A novel procedure for the evaluation of new refractories for aluminium furnaces

Iban Vicario (1); Leonor de Landía (2); Gorka Mendizabal (3); Nagore Toledo (2); Eva Anglada (1); Patxi Rodriguez (1)

(1) TECNALIA. Industry and Transport Division, Mikeletegi Pasealekua 2, Donostia-San Sebastián E-20009, Spain. Iban.vicario@tecnalia.com; eva.anglada@tecnalia.com; patxi.rodriguez@tecnalia.com

(2) REFRACTARIOS KELSEN S.A. Belen Industrialgunea 1, Aduna E-20150, Spain. ldelandia@calcinor.com; ntoledo@calcinor.com

(3) CALCINOR S.A. Egileor auzoa, 1010. Altzo E-20268, Spain. gmendizabal@calcinor.com

ABSTRACT

Refractory materials for aluminium industry are designed to be resistant to different degrees of thermal, mechanical and chemical wear. The refractory wall thickness reduction during service life increases the heat losses through walls, decreasing the thermal efficiency of the furnace. Last developments are focused on obtaining refractories with better performance and improved insulation properties.

On this regard, a novel procedure has been developed to compare the thermal and chemical performance of different refractories. This procedure includes comparing the resistance of the refractory to molten aluminium, determining corundum and cracks appearance, and measuring the internal and external wall temperatures of a testing furnace using thermography. These temperature measurements make possible to estimate the wall thermal conductivity together with its evolution in time and also validate its simulation modelling in order to be used in future furnace designs. Two refractories have been tested by this procedure for comparative purposes; a commercial alumina castable and an improved alumina castable with better insulation properties

Keywords: Refractories; aluminium furnaces; simulation; end use properties; corrosion resistance; insulation behaviour

1 INTRODUCTION

Refractory degradation and failures in aluminium melting furnaces can be caused by several mechanisms [1,2] such as: chemical reactions between the molten aluminium and the refractory material (corrosion); mechanical degradation of the material by the process environment (erosion and abrasion); ingot loading (impact) or by thermal shocks. All these mechanisms tends to reduce the refractory wall thickness promoting heat losses (insulation of the furnace is reduced) and also increasing the refractory maintenance and repairing [3]. In order to have a good efficiency of the furnace, low thermal conductivity refractories are being continuously developed.

In the furnace, there is an area where the aluminium is in contact with the furnace atmosphere (Bellyband area). In this area there is a triple interface (gas atmosphere, refractory and molten metal), with the presence of a thermal gradient between them. In the area over the molten metal, corrosion of the refractory is produced by the action of the corundum growing from the metal line [4].

In the area of contact between refractory and liquid aluminium is where corundum is created by an oxidation and/or corrosion mechanism, but also by the mechanical cleaning and de-drossing of the furnace. At the surface of the liquid aluminium, aluminium is oxidized with the oxygen presented in the furnace atmosphere (see equation (1) and reference [5]).

$$2/3\ Al_2 + O_2 \Rightarrow 2/3\ Al_2O_3 \quad (1)$$

Liquid aluminium and its alloys react with refractories to form corundum by reducing the silica present in them (see equation (2)).

4 Al + 3SiO2 => 2Al2O3+ 3Si (2)

We can observe in Figure 1 the corrosion of the refractory in the bellyband area.

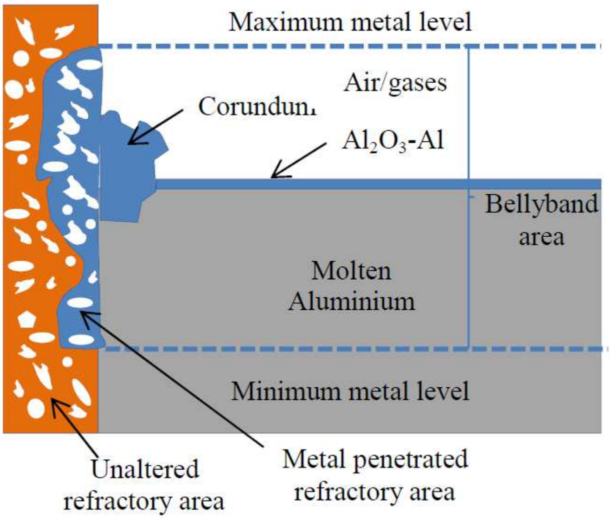


Figure 1. Bellyband area with corrosion of the refractory

In some cases the performance of a refractory material can be predicted from the results of different laboratory tests. However, in other cases, due to a lack of direct correlating tests, some properties are predicted by experience. Hence, it is of a great importance to have validation tests that closely predict the performance of refractories. A common standard procedure to test the new refractories implies the introduction of refractory test samples, at a specific temperature, in a closed furnace where the temperature is equal over the sample. In this procedure, the thermal stress and the thermal conductivity are not determined as in real industrial conditions, neither is the chemical corrosion resistance.

The most important properties to essay for a refractory are:

- Density and porosity.
- Mechanical resistance.
- Thermal conductivity.
- Thermal shock resistance.
- Chemical resistance.

Density is sometimes used as a “rule-of-thumb” indicator of the insulating ability of a refractory, but this can be misleading [6] since other material properties can also affect this behaviour.

The thermal properties of refractories such as thermal conductivity and thermal shock resistance can be measured following different standards (EN-993, ASTM C-182, ASTM C-1171). Thermal data from commercial refractories given by producers have several drawbacks to estimate the real behaviour of refractories on working conditions, being difficult to make comparisons for the selection of the refractories because the lack of information about the test procedures and complete characterization of properties at different temperatures. A similar situation happens when comparing the chemical resistance of the refractories. In this case, only laboratory scale qualitative methods are available (i.e. PRE/R34) which not always totally replicate the real behaviour of the material during service conditions.

This work deals mainly with the determination of the real behaviour of refractories during end use. The objective is to obtain a better refractory testing procedure to determinate by comparison their chemical attack and thermal shock resistance and their thermal conductivity. The resistance to liquid aluminium will be evaluated by determining the corundum and crack’s appearance on the refractory. For the thermal behaviour, the internal and external temperatures of the furnace walls will be measured to determine an equivalent thermal conductivity of the refractory, that can be translated to a heat loss during real application and therefore to an energy and refractory cost. These temperatures will make also possible to evaluate if the wall thermal conductivity is affected by the refractory wear and in addition to adjust/validate the modelling of these materials to be used in numerical simulations. The modelling validation will permit the inclusion of these materials in the future simulation models for furnaces design, with a higher level of confidence.

2 DESIGN OF EXPERIMENTS

A method to simulate the behaviour and properties of refractories is described. An iron vessel with a total capacity, once the refractory lining installed, of 60kg of aluminium works as a container for testing 4 identical walls with different refractories. By using a top cover with electrical resistances, a temperature of 750ºC is obtained in the liquid aluminium, with an internal room temperature of 850ºC, in order to promote corundum formation like in industrial conditions. As the external wall of the vessel is in contact with air, there is a gradient of temperature, like in the industrial furnaces.

2.1 MATERIALS

The molten material for the tests have been chosen from the typical material used for high pressure die casting aluminium, which is the alloy AlSi9Cu3(Fe) according to standard EN AC-46.000, included in the EN 1706:2010 standard.

The refractories selected for the study are dense alumina castables containing a hydraulic binder. Two different refractory formulations were chosen for comparative purposes. On one hand, RCAST A is a standard refractory castable used in contact with molten aluminium. On the other hand, RCAST B is an improved refractory castable designed to obtain better insulation properties and a positive impact on energy savings. Both castables must resist the chemical wear caused by being in contact with molten aluminium but also their thermal properties must be appropriate to endure the thermal and mechanical shocks during service operations.

The refractories tested in the furnace are summarized on Table 1.

Table 1. Refractory materials tested

Material	Thickness
RCAST A – Reference	80 mm
RCAST B - Improved material	80 mm

The base composition for the dense castable are summarised in Table 2.

Table 2. Composition of tested refractories

Chem. Comp.	RCAST A - Reference	RCAST B - Improved mat.
Al2O3 (%)	60.0	68.0
SiO2 (%)	21.0	22.0
Other (%)	19.0	10.0
Density (kg/m^3)	2500	2500

On a first stage, several formulations were developed in REFRACTARIOS KELSEN with the aim of obtaining a new refractory castable with improved insulation properties while maintaining its chemical resistance. Modifications on composition and adjustments on the manufacturing procedures were done to obtain an improved refractory castable.

2.2 EXPERIMENTAL

Once the vessel is prepared, a polystyrene cubic mould is introduced into the centre of the vessel to help create the refractory walls. The different refractories are prepared and poured into the vessel's walls. After one day, the polystyrene is removed and the refractory curing process starts. Refractory is cured during 2 days at 180ºC and after the temperature is increased, at a rate of 100ºC per day, until a maximum temperature of 1.000ºC is reached. In Figure 2 (a) we can observe the refractory after the curing process.

Solid aluminium is introduced in the furnace, and then is melted. The holding temperature for liquid aluminium is set to 750ºC. Every day the furnace is cleaned and de-drossed. Every week the furnace is emptied and the lining is revised for crack detection. Pictures are taken to each of the walls and different behaviours between materials are checked.

On that basis, a Thermographic Camera is used to measure the internal and external temperature of the furnace. The thermal image is adjusted by measuring wall temperatures with a calibrated thermocouple. A FLIR Systems ThermoVision A320 camera is employed to obtain the thermal images, and a calibrated contact pyrometer to determine the real temperature of the internal and external walls of the furnace. In order to analyse the images and to adjust the images with real temperatures, the IR monitoring and Thermo Cam researcher professional 2.9 software packages are employed. We can observe in Figure 2 (b) how is recorded the internal temperature and the adjusted thermal image in Figure 2 (c).

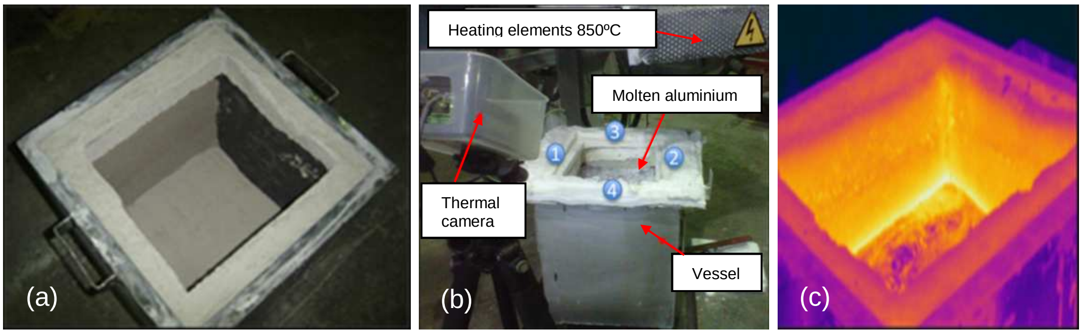


Figure 2. (a) Refractory lining after sintering; (b) Test equipment; c) Thermal image.

This methodology is applied during 6 months, and the test is stopped if important cracks are detected in the lining before that time.

3 RESULTS AND DISCUSSION

The disclosed procedure was used to evaluate the two aforementioned refractory materials which were installed on opposite walls of the testing vessel.

3.1 AS MANUFACTURED – BEGINNING OF THE TEST

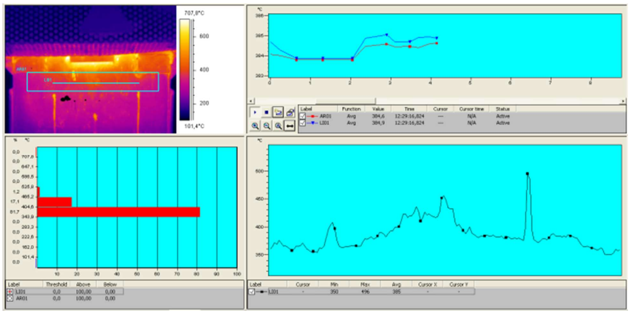
The results in the external area of the furnace are summarized in Table 3.

Table 3. External average temperature of the furnace wall

Reference	Temperature (ºC)
RCAST A - Reference	397ºC
RCAST B - Improved mat.	385ºC

The emissivity parameter was established with a value of 0.44 in the thermal analysing software. With this value, the difference between pyrometer measured temperature and temperature obtained with the thermal camera was less than 1ºC. The minimum fluctuations of temperature in function of time are also available.

In Figure 3 we can observe how the external temperature of the refractory, the average external temperature, the standard deviation and the distribution of temperatures can be determined.



Label	Function	Value	Time	Cursor	Cursor time	Status
AR01	Avg	384,6	12:29:16,824	---	N/A	Active
LI01	Avg	384,9	12:29:16,824	---	N/A	Active

Label	Threshold	Above	Below
LI01	0,0	100,00	0,00
AR01	0,0	100,00	0,00

Label	Cursor	Min	Max	Avg	Cursor X	Cursor Y
LI01	-	350	496	385	-	-

Figure 3. External refractory temperature determination

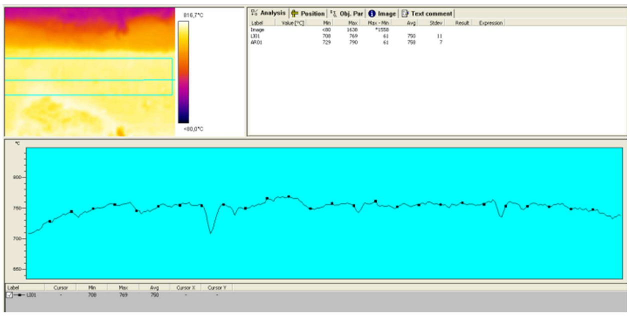
Depending on the surface quality of the steel surface of the vessel, some points showed punctually higher or lower temperatures. In order to equilibrate these variations a square analysis area is defined to obtain the average temperature values and compare them with the linear values.

The results in the internal area of the furnace are summarized in Table 4

Table 4. Internal average temperature of the furnace wall

Reference	**Temperature (ºC)**
RCAST A - Reference	714ºC
RCAST B - Improved mat.	750ºC

We can observe in Figure 4 how is determined the internal temperature of the refractory, the average internal temperature and the standard deviation. In this case, the emissivity parameter is established as 0.88. With this value the difference between the pyrometer measured temperature and the temperature obtained with the thermal camera is less than 2ºC. We can observe the fluctuations of temperature in function of time.



Label	Value [ºC]	Min	Max	Max - Min	Avg	Stdev	Result	Expression
Image		<80	1638	*1558				
LI01		708	769	61	750	11		
AR01		729	790	61	758	7		

Label	Cursor	Min	Max	Avg	Cursor X	Cursor Y
LI01	-	708	769	750	-	-

Figure 4. Internal refractory temperature determination

The best results are obtained with the new improved castable refractory, based on the smaller external temperature and the higher internal temperature that this material showed during tests in the furnace.

These temperature measurements make possible to estimate the thermal conductivity of the refractories by means of an analytical modelling. Heat transfer across the wall is determined by equations (3) to (5). Equation (6), derived from these, permits to estimate the value of the thermal conductivity.

$$Q_{across\ wall} = K(W/mK) \cdot \frac{A\,(m^2)}{L(m)}\,[T_{int\ wall} - T_{ext\ wall}](K) \qquad (3)$$

$$Q_{across\ wall} = K(W/mK) \cdot \frac{A\,(m^2)}{L(m)}\,[T_{int\ wall} - T_{ext\ wall}](K) \qquad (4)$$

$$Q_{across\ wall} = Q_{ambient\ dissip} \qquad (5)$$

$$K\,(W/mK) = L(m) \cdot h\,(W/m^2K)\,\frac{(T_{ext\ wall} - T_{ambient})(K)}{(T_{int\ wall} - T_{ext\ wall})(K)} \qquad (6)$$

This is especially interesting for the case of the new improved refractory, as it is a new developed material which has not ever been modelled before now. It permits to validate the expected thermal conductivity value increasing the level of confidence in its thermal behaviour. The thermal conductivity estimation for the case of this newly developed refractory has been done considering the measured temperatures ($T_{int\ wall}$ = 750 ºC, $T_{ext\ wall}$ = 385 ºC), a wall thickness of 80 mm and typical values for ambient temperature (20 ºC) and convection coefficient (between 5 and 25 W/m^2K, typical values for natural convection). The value obtained for the thermal conductivity is consistent with the obtained by physical characterization at laboratory scale.

In addition to the analytical model, these results have been also validated by means of numerical simulation. A steady state heat transfer simulation with NX Nastran thermal software package using finite elements (FEM) has been performed confirming the results obtained by the analytical model. This numerical simulation permits also to evaluate the thermal gradient across the wall (see Figure 5).

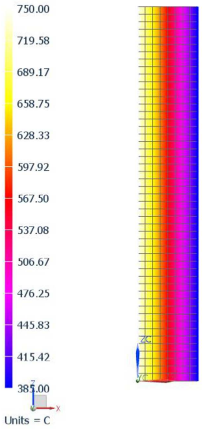


Figure 5. Temperature results from FEM simulation for RCAST B.

These performed validations make possible to include this new refractory material in future simulation models for furnace design with a higher level of confidence. This fact has been verified during the test phase of an industrial melting chamber prototype.

In this case, chamber prototype walls are not composed by only a refractory layer. Instead, they are formed by several layers of different isolating materials (see Figure 6) as it is usual at industrial furnaces. An identical configuration is employed on the opposite wall of the prototype with refractory material RCAST A used instead of RCAST B. In order to validate the numerical simulation model, temperature measurements have been taken from the molten alloy, at the external surface and at one more point in the middle of the refractory linings. The temperature results obtained from simulation, once the model has been properly adjusted, agree well with experimental data, confirming the validity of this modelling to be used for future designs for this type of furnaces. Table 5 collects the temperature values registered experimentally during the industrial test execution together with the values obtained from simulation for RCAST B wall. Figure 7 shows the temperature distribution across the wall obtained by simulation and the thermocouple position into the wall.

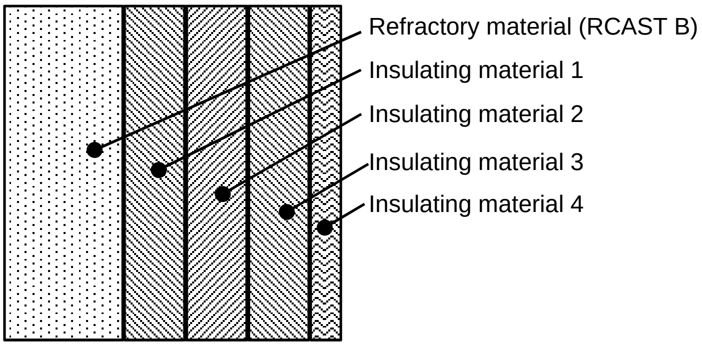


Figure 6. Materials of the industrial furnace prototype wall

Table 5. Temperature of the industrial prototype. RCAST B wall

Case study	**Alloy temperature (ºC)**	**Temperature into the wall (ºC)**	**Temperature at external surface (ºC)**
Experimental measurements	815	460	38
Simulation results	815	458	41

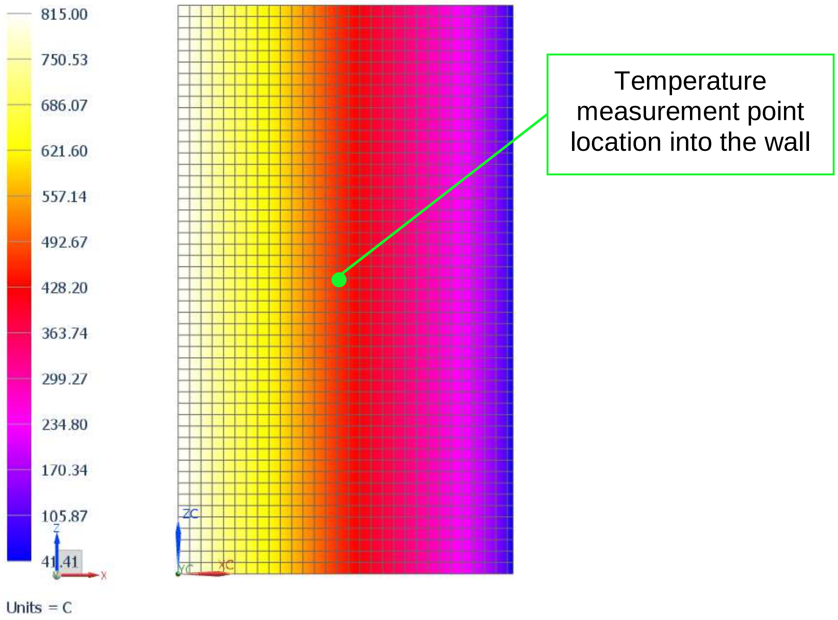


Figure 7. Temperature results from FEM simulation. Industrial prototype

3.2 SERVICE LIFE – END OF THE TEST

The data collected during the test campaign for wall temperatures (internal and external) are shown at Table 6. The wall thickness reduction during this time, which would decrease the thermal efficiency, has not been significant (thickness reduction from 80 mm to 77 mm).

Table 6. Data collected during test campaign

DATA	Test start	4 months	5 months	6 months
RCAST A - Reference				
$T_{internal}$ (ºC)	714	702	669	710
$T_{external}$ (ºC)	397	316	244	240
RCAST B – Improved mat.				
$T_{internal}$ (ºC)	750	707	658	725
$T_{external}$ (ºC)	385	261	213	219

The same procedure for thermal conductivity estimation based on the analytical model, has been used to study the evolution of the wall thermal conductivity during time. Figure 8 shows the fall in the wall thermal conductivity that takes place during the test for the studied refractories. As can be observed, tendency is similar for both refractories. The thermal conductivity of the new improved refractory wall is maintained below the values of the standard refractory during the whole test. It is though that the drop observed in the wall thermal conductivity, is related with the chemical attack suffered by the refractory in contact with the aluminium including the oxide/corundum creation (see Figure 9).

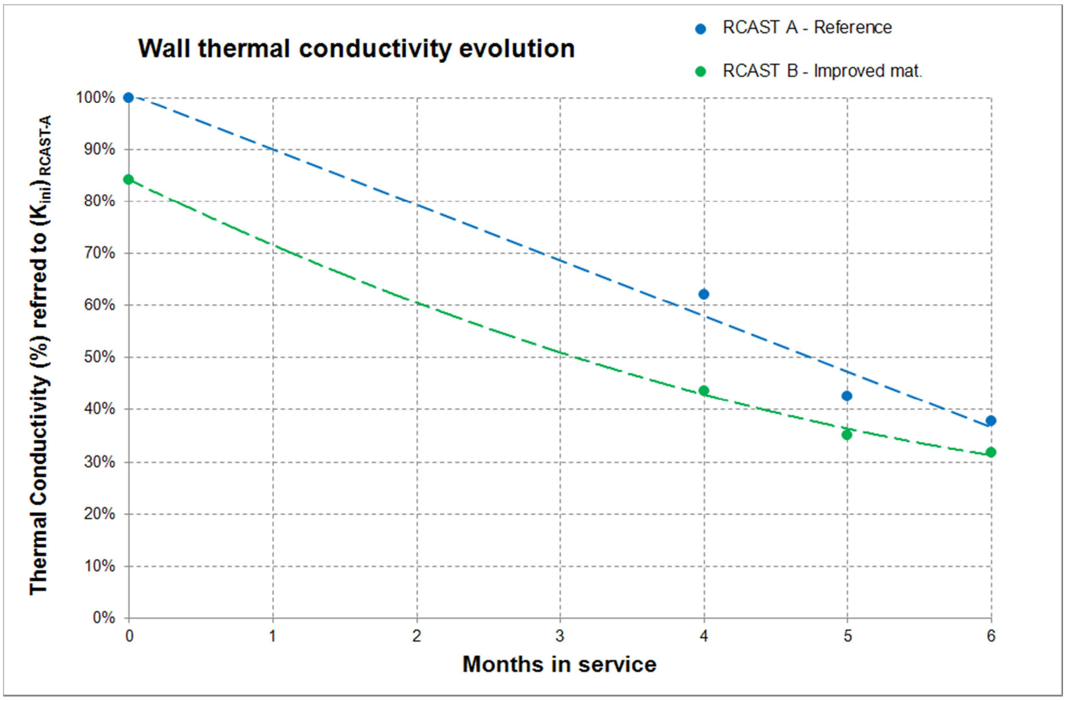


Figure 8. Wall thermal conductivity evolution

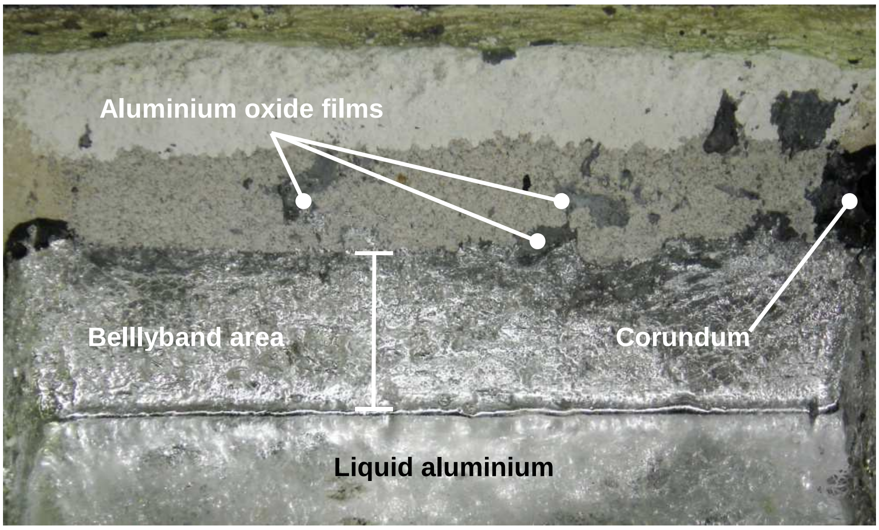


Figure 9. Furnace walls in service

An example of the measurement of the corrosion by image comparison method is shown on Figure 10. We can observe that corundum formation is higher in the reference material RCAST A, and that the improved refractory RCAST B has a better resistance to chemical attack and a lower corundum formation.

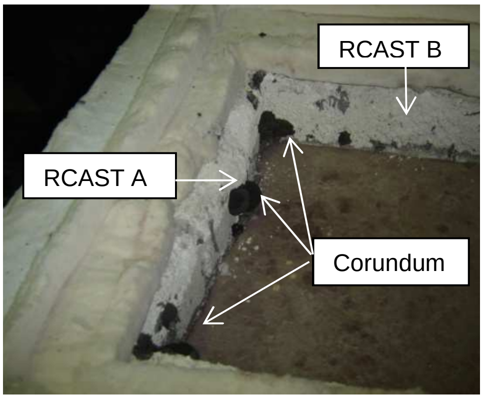


Figure 10. Corundum and cracks formation comparison between different walls

4 CONCLUSIONS

There are standard tests in order to determinate refractories' properties, but they are not always capable of determining their behaviour in real end user conditions. The work reported in this paper has permitted to develop a test procedure that can compare different refractories in semi industrial operation. The main properties that can be compared are:

- Thermal conductivity.
- Corrosion resistance.
- Thermal stress resistance.

For that purpose, a special vessel that replicates the real operation conditions of an aluminum furnace has been used to carry the refractories and test them.

By using and adjusting a thermal camera with real measured temperature it's possible to determinate the internal and external temperature distribution and quantify differences between different refractories. With this procedure thermal conductivity of materials is tested in as built condition and also during the whole service life. Results showed improved insulation properties of the new developed castable refractory.

The system permits also to adjust/validate simulation models with real data, increasing the accuracy of simulation results and providing a good designing tool for the development of new refractory linings.

Finally, and using the same testing vessel, the chemical corrosion resistance of the installed refractories can also be evaluated by determining the corundum formation and crack's appearance on the refractory surface. The newly developed refractory castable was this way validated on its improved chemical resistance.

Industrial validations confirmed the improvement of performance in real industrial conditions by comparing the two refractories in the same furnace at the same time.

5 ACKNOWLEDGMENTS

This work has been has been developed in the framework of the 'New Designs of Ecological Furnaces' - EDEFU project (Grant Agreement 246335) co-financed by the European Commission under the Seventh Framework Programme for Research (FP7).

The authors are also much grateful to the personnel of REFRACTARIOS KELSEN S.A, CALCINOR S.A and GIS2003, companies that collaborated in aspects related to the design and analysis of results.

6 REFERENCES

[1] T.W. Dunsing, Tips on Good Refractory Practice for Aluminum-Melting Furnaces, Industrial Heating. April (2002) 27–29.

[2] P. Bonadia, M.A.L. Braulio, J.B. Gallo, V.C. Pandolfelli, Refractory Selection for Long-Distance Molten – Aluminum Delivery, American Ceramic Society Bulletin. 85 (2006) 9301.

[3] K.-M. Peters, R.M. Cravens, J.F. Hemrick, Advanced ceramic composites for improved thermal management in molten aluminum applications, in: Energy Technology Perspectives, TMS (The Minerals, Metals & Materials Society), 2009: pp. 241–247.

[4] A.D. Porterfield, Aluminium resistance refractory composition, U.S. Patent 4,806,509, 1989.

[5] R.N. Nandy, R.K. Jogai, Selection of Proper Refractory Materials for Energy Saving in Aluminium Melting and Holding Furnaces, International Journal of Metallurgical Engineering. 1(6) (2012) 117–121.

[6] A. Wynn, M. Marchetti, E. Magni, Insulating firebrick – maximising energy savings through product selection, (n.d.).