

Development of a new process to recover aluminium from thin complex aluminium scrap to employ for primary aluminium alloys

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Current thermal processes for the treatment of metals contaminated with impurities, especially with high contents of organic compounds, present problems in obtaining the metal fraction: the metal fraction oxidises with increasing process temperature and the contact with organic compounds. Often, the quality of aluminium obtained from "dirty" raw materials does not allow using it for applications requiring high metal purity or low metal oxidation (alloying tablets, aluminium shots, ...), or for HPDC structural parts. Some aluminium scrap streams cannot be recovered with existing processes and new developments are needed.

A new two-step continuous process has been developed. It consists of a heating process at 300°C in a controlled oxidising atmosphere furnace, followed by a second heating process at 400-550°C in an inert atmosphere, which results in a combined thermo-mechanical action. The obtained clean material can be continuously sintered under pressure, obtaining a product with a density close to 2.7 g/cc. Several "dirty" materials with initial contents up to 15% wt. C and up to 1.5% wt. O have been studied, reaching to final products containing as low as 0.15% wt. C and 0.3% wt. O. An example with a filter dust from shredding of aluminium profiles is described. The final composition of the product obtained after the treatment allows using the recycled aluminium as a high quality secondary raw material for aluminium alloying tablets, aluminium powder and high-performance aluminium casting alloys.

KEYWORDS: ALUMINIUM RECYCLING, COMPLEX ALUMINIUM SCRAPS;

INTRODUCTION

Aluminium processing residues, in the form of fine particles, sheets and chips, are difficult to recycle due to their high surface/volume ratio, small size with a high oxidising capacity and the presence of contaminants. Recycling aluminium leads to significant savings in energy and greenhouse gas emissions, with only a 5% of the energy required compared with the conventional molten salt electrolysis [1]. Aluminium is a critical raw material and multi-objective design approaches have been proposed to evaluate the environmental impact and the criticality issues linked to raw materials [2]. Aluminium scrap from aluminium profiles contains not only lacquers, but also plastic insulation materials. These profiles are shredded, and aluminium and plastic are separated. Fine particles are sent to a filter with a metal content between 80 and 90% wt. When melting aluminium scrap in a furnace, an

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oxide layer forms on the surface, which results in metal loss and metal yield can be low (loss of material as high as 20%, with the thinner the scrap, the higher the losses). Conventional aluminium recovery methods based on scrap melting are the most widely used by secondary metal recyclers, involving high energy consumption and CO₂ emissions. Induction melting with protective atmosphere can also be used, but due to the higher cost of electricity compared to natural gas, induction furnaces are less used [3]. In Vortex melting system, which consists of a vortex that is generated by a stirring mechanical or electromagnetic device chips are added to the vortex and due to the speed and movement produced in the vortex, chips are immersed in the molten metal and they are melted in a short period of time. This system presents problems for melting lacquered aluminium or aluminium containing plastics, unless a protective atmosphere is employed. The study of a method combined vacuum pyrolysis with dilute sulfuric acid leaching to delacquer pyrolysis shows that the higher the pyrolysis temperatures, the less carbon presents in the solid residue, while the aluminium yield remains [4]. Direct hot extrusion technique in recycling of

aluminium chips shows that properties of the extrudates depend on the process parameters and the alloying elements. High shear strain is required to disperse the oxide layer on the surfaces of the chip [5]. Friction Stir Extrusion recycling process based on the plastic deformation generated by the heat from the friction of the rotation and the chips was employed to obtain extruded products. The energy required is lower, but cracks were detected [6]. A procedure of cold compaction for chips and powder using uniaxial cold compaction followed by a sintering process allows obtaining a sintered product with a high density [7]. Though these processes represent a reduction in energy consumption, they are not industrialized.

MATERIALS AND METHODS

Materials

Filter dust from shredding of aluminium profiles and aluminium parts with high percentage of plastics was tested for organics removal with the developed process. The bulk density was only 0.3 kg/dm³. Particles were long and fines, and they contained organic plastics (PVC, PU...), wood, lacquers, paints... as shown in Fig. 1.



Fig.1 - Filter dust from shredding aluminium profiles.

Methods

Thermal Gravimetric Analyzer (TGA), model SETSYS Evolution, (SETARAM, France) was employed to determine the variation in mass of the samples subjected to a temperature gradient. Samples were heated in air from 20 to 300°C (5°C/min) with a residence time of 1 hour at 300°C with a subsequent heating in an argon atmosphere

(99.998% pure) from 300 to 550°C (5°C/min) and a residence time of 1 hour at 550°C. Tests were performed also in a laboratory muffle, model L9/C6, (EHRET, Germany) with 200 gr of every sample, combining a first process at 300°C in an oxidant atmosphere and a second one in an argon atmosphere at 500°C. To sieve the raw material an industrial circular sieve shaker, model FTI-0550, (Fil-

tra Vibración SL, Spain) was employed. Carbon wt.% was determined with an Automatic Analyzer, model CS-400, (LECO, USA) and Oxygen wt.% with a N, O, H Automatic Analyzer, model TCH-600, (LECO, USA). To simulate at the semi-industrial scale a continuous recycling process, 2 rotatory drum type furnaces were designed and constructed by TECNALIA. Both were equipped with an external electrical heating cover of 200 KW, vibratory feeders and fume extractions systems connected with a filter. The rotation speed of the furnace was adjustable from 1 to 80 r.p.m., with a calculated residence time for the material from 4 to 60 minutes. Temperature thermocouples were installed to automatically control by a PID system to maintain the objective temperature of the furnace camera and of the exhaust gases. Thermocouples were added to control temperatures. To avoid explosions, a continuous CO and O₂ gas concentration measurement system was employed. The inert furnace atmosphere was held inert by a continuous addition of N₂ gas, at a rate of about 60 l/min. The first drum furnace with an oxidizing atmosphere was constructed with internal helixes to allow a good mixing and movement of aluminium particles and swarf inside the furnace, avoiding particles agglomeration and

a regular distribution of the heat. The second furnace was constructed in a similar way, but with a gas inlet and a near sealed vessel.

RESULTS AND DISCUSSION

The average wt.% composition and the standard deviation of as received sample have a C wt.% of 5.0±0.5 and O wt.% of 2.1±0.6. For employing the obtained material for alloying tablets / aluminium powder 0.5-1 %wt. for C and 0.3-0.5 %wt. for O are the acceptable values. TGA analysis showed a clearly defined peak in the oxidizing and in the inert atmosphere. Mass loss starts at approximately 200-250°C with a maximum at 289°C. Mass loss during this stage is a 3%. The second peak occurs in an inert atmosphere starting at 330°C. Mass loss in this case is a 4.5%. The total mass loss attributable to organics is 7.5%, in line with the C obtained by LECO and it would indicate that C present in the initial material is susceptible to being eliminated. Based on these results, seven different tests were performed in a laboratory muffle, combining a stage in an oxidant atmosphere (air) and in an inert atmosphere, as shown on Tab. 1:

Tab.1 - Laboratory tests at different temperatures with loss weights (LW).

Sample	Temp. (°C)	Time (Min)	% C	% O	LW (wt. %)	Temp. (°C)	Time (Min)	% C (wt.%)	%O (wt. %)	LW (wt. 5)
	Oxidant					Inert				
Test 1	300	0	-	-	-	550	120	0.19	1.13	4.5
Test 2-3	300	15	3.66	1.98	3.9	550	120	0.77	2.15	4.2
Test 4-5	300	30	3.11	1.73	4.1	550	120	0.59	1.70	4.3
Test 6-7	300	60	3.01	1.60	4.3	550	120	1.00	1.63	4.4

When a pre-treatment in an oxidising atmosphere it's performed at 300°C, partial C content is reduced in about 1.5-2%. During the subsequent treatment in an inert atmosphere, about a 2.5% of C% is eliminated. A possible explanation is that test material has a large helicoidal shape, so the dilatation of the chips could lead to an easier elimination of carbonaceous depositions from the chip

surface. The minimum obtained O wt.% is 1.1% in the inert atmosphere. When a previous oxidising stage is introduced, there is an increase in the final O content, but the values obtained after the inert stage are similar to those obtained in the oxidising phase. It can be deduced that oxidation takes place during the first phase. The original oxidation cannot be reduced by only thermal treatments.

At 300°C an oxidation process starts, since it is not possible to reduce the value reached afterwards. It was so defined a maximum treatment temperature at the oxidant stage of 300°C.

Seven tests were performed to determine the maximum

treatment capacity of the furnace in its oxidising atmosphere treatment stage and the effect of the inert atmosphere treatment stage in the designed pilot plant. Different feeding rates and process parameters were defined at the semi-industrial trials, as shown in Tab. 2.

Tab.2 - Process parameters used at the semi-industrial scale tests.

Parameter / Sample	Test 8	Test 9	Test 10	Test 11	Test 12	Test 13	Test 14
Feeding rate (Kg/h)	30	60	90	120	90	120	120
Oxidant furnace temp. (°C)	450	450	450	450	300	300	300
Residence time at oxidant furnace (min)	10	5	5	5	15	30	30
Product temp. at the exit of oxidant furnace (°C)	265	380	440	480	135	135	135
Inert furnace temp. (°C)	20	20	20	20	-	-	450
Residence time at the inert furnace (min)	4	4	4	4	-	-	15
C (wt.%)	0.28±0.0	0.46±0.2	0.34±0.1	0.39±0.1	4.77±0.4	4.33±0.9	1.03±0.1
O (wt.%)	1.11±0.3	1.48±0.3	1.60±0.5	1.58±0.4	2.33±0.4	2.75±0.7	1.12±0.3

In Test 8, an oxidising stage with flame has been obtained inside the furnace, with a small reduction in the O wt.% in comparison with the original sample. The obtained O wt.% value was 1.1%, a value that coincides with that obtained in the laboratory in an inert atmosphere, and which could be interpreted as the percentage associated with the intrinsic oxidation of the aluminium in the original product. When the feed flow rate increases, the oxidation of the obtained product increases. This may be due to the higher temperature that the aluminium reaches when the material feed to the furnace is increased, containing a higher organic charge per time unit. Temperature of the material at the exit of the oxidising furnace rises from 265°C with 30 kg/h of feed to 380°C at 60 kg/h, with values that reach 440°C and 480°C with feeds of 90 and 120 kg/h respectively. In Test 11 material, a flameless

oxidising stage was followed by an inert stage. The feed was progressively increased up to 120 kg/h. Under these conditions, a very small reduction of C occurred in the oxidising. Further treatment in an inert atmosphere reduced the carbon content to 1 wt.%. With regard to O, it has been observed that the final value obtained (1.1%), after the inert stage, is similar to that obtained in the previous test (with flame inside the furnace) and in the laboratory test in the muffle furnace with inert atmosphere. On this basis, it is possible to confirm the existence of a limiting value for oxygen, which is approximately a 1% and which corresponds to an intrinsic oxidation of the aluminium fed to the furnace. Under these working conditions, the exit temperature of the product from the oxidising furnace was 135°C, which is far from the upper limit of 300°C established for the process. For a more adequate opera-

tion, this value should reach at least 200-250°C, which is the range where the first important mass reduction peak occurs according to thermogravimetry.

To continue with the study, a new lot of the studied material was employed. A reduction in the average of C wt.% was observed with C 1.4±0.5 wt.% and O 2.9±0.6 wt.%. These variations are very common in the recovery indu-

stry. The aim of these new tests trials was to work at a product outlet temperature of the oxidising furnace close to 300°C. Two setpoint temperatures were tested in the oxidising furnace (375 and 400°C). The process was carried out without flames inside the furnace. Process parameters are summarized in Tab. 3.

Tab.3 - Process parameters at the semi-industrial scale tests.

Parameter / Sample	Test 15	Test 16	Test 17	Test 18	Test 19	Test 20	Test 21	Test 22	Test 23
Feeding rate (Kg/h)	90	90	90	90	53	53	70	53	53
Oxidant furnace temp. (°C)	400	375	375	375	375	400	400	375	400
Residence time at oxidant furnace (min)	10	10	10	10	13	13	10	13	13
Product temp. at the exit of oxidant furnace (°C)	397	183	215	206	220	270	270	220	270
Inert furnace temp. (°C)	-	-	450	500	-	-	-	500	500
Residence time at the inert furnace (min)	-	-	15	30	-	-	-	30	30
C (wt.%)	1.6±0.4	2.1±0.1	0.3±0.1	0.2±0.0	1.6±0.1	2.0±0.2	1.7±0.1	0.4±0.1	0.6±0.1
O (wt.%)	1.1±0.3	1.1±0.1	0.8±0.2	0.4±0.2	1.4±0.2	1.0±0.2	1.3±0.1	0.7±0.0	0.9±0.1

The best results have been obtained in test 18, with a C of 0.24 wt.% and O of 0.45 wt.%. The oxidizing furnace remains in operation for a 24% of the operating time, with the temperature of the furnace oscillating between 371 and 390°C, for a set point of 375°C. The inert furnace remains in operation for a 29% of the operating time, with the furnace temperature ranging from 496 to 510°C, for a set point of 500°C. In the case of C wt.%, there is a clear reduction in the inert treatment, decreasing the final C wt.% by increasing resident time with the same operating parameters. A near 2% wt. of reduction in the O wt.% was obtained in the oxidizing furnace, probably due to the liberation of the aluminum oxide in the surface of the material by the induced dilatation of the increase of the tem-

perature and by the mechanical attrition induced over the treated material. It can be observed a higher decrease in the final O wt.% by increasing the treatment temperature and the residence time.

The low C wt.% could be explained because when the material rotates in the interior of the drum, there is a combined thermo-mechanical effect over the treated material, promoting the collection of fines (inorganics and C) in the filter. This effect promotes a reduction in inorganics and C wt.% in the material treated, especially in the inert furnace. Comparing the results obtained between the sample treated in the muffle furnace and the tests carried out at a semi-industrial level, C wt.% variates from 0.19-1.00

% in the muffle to an interval between 0.28-0.46 in the semi-industrial process with the same lot, and therefore there is a considerable reduction in C %wt. This is due to the fact that in the semi-industrial process, aluminium particles rotate inside the rotary furnaces, with friction between the aluminium particles and falling from the interior wall of the furnace when the material rises inside the furnace during the rotation, allowing part of the fine C particles to be extracted by the filtering system. In the case of the O wt.% with the first lot, the obtained values varied from 1.13-2.15 in the muffle to a range of 1.11-1.59 in the semi-industrial process. The value obtained in the muffle is slightly higher because in the semi-industrial process some of the oxides are collected by the filter.

CONCLUSIONS

The use of a continuous recycling process involving a first oxidizing furnace combined with a second furnace, where the remaining organics are pyrolyzed under an inert atmosphere, allows recycling complex aluminium scraps. The waste known as filter dust from shredding of aluminium profiles has very different levels of organic contamination depending on where it comes from. This fact, demonstrated in the analysis of the two lots, makes difficult to establish optimum general conditions for the treatment of this type of waste. The capacity and process parameters to be used in the treatment of a waste in the furnace is influenced by its organic matter content:

- If the organic content is medium-high, it is possible to work with flame or without flame inside the furnace. When working with flame, the treatment capacity for the defined composition interval decreases, as the product overheats and tends to oxidise. If operating without visible flame, the treatment conditions can be wider, but unburned gases are generated, and they will need to be treated.
- If the organic content is very low, it is not possible to cause a self-ignition of the gases. Under these conditions, the treatment capacity can be increased but the generation of unburned gases that need to be treated is also increased.

In the second furnace with the inert atmosphere a final pyrolysis of the remaining carbonaceous is produced. A thermo-mechanical effect takes part in the furnace, combining an attrition of the aluminium particles among them and the surface of the rotating furnace at high temperatures. This mechanical effect allows removing carbonaceous particles from aluminium scrap and these fine particles are collected in the baghouse filter. This mechanical effect allows reducing the content of C in the final material.

The final quality of the product obtained depends on the initial quality of the product. If there is oxidised aluminium in the original waste, it will not be possible to reduce all the oxygen content. The technology can only reduce some of the oxygen liberated by the thermo-mechanical processing and liberated as fine particles.

The obtained final composition of the tested filter dust particles shows a best C %wt. less than 0.25 %wt., below the optimum of a 0.5 %wt. target. In the case of O, the best obtained values are below the maximum of a 0.5%wt. target. Thus, the semi-industrial trials confirmed the researched process as a valid technology for the obtention of aluminium material with compositions fulfilling the severe requirements to be used for aluminium alloying tablets and for high valuable aluminium alloys.

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