches, sheet foams or products with simple shapes. Another method is casting with a filling. Salt grains can act as fillers being then washed into the aluminum matrix. The porous structure appears in place of the removed filling. Polyurethane foam having a heat-resistant coating layer can play the role of filling material being then burned during the heat treatment. Other making processes of aluminum foam use hollow metal or ceramic balls that are introduced into the metal matrix. Injection molding technique is based on polymer grains using for pores formation. The grains do not decompose at high temperature due to the very short processing time remaining as filler into metal matrix. Other production way of aluminum foam is the powder metallurgy. First, a mixture including aluminum powder and special additives is performed. By its heating, foaming and sintering processes occur and after cooling aluminum foam is formed. The most modern way to obtain metal foam (including also aluminum foam) is additives technology. The 3D shape and structure of the material can be high precision-controlled. This procedure is still very expensive and can be applied only to a low number of materials (Aluminum foam, 2021).

Several papers presented in the literature refer to the choice of adequate expanding agent in the making processes of aluminum foam by the powder metallurgy. In the paper of Koizumi et al., 2011 it is shown that metal foams are usually produced using hydride foaming agents (TiH_2). Less expensive, less dangerous during the use and easier to handle variants, carbonates are recommended, but their performance in this type of process is not sufficiently evaluated. According to the experimental results, it was shown that the agent that begins to decompose after melting the matrix is suitable to lead to obtaining a fine and homogeneous porous structure. Therefore, magnesium carbonate ($MgCO_3$) and dolomite [$CaMg(CO_3)_2$] were chosen as optimal expanding agents for the Al-Si-Cu aluminum alloy. The foam obtained by foaming with dolomite had the density of 1.19 $g\cdot cm^{-3}$ and a homogeneous structure.

The efficiency of using $CaCO_3$ powder as an alternative to TiH_2 foaming agent in the manufacturing process of closed-cell aluminum foam was experimentally analyzed by Kevorkijan in its paper in 2010. The foams were produced from sintered precursors by powder metallurgy and melt route, respectively. In both techniques, $CaCO_3$ powder with 38-120 μm grain size was used as a foaming agent mixed with fine powder aluminum (63 μm). In the first making method, the mixture was compacted by cold pressing into a mold. In the melt route, aluminum powder was melt by induction, following by the addition of $CaCO_3$ powder (between 3-12 wt. %). After heating up to 750 ºC, the melt was mixed and 1/2 ratio of $CaCO_3$ and aluminum powder was added. Finally, the obtained paste was poured into a mold at room temperature and cold pressed. The experiment confirmed that the compressive strength, the energy absorption ability and the microstructure of aluminum foam samples are comparable to the characteristics of products made by TiH_2 foaming.

The experimental manufacture of closed-cell aluminum foam using dolomite as an expanding agent was presented by Papadopoulos et al. in 2011. The raw material was commercial aluminum powder (99.8 % purity), which was melted in a conventional furnace. At 650 ºC, dolomite powder was added to the melt (in variable weight ratio between 1.5-3 %) and the mixture was stirred at 1200 rpm for 1.5 min. Keeping the melt inside the furnace at 720 ºC for 13 min allowed the thermal decomposition of dolomite and the release of carbon dioxide (CO_2), which caused the formation of gas bubbles into the melt mass. After cooling, a porous structure was obtained. The foam density using 2 % dolomite was 0.773 $g\cdot cm^{-3}$ and the porosity reached 71.4 %. By comparison with the foam produced with TiH_2, the microstructure of the experimental material was finer with smaller cell size.

Innovative technical solution of some authors of the current work on the energy efficiency of heating process for aluminum foaming making was shown in the work (Paunescu et al., 2019). The fast and economic method of heating by microwave irradiation was applied in this experiment, in which recycled aluminum waste atomized with jets of nitrogen gas by an own original technique as well as dolomite powder as an expanding agent were mixed and pressed into a metal crucible with removable wall inserted inside a silicon carbide (SiC) crucible thermally protected with ceramic fiber. The process of melting and foaming the mixture took place in a microwave oven adapted for high temperature operation. The decomposition of dolomite occurred in two stages at over 440 ºC and then at over 740 ºC. The duration of the foaming process was very short (9.17-10.92 min). The main characteristics of the foam were: density between 1.16-1.19 $g\cdot cm^{-3}$, porosity within the limits 55.9-57.0 %, compressive strength in the range of 6.83-7.01 MPa, and thermal conductivity between 5.71-5.84 $W\cdot m^{-1}\cdot K^{-1}$. The pore size was between 0.4-0.9 mm corresponding to the highest weight ratio (2 %) of the foaming agent.

The literature offers poor information on the use of microwave heating for obtaining foams of aluminum or aluminum alloys. Alloy foams based on aluminum and titanium were experimentally produced by combustion synthesis of the powder mixture containing the two component metals using microwave heating (Yamamoto et al., 2012). The exothermic powder of titanium and boron carbide was added to the starting powder to increase the burning temperature. Microwave heating has been investigated as a precursors ignition method. It was observed that their expansion is not achieved when the amount of exothermic powder is below 5 wt. %, while when the exothermic powder reaches 10 wt. %, the raw material compaction (at 25-200 MPa) does not affect the foam porosity.

Other experiment in which the microwave heating was used for producing aluminum foam was exposed in the work (Kumar et al., 2021). The precursors were processed by friction. The aluminum foam had pore size between 0.24-1.35 mm and porosity between 41-56 %. These physical characteristics were almost similar to those obtained by heating in a conventional oven. However, the results showed that the manufacturing process by microwave heating is much faster.

In the current paper, the authors aimed to test the production of closed-cell aluminum foam by the own method of unconventional microwave heating applied in the work (Paunescu et al., 2019), under the conditions of replacing dolomite with calcium carbonate as an expanding agent. Also, the work originality is the use of recycled aluminum waste melted by microwave heating and atomized with nitrogen jets for obtaining fine powder of aluminum used as raw material in the experiment.

2. MATERIALS AND METHODS

2.1. Materials

As mentioned above, the making process of aluminum foam aimed at reducing production costs by using cheap materials and forms of energy compared to those of techniques known in the literature. The basic raw material was recycled aluminum waste (generally, from post-consumer drinking doses). After melting the waste in a SiC crucible through microwave irradiation, the liquid metal was atomized with nitrogen jets in a water-cooled installation for capturing very fine aluminum grains (below 32 μm).

$CaCO_3$ purchased from the market in the form of a fine powder, had the grain size below 10 μm.

2.2. Methods

The decomposition reaction of $CaCO_3$ in contact with the molten aluminum at above 700 ºC has been found that is a much more complex reaction. The well-known basis reaction (1) occurs in the range of 800-900 ºC (Karunadasa et al., 2019).

$$CaCO_{3\,(solid)} = CaO_{\,(solid)} + CO_{2\,(gas)} \quad (1)$$

However, the chemical conversion occurs at the $CaCO_3$-liquid aluminum interface, where other chemical reactions could take place:

$$2Al_{\,(liquid)} + 3CO_{2\,(gas)} = Al_2O_{3\,(solid)} + 3CO_{\,(gas)} \quad (2)$$
$$8Al_{\,(liquid)} + 3CO_{2\,(gas)} = 2Al_2O_{3\,(gas)} + Al_4C_{3\,(solid)} \quad (3)$$

These reactions are possible even at temperatures under the melting point of aluminum. By melting, the foam is formed due to the CO_2 released by $CaCO_3$ only if the partial pressure of CO_2 inside the cell is kept below its equilibrium value by reaction (2). The minimum proportion of gaseous CO_2 required to participate in the melt foaming was studied by Gergely et al., 2003 showing that the thermal decomposition below 30 % of the available carbonate could be sufficient for the production of foam with high porosity.

The working method adopted for this experiment included several stages. Aluminum powder and $CaCO_3$ in four weight proportions of 2, 5, 8 and 11 % were dosed, mixed, and introduced into a metal crucible with removable wall. The total amount of the solid mixture was kept constant at 100 g. The mixture wetted with 10 % water and pressed was introduced into a ceramic crucible made of SiC and Si_3N_4 (high microwave susceptible) and thermally protected with ceramic fiber mats. An 800 W-microwave oven of the household type, but adapted for high temperature operation was used (Figure 1) to heat the mixture to 750 ºC. The temperature was measured with a radiation pyrometer through the upper wall of the oven provided with a 30 mm-central hole. The heating duration was 11 min, corresponding to a heating rate of 66.4 ºC/min. Then, the energy supply to the oven was stopped, the two crucibles were together taken out and the melt was stirred (1000 rpm) with an electrically operated device for 2 min. The cooling of the foamed material was freely carried out at room temperature.

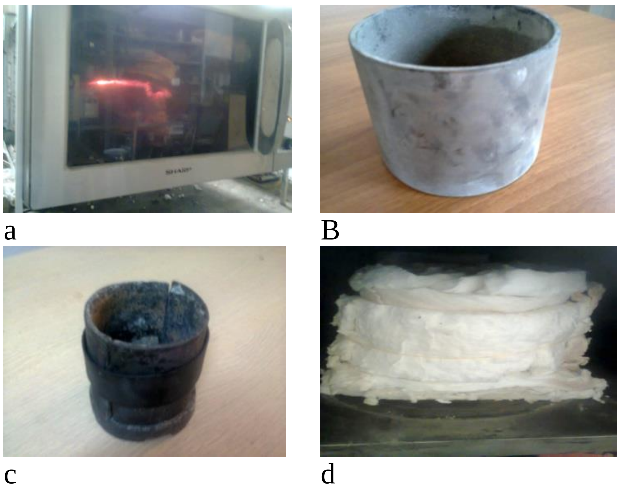

Fig. 1 Components of the experimental microwave equipment

a – microwave oven; b – SiC and Si3N4 crucible; c – metal crucible; d – heat protection of ceramic crucible.

2.3 Characterization methods of the aluminum foam specimens

The density was measured using the gravimetric method (Metrology, 2015). Porosity was calculated as a percentage of the difference between the estimated density of aluminium foam without pores and the measured apparent density including the pore volume related to the density of foam without pores, according to NE 012-99 (Code, 1999). Determining the heat conductivity (at 30 ºC) was made with HFM 446 Lambda apparatus based on the heat-flow method (SR EN 1946-3: 2004). The compression strength was measured using 100 kN-hydraulic axial press machine (EN 826: 2013). The microstructural characteristics of specimens were investigated with Biological Microscope MT5000 model with captured image, 1000 x magnification.

3. RESULTS AND DISCUSSION

3.1. Results

As mentioned above, four experimental variants were chosen, in which the variables were the weight proportions of the aluminum powder and the expanding agent ($CaCO_3$), while the heating temperature, the duration of the process, and the heating rate were kept constant. In Table 1 these data are centralized.

Table 1. Data of experimental variants

Characteristic	Variant			
	1	2	3	4
Apparent density (g·cm-3)	1.10	1.00	0.93	0.88
Porosity (%)	59.3	63.0	65.6	67.4
Heat conductivity (W·m-1·K-1)	5.65	4.88	3.91	3.30
Compression strength (MPa)	7.4	6.1	4.9	3.9
Cell size (mm)	0.04-0.08	0.11-0.24	0.13-0.36	0.14-0.45

Applying the characterization methods of the aluminum foam specimens, their main characteristics were determined and are shown in Table 2.

Table 2. Characteristics of aluminum foam specimens

Data of variants	**Variant**			
	1	**2**	**3**	**4**
Aluminum powder (wt.%)	98	95	92	89
CaCO3 (wt.%)	2	5	8	11
Heating temperature (ºC)	750	750	750	750
Heating time (min)	11	11	11	11
Heating rate (ºC/min)	66.4	66.4	66.4	66.4
Specific energy consumption (kWh/kg)	1.15	1.15	1.15	1.15

The data in Table 2 indicate that with the increase of expanding agent proportion from 2 to 11 wt. %, the density and thermal conductivity decrease, showing the improvement of thermal insulation properties of the foam. Also, the porosity of the material is increasing, as is the cell size range. The compressive strength reaches a high value (7.4 MPa) corresponding to variant 1 and significantly decreases to 3.9 MPa (in the case of variant 4). The microstructural aspect of specimens was analyzed under the microscope, the images being presented in Figure 2. An extremely fine and homogeneous microstructure (cell size between 0.04-0.08 mm) corresponds to variant 1 in which the $CaCO_3$ proportion was the lowest. The microstructural homogeneity is kept also to the other specimens, but cell size is increasing up to 0.14-0.45 mm (in the case of variant with the highest $CaCO_3$ proportion).

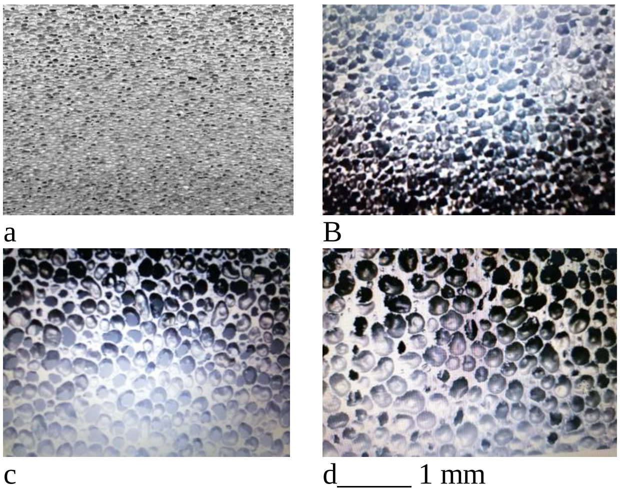

Fig. 2 Microstructural configuration of aluminum foam specimens

a – variant 1; b – variant 2; c – variant 3;d – variant 4.

3.2 Discussion

The technique adopted in this paper for producing closed-cell aluminum foam meets several very current requirements. One of them is the tendency to recycle the material waste. The metallic aluminum required in the process as a raw material was recycled from post-consumer drinking doses. The heating method of metal waste based on microwave irradiation is a significantly more energy efficient procedure compared to conventional heating methods. The specific consumption of the microwave heating process was determined at 1.15 kWh/kg, but it could be considerably lower due to the small amount of raw material reported to the installed power of the microwave oven. Also, the process is "clean", environmental friendly, obviously advantageous compared to thermal processes based on burning a fossil fuel that emits pollutants into the atmosphere. The use of $CaCO_3$ as an expanding agent substitute for TiH_2 was adopted primarily for economic reasons. It was also known from the literature that TiH_2 is a material with a high degree of danger during its use and more difficult to handle. The application of $CaCO_3$ in the experiment described above followed the effect on the quality of the aluminum foam and this effect was beneficial because the characteristics of the foam were considered almost similar to those of cellular aluminum products manufactured with TiH_2.

4. CONCLUDING REMARKS

Attractive lightweight metal foam (closed-cell aluminum foam) for applications in the aeronautical and automobile industries especially, was produced experimentally. The raw material (aluminum powder) was selected from recycled aluminum drinking doses, which was processed through an original technique of melting by heating under the influence of microwave irradiation followed by atomization of the aluminum melt in contact with nitrogen jets in a water-cooled metallic installation. The other component of the starting mixture was $CaCO_3$ as an expanding agent adopted as a substitute for the commonly used TiH_2 that is expensive and dangerous during the operation. The mixture foaming was achieved through applying the microwave heating technique at 750 ºC. The remarkable energy efficiency of this method allowed reaching very high heating rate of 66.4 ºC/min. The closed-cell aluminum foam obtained in this experiment had: apparent density between 0.88-1.10 $g \cdot cm^{-3}$, porosity in the range of 59.3-67.4 %, thermal conductivity within the limits 3.30-5.65 $W \cdot m^{-1} \cdot K^{-1}$, compression strength between 3.9-7.4 MPa, and homogeneous microstructure with cell size below 0.45 mm, almost similar in terms of quality with products made with TiH_2. Practically, all the four foam specimens having characteristics within the limits mentioned above are adequate for applications in aeronautic and automobile domains.

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