Oxidation resistance and corrosion resistance of silicon carbide side lining

Andrey Yurkov

Chief specialist – refractory development,
PJSC “Voljsky Abrasive Works”, Voljsky, Volgograd region, 404130, Russia
Corresponding author: yurkov_al@vabz.ru

Abstract

Si_3N_4-SiC materials have considerable strength, yet in Al reduction cells the decay of SiC side lining takes place due chemical corrosion and oxidation, sometimes accompanied and enhanced by physical erosion. N-SiC lining materials in reduction cells should be corrosive resistant to electrolyte and oxidation resistant. In aluminium reduction cell the upper part of SiC lining is subjected to interaction with oxygen, carbon dioxide and carbon monoxide from air. However other parts, covered with side ledge and near the bus bar windows, also may be oxidized during the service. Probably oxidation resistance and corrosion resistance of Si_3N_4-SiC materials may be governed by different factors. The priority of oxidation resistance and corrosion resistance to cryolite of silicon carbide over silicon nitride leaves many questions. Anyway, silicon nitride and silicon carbide slowly oxidize during the service in N-SiC side lining of reduction cell. The question on priorities of oxidation and corrosion resistance of one over another has aspect not only in the service life time in reduction cell, but also in uncertain optimum of concentrations, slight porosity and concentration gradients.

Keywords: Silicon carbide; silicon nitride; oxidation; corrosion.

1. Introduction

Silicon carbide side lining of aluminium reduction cells has been well known for many years. In the scientific literature, other materials are discussed as potential candidates for side lining elements - SiC based [1, 2, 3, 4] and non-SiC based [5]. However, the change of N-SiC side lining in the construction of aluminium reduction cell for another more resistant material is a problem of the future. Probably, when inert anodes will be implemented, profound research on new side lining materials will be required, and a new material will replace nitride bonded silicon carbide.

Currently there is well-known SINTEF test for the corrosion resistance of N-SiC [6, 7], and it is accepted by many smelters. The procedure of this testing simulates the behaviour of side lining during the first hours of electrolysis. There are the variations of this test for the corrosion resistance [8, 9, 10]. Also there is a corrosion resistance test without application of the electric current [11]. It is performed with preliminary oxidation of N-SiC samples.

Usually the decision on the possibility to use the material in a lining of the cell is based on the corrosion test results.

2. Experimental

Standard industrial N-SiC side lining refractories, produced at VAW Voljsky Abrasive Works, Russian Federation (VAW) were used for the analysis. The properties of materials (porosity, density, strength) were determined using standard methods. The chemical composition of the materials was determined by standard wet chemistry methods (SiC, Si, SiO_2, CaO, Fe_2O_3, Al_2O_3) and by XRD (SiC, Si, SiO_2, Si_3N_4). Nitrogen and oxygen were determined using the LECO method. Silicon oxynitride, Si_2ON_2, α- and β- modifications of silicon nitride were determined by XRD. Structures were analyzed at Nikon Eclipse optical microscope and Tescan X-max SEM with EDX Oxford instruments microprobe analyzer.

3. Results and discussion

Although there are no unique specifications of nitride bonded silicon carbide side lining material for aluminium reduction cells, the composition is more or less standard. Silicon carbide content varies from 72 % to 75%, silicon nitride and silicon oxynitride content is in the range of 15 – 25 %. Silica content, that is supposed to be non-desirable admixture, is usually below 1 % or even below 0.5 % (in reality it is in the range below 0.3 %).

During the service of N-SiC side lining in Al reduction cell, the concentration of silica slowly increases. Unfortunately, there are no well proven data on the increase of silica in the lining with service time. However, this may be explained by the fact, that it is impossible to make such dependence, because there are too many variables. In different parts (Figure 1) (above the melt, under the melt and in the lower part), N-SiC side lining undergoes different transformations.

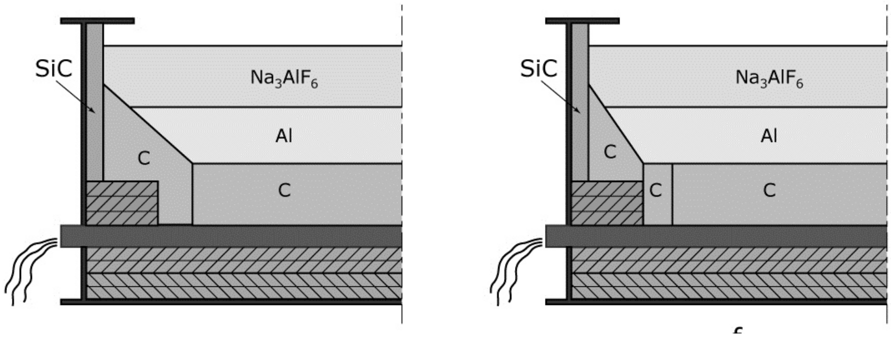

Figure 1. Different zones in SiC side lining.

Another variable is the existence of side ledge (Figure 2), absence of the side ledge and thin side ledge. The consequence is different temperature in one and the same zone of the side lining and different kind of interaction - in case of absence or existence of side ledge.

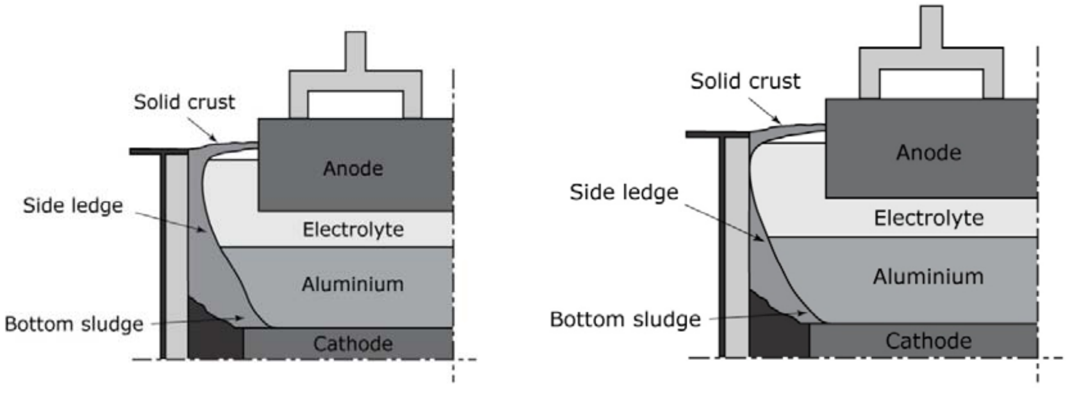

Figure 2. Cross section of the reduction cell: a) with side ledge;b) almost without side ledge in the upper part.

Several kinds of interactions take place between N-SiC refractory material during the service in aluminium reduction cells.

The questions on priority of the direct reactions of silicon carbide and silicon nitride with the constituents of electrolyte in liquid and vapour phase over the oxidation reactions of silicon carbide and silicon nitride with the following reactions of appearing silicon oxide with the components of electrolyte and following dissolution of the products in electrolyte has not only theoretical value.

Microstructure of N-SiC material may have considerable influence on oxidation resistance, especially with respect to gas permeability and pore size distribution.

5. References

1. D. John, R. Maurage, “Sialon Bonded Silicon Carbide Sidewall Pieces for the Aluminium Reduction Cell”, Ceram. Ind. Int., 1992, 182 (1992), p. 42.
2. A.F. Flickel, M. Volker, On the Technological Potential of SiC-TiB2-Si3N4 Compounds in Al-electrolysis Cells”, Light Metals 1994, pp 493-499.
3. M.I. Jones, R. Etzion, J. Metson, Yu. Zhou, H. Hyuga, Y. Yoshizawa, K. Hirao, “Reaction bonded Silicon Notrode – Silicon Carbide and SiAlON – Silicon Carbide Refractories for Aluminium Smelting”, Key Engineering Materials, 403 (2009), pp 235-238.
4. Zhigang Huang, “Excellent Cryolite Resistance and High Thermal Conductivity SiC Sidewall Material for High-amperage Aluminium reduction cells”, Light Metals 2010, pp 889-894.
5. X.Y. Yan, R.Z. Muk hlis, M. A. Rhamdhani and G.A. Brooks, “Aluminate spinels as Sidewall Lining for Aluminium Smelters”, Light Metals, 2011, pp 1085-1090.
6. E. Skybakmoen, H. Gudbransen and L.T. Stoen, “Chemical Resiatance of Sidelining materials based on SiC and Carbon in Cryolitic melts – a laboratory study”, Light Metals 1999, pp 215-222.
7. E. Skybakmoen, L. Stoen, J. H. Kvello and O. Darrel, “Quality evaluation in Nitride bonded Silicon Carbide Sidelining Materials”, Light Metals 2005, pp 773-778.
8. J. Zhao, Z. Zhang, W. Wang, G. Liu, “Test method for Resistance of SiC material to Cryolite”, Light Metals, 2006, pp 663-666.
9. B.L. Gao, Z.W. Wang and Z.X. Qiu, “Corrosion Tests and Electrical Resistivity Measurement of SiC-Si3N4 Refractory Materials”, Light Metals 2004), pp 419-424.
10. C. Cao, B. Gao, Z. Wang, X. Hu, Z. Qui, “A new test method for evaluating Si3N4-SiC bricks corrosion resistance to aluminium electrolyte and oxygen”, Light Metals 2006, pp 659-661.
11. R. Laucournet, V. Laurent, Didier Lombard, Chemical resistance of sidelining refractory based on Si3N4 bonded SiC, Light Metals 2008, pp 961-966
12. J. Schoenhahl, E.Jorge, O. Marguin, S. Kubiak, P. Temme, Optimization of Si3N4 bonded SiC refractories for aluminium reduction cells, Light Metals 2001, pp. 251-255.
13. A.Proshkin, V. Pingin, P. Polyakov et ali, Study of the state and dynamics of Wear of the side lining in the cathods of Aluminium Cells, J. Siberian Federal University. Engineering and technologies, , 2, 2013 6, pp. 276-284.
14. A. Yurkov, O. Danilova, A. Dovgal, V. Doroganov, E. Evtushenko, Investigation of corrosion of N-SiC material by the melt of copper and the copper slag, to be published in issues of UNITECR 2015, Vienna.
15. G.X. Wang, G.Q. Lu, B. Pei, A.B. Yu, Oxidation mechanism of Si3N4-bonded SiC ceramics by CO, CO2 and steam, J. Mater. Sci, 33 (1998), pp 1309-1317.

16. R. Paulek, “SiC in Electrolysis Pots: an Update”, Light Metals 2006, pp.655-658.
17. R. Etzion, J.B. Metson, “Factors Affecting Corrosion Resistance of Silicon Nitride Bonded Silicon Carbide Refractory Blocks”, Journal of the American Ceramic Society, 95 (2012), pp 410–415.
18. T. Tonnesen, R. Telle, M. Breuers, Oxidation resistance of SiC based refractory lining in steam atmospheres - material properties and testing methods, 47th Int. Colloquium on Refractories, Aahen, 2004, pp 214-217.
19. T. Tonnesen, E. Goerenz, R. Telle, Examinations of different bonding characteristics of refractory SiC and the influence on oxidation resistance, Interceram, Refractories manual, 2007, pp.14-18.
20. T. Tonnesen, R. Telle, Refractory corrosion in industrial waste incineration process, Refractories Worldforum, 2009 (1), 1, pp. 71-76.
21. ASTM C863 – 00 (2010) Standard Test Method for Evaluating Oxidation Resistance of Silicon Carbide Refractories at Elevated Temperatures.