

SPENT POTLINING: AN UPDATE

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Abstract

This paper continues of a review given in 1999 and resumes practices to treat the waste from used cathodes of the primary aluminium industry. The waste cathode lining consists of a carbon part and a refractory part. Spent potlining contains water-soluble fluorides and small amounts of leachable cyanides, and is therefore classified as hazardous.

Besides inertization - such as treatment at high temperature, by vitrification, or by leaching and flotation - the refractory component can also be used for instance in the manufacture of cement, of concrete and brick, or as a road pavement additive, or and as a potline electrolyte additive for the production of Al-Si pre-alloys.

The carbon components can be used as a fuel additive in cement kilns, but also as an additive in the pig iron and steel industry.

Various ways to treat and use spent potlining have been published in the meantime, and will be reviewed here.

Introduction

Besides some 40m tpy of primary aluminium, the world's smelters produce about a million tonnes per year of toxic waste in the form of spent potlining. This means about 25 kg of SPL per tonne aluminium produced. Hitherto, most of this SPL has gone to landfill. This must change if the industry wants to claim a reasonable degree of sustainability and environmentally tolerable emissions.

During the last years, methods for processing spent potlining (SPL) were reviewed several times [1-8]. However, an excellent overview was presented by Sørli and Øye [9]. The following update is a continuation of [1].

Liu and Lou [10] found that the carbon hearth made of anthracite and/or graphite undergoes great changes in its various physical properties, chemical compositions, internal microstructures, carbon morphology and crystalline structure during the aluminium smelting process. The generally applied physical, chemical, and thermal processes to process spent potlining are inefficient, but they could be reduced or possibly eliminated, according to Young [8], if the pots were designed for recyclability using a management technology which has been elaborated. So, as proposed by Sturm et al. [11] thermal insulation of electrolysis pots uses alumina instead of refractory bricks. Typical analyses of spent potlining have recently been published several times [12-15]. The analysis of Krüger [12] is presented in table 1.

Table 1				
SPL typical components and concentrations [12]				

Component			
	Both cut	2 nd Cut	
	together	Carbonaceous	Refractory
	100%	56%	44
Carbon	33.1	54-64	18.2
Total fluorides	15.7	6-20	4-10
Free alumina	22.3	0-15	10-50
Total aluminium	15.1	5-15	12.6
Total sodium	14.2	5-12	12.0
Calcium	1.8	0.5-4	1-6
Quartz	2.7	0-6	10-50
Phosphorous	0.3	0-650 g/t	0-300 g/t
Sulphur	0.1	0.1	0.1

Thermal processes

Crushing SPL on site and exporting it in bulker bags was proposed by [16]. As SPL is rich in carbon, this can be used as a fuel for instance in the brick, cement and pig iron industries. For crushing SPL, Chen and Li [17] determined its physical behaviour. They observed that graphite and sodium in SPL make it sticky, slippery and difficult to crush. They also carried out tests to determine the chemical stability of the fluorides in the SPL [18].

Crushed first cut of SPL can be used as collar paste for protecting anode stems [19].

In the USA landfill disposal of SPL without inertisation is forbidden [20, 21]; it is treated as hazardous material due to its water soluble fluorides and cyanides. Possible treatments include heating a crushed SPL mix to about 1000°C and adding lime to oxidize cyanides and to bind the fluoride, as proposed by Karpel [22] and by Li and Chen [23]. Oliveira et al.[24] proposed to heat second cut SPL up to more than 750°C to remove molten and volatile impurities, while Utkov et al. [25] treat the carbon rich part with an FeSO₄- solution in order to neutralise water soluble NaCN. Courbariaux et al. [26] propose to treat crushed SPL in a circulating fluid bed.

Balasubramanian et al. [27] proposed vitrification of SPL by adding small additions of glass former along with traces of nucleation agents to aid crystallisation, and then melting at around 1300°C.

Then Lazarinos [28, 29] treated SPL in a gasification combustion, the main reason being to destroy cyanide compounds mostly contained in the carbon fraction.

Because of its high energy and fluoride content, Blinov and coworkers [30] proposed a pyrohydrolysis process to recover fluorine as HF, while the carbon rich part is used in pig iron manufacture.

The development of the Ausmelt process to treat SPL was reviewed several times [22, 31-40] during 2001 and 2004. In 1995 Portland Aluminium of Australia invested about US\$16m to construct a research and development processing facility to treat SPL. Alcoa, in cooperation with Ausmelt Ltd., used a pyrometallurgical process to combust and melt SPL. On one side this produces hydrogen fluoride laden gas, which is cooled and converted into aluminium fluoride, and on the other side it produces an inert sand. Since 2001 the Ausmelt SPL treatment plant is in operation and can process 12,000 tpy of SPL.

Chemical processes

A low-technical method is needed to treat SPL material, but processing the cathode wastes (carbon cut and refractory cut) only with water [37] does not seem enough to eliminate all harmful elements. Therefore Baranovskii [41] proposed to crush first cut material, to mix it with limestone, and then to add this mixture to an aqueous slurry in order to recover soda and potash.

As aluminium electrolysis carbon froth and spent potlining are inevitable, Lu et al. [42] proposed to separate these components by flotation. In this respect Mirsaidov et al. [43] propose to use kerosene and pine oil as flotation agents. On one hand the scientists separated a cryolite- alumina concentrate, and they burnt out the remaining carbon at 800°C in a rotating furnace.

The principles for leaching SPL were elaborated by Silveira et al. [44, 45]. They observed that for all the pots studied the pH of SPL is at 10-11.8, and the total determined fluoride content at high pH was 5.13-11.41%. But the total fluoride content in leached fluid at pH 5 was only 0.26-3.46%. Therefore leach liquid must be alkaline above ph 12 in order to effectively process SPL.

Leaching SPL with water and concentrated H_2SO_4 was proposed by Zhao [46] to recover HF. The liquids are filtered for the manufacture of graphite powder, aluminium hydrate and alumina, while fluorides and sulphates are manufactured from the filtrate.

Lisboa et Steel [47] examined the leachability of NaF, CaF_2 and cryolite from spent potlining. By manipulating solution equilibira, they precipitated fluoride in a form that can be recycled back into the pot. Following removal of fluoride, they achieved high degrees of separation of graphite from the remaining refractory compounds by using a density separation technique.

Platt [48] proposes the complete treatment of spent potlining and salt slag in order to avoid landfill. Electrolysis produces 22-24 kg/t of spent potlining per tonne of aluminium. SPL is hazardous due to its high cyanide and fluoride contents. Befasa collects such material together with salt dross from remelters to treat chemically at special sites. Besides gases such as hydrogen, ammonia, methane, etc., at the end of the reaction stage Befasa obtains solids which are pumped to a vacuum belt filter. Here the resulting oxides and solids undergo in-process testing to ensure reduced levels of water-soluble chlorides in line with customer requirements.

Alcan identified its low caustic leaching and liming (LCL & L) hydrometallurgical process as the most efficient means of recuperating the chemical values contained in SPL. This process Alcan has reviewed several times [49-50]. An initial water leach was followed by a dilute caustic extraction and washing steps. Cyanides leached as NaCN in the initial water slurry were precipitated as solid ferrocyanide compounds which underwent tests for leachable fluorides. SPL leachate of the LCL&L process yields valuable crystalline NaF and caustic liquor for recycling into the Bayer plant. In December 2000 [51], Alcan deposed a project at the ministry of Quebec for the treatment of up to 80,000 tpy of SPL using the LCL&L process. To construct the plant Alcan put aside some C\$150m [52, 53]. Schaffer reported [54] that this plant would create more than 50 jobs, would reduce

Alcan's treatment costs considerably [55], and would also be able to treat SPL from third parties. In the meantime the investment costs increased to US\$180m [56] and construction started in the Autumn of 2006. Alcan reported about the progress of the construction [57-59] and after the plant was finished in February 2008 [60], the treatment centre was officially inaugurated in June 2008 [61]. Then Hamel and co-workers [62] reported about the valorization potential of the main by-products of LC&L, which are: a carbonaceous by-product (CBP); a concentrated caustic liquor; and fluorides including NaF or CaF₂. Once operating at its full capacity of 80,000 tpy, the plant will produce 78,000 t of CBP (30% water); 80,000 t of caustic liquor (27% NaOH) and 54,000 t of CaF₂ (50% water). In 2009 about 26,000 t [63] of SPL were already processed in this new plant.

Final use of SPL

The literature relates that untreated spent potlining can be used especially in the klinker (cement) industry and also in pig iron or steel making. For first and/or second cut SPL other special solutions have been developed.

Cement kiln

Processing SPL in klinker furnaces has been proposed several times [14, 22]. In general the first cut serves as fuel while the second cut of SPL serves as klinker aid [64-66]. Venancio et al. [66] determined the calorific power in the first cut of SPL to be up to 5 MJ/kg. Therefore the coke fuel for the klinker furnace could be reduced by 111 tpm, and the klinker forms at a lower temperature by 80°C allowed the use of a cheaper coke with higher sulphur content, with no impact on emissions.

Hillside and Bayside Aluminium [65] have worked for years with Pretoria Portland Cement to develop a one-step solution. Introducing SPL into the cement kiln at high temperature destroys cyanides and binds fluorides with calcium which is present in the cement or lime.

The carbon rich first cut of SPL is, according to Cardoso [67], a high calorific material with an heat input of 16 MJ/kg, and the second cut is used in large quantities in the cement industry.

For Chen [68] it is only inert SPL, which can be recycled and forwarded as raw material for cement production.

Pig iron and steel

The use of SPL in the pig iron industry both as fuel and for decreasing the slag viscosity was reported several times [69, 70]. SPL is added in amounts of between 5-25 kg/t pig iron [69] and the SPL refractory part and bath material dissolves in the slag. Olsen [70] points out that the slag is designed so that any leaching of environmentally dangerous elements is minimised.

Rusal [71] signed a cooperation agreement with the administration of the Kemerovo Region on recycling waste to innovative products for ferrous metallurgy. In a first step the Novokuznetsk Aluminium Smelter will supply up to 3,000 tpy of SPL to the environmental regional centre for processing.

For the manufacture of special steels von Krüger [12] proposes to use only the carbon-rich first cut SPL material. Volynkina and coworkers [72] see the possibility to use first cut material as a fuel additive.

2nd cut special products

Istomin [73] performed an in depth literature review on the recycling and use of fluorine containing wastes from the aluminium industry, and found out that the refractory-rich 2^{nd} cut of SPL can be used in the production of aluminium silicon alloys. Moxnes et al. [74] also used 2^{nd} cut material and tried this in the manufacture of Al-Si alloys directly in the electrolysis pots. Crushed refractory-rich SPL is mixed with alumina but does not exceed about 15 wt % that of normal industrial smelter grade alumina. This means that large and concentrated batches should be avoided.

Using 2^{nd} cut material in foam-silicate manufacture was proposed by Proshkin et al [75]. This can be used as thermal insulation filling powder and as raw material for the manufacture of nonformed thermal insulation items.

And the Alouette Aluminium Smelter in Canada [76] uses refractory material of spent potlining for the manufacture of special movable concrete blocks, used for bordering highways, but also used as removable walls for rapid access to buildings' basements.

Conclusions

There are now several thermal and chemical processes available for processing and treating spent potlining. The most basic of these seem to be inertisation followed by landfill, but this does not seem an efficient and sustainable solution. A better way is to cooperate with cement manufacturers, who can add spent potlining to both reduce fuel input and to improve slag conditions. The Rio Tinto Alcan LCL & L process has been developed and an industrial pilot plant is now in the start-up stage and expects reach its optimal drive to process up to 80,000 tpy of spent potlining in the near future.

A million tonnes per year of SPL represents a considerable environmental hazard and an expense to contain or neutralise. But it also represents a great opportunity to use or recycle SPL as a resource.

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