DEALING WITH SPENT REFRACTORY MATERIALS

by

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ABSTRACT

The disposal of spent refractory materials has become a major problem for metals producers. The main issue surrounding refractory disposal is the elimination of the hazardous components of refractories, most commonly chrome-bearing materials and fluoride and cyanide in spent pot-liners. The various approaches to pollution prevention involve re-design to eliminate or significant reduction in wastes, recycling or reuse of the materials, and the least valuable approach, abatement. In the present work, the current status of spent refractory disposal is related to the various pollution prevention approaches.

INTRODUCTION

As disposal costs escalate and hazardous waste regulations become increasingly stringent for a wide array of industrial wastes, industry is beginning to recognize the potential returns of investing in a pollution prevention policy. The most effective approach to prevent pollution, which often results in the greatest savings, is to design for zero waste; that is, develop a process which does not create (or significantly reduces) the waste material, or creates no unusable byproducts. The next best policy requires a fundamental shift in thinking as it demands that waste streams be thought of as economic and material resources as opposed to economic and societal burdens. Spent refractories and potliners, like virtually all wastes, potentially offer resource opportunities such as valuable material reclamation, and recycling. The least valuable approach, abatement, aims to eliminate any negative impacts of the waste material prior to disposal.

Refractories and spent potliner solid wastes are two significant environmental issues currently facing metals producers. Efforts are being made to reduce the waste, or eliminate its hazardous components. In this paper, the various approaches to pollution prevention will be described. With each approach, some current pollution prevention practices for metallurgical refractory and spent potliner will be discussed. The present work provides examples of current efforts, and discuss further possibilities for the future. A methodology is given for implementation of the zero waste approach to pollution prevention.

CURRENT STATUS

Pyrometallurgical Refractories

The annual production of refractory raw materials in the United States alone is over 3 million metric tonnes, representing a value of $2 billion (US$)[1]. Typically, refractory materials for pyrometallurgical operations are composed of MgO, Al_2O_3, CaO and other stable, high melting temperature oxides. In addition to these matrix materials, other components are added to alter the refractories physical or chemical properties. These additives include graphite, tar and pitch resin, chromite ore, and various silicates among many others. Together there are over 10,000 refractory products available for use in the metals, glass, chemical and petroleum industries. The iron and steel manufacturers consume about 50 percent of the refractories while the non-ferrous sector uses another 7 to 8 percent[2].

Currently, virtually all of the refractory materials employed in the metallurgical industries are disposed of in landfill sites. The main environmental issues regarding the disposal of pyrometallurgical refractories concern chromite as well as any other materials which inevitably infiltrate into the brick during operation. Chromium is the target of stringent government regulation due to the highly toxic nature of hexavalent chromium (Cr^{6+} or CrO_3). Although the chromium contained in refractories is associated with complex, stable, solid oxide solutions, and is unlikely to leach into groundwater, regulations stipulate that these materials must be disposed of in a hazardous landfill site. The costs of this disposal are rapidly increasing due to the difficulty in opening new sites and anxiety over future liabilities. The primary environmental issue regarding landfilled refractories is the materials that are entrained from the operation. For example, a refractory brick employed in a copper smelter will contain various sulphides, arsenic and selenium among many others. An additional environmental issue with refractories involves the high energy consumption required for their manufacture.

Spent Potliner (SPL)

Spent potliners are a significant concern for the aluminum industry. Spent potliner (SPL) is the cathode lining removed from the electrolytic cells used to produce the aluminum metal, and comprises mainly two layers, carbon block and refractory brick materials. The carbon acts as the cathode at which the aluminum accumulates, while the refractory brick provides insulation and strength. The average potliner must be replaced approximately every 6 years, due to deterioration of the materials. The aluminum industry generates up to 35 kg of spent potlining per ton of aluminum produced from its smelting operations[3]. Over 500,000 tonnes per year of SPL are generated worldwide[4].

Spent Potlining is considered hazardous due to soluble cyanide, fluoride and metal carbides which infiltrate the material during aluminum production. They are a concern due to the possibility of leaching to the groundwater system surrounding a landfill. SPL is also known to generate explosive gases (hydrogen, methane and ammonia) when wet. During aluminum production, the refractory bricks are slightly contaminated with fluoride, while cyanide, carbides and fluorides are found in the carbon lining layer.

Spent Potliners continue to be a significant waste challenge to aluminum producers. Pawlek reported in 1993, that 61 percent of spent potlining was landfilled as is, while another 17 percent was stored in expectation of recycling[5]. However, spent potliner has value for recovery and reuse of chemical elements (F, Al, C, Na) as well as its energy (C) values. The development of new regulations for spent potlining (SPL) means that the percentage landfilled will likely decrease in the near future. As of January 1997, all spent potlining will require a pre-treatment prior to landfilling, providing a strong incentive to aluminum producers to eliminate hazardous aspects of the liner and identify recycling opportunities[6].

DESIGN FOR ZERO WASTE

The most obvious way to eliminate costly disposal fees and other serious health and social issues surrounding waste is to avoid creating it. While it requires a significant emphasis on research and development of new technologies, it is, in the long-term, the most cost effective and environmentally advantageous route for industry. Sustainable design emphasizes energy and resource efficiency, reducing waste, material recyclability, and reduction of persistent toxic substances. As new designs are proposed, they need to be examined from a life cycle point of view. What materials are most environmentally benign from mining through to end use? Can the design allow for ease of reuse or recycle, so that components remain in their life cycle? Although certain materials may have excellent technical properties and price, is it possible to use the next best material that has fewer environmental impacts (including costs), without compromising the product integrity?

Sustainable design positively impacts the community and the bottom line. Landfill and disposal expenses are avoided, while capital expenditures (for the new design developed, or for the recycling process) are recovered by reclaiming valuable components of process by-products.

Reduction in Amount of Refractories Consumed

The amount of refractories consumed annually in the operation of a metallurgical vessel has been decreasing for many years. Figure 1 presents the progressive decrease in the refractory consumption rate in the U.S. steel industry. The amount of refractories consumed per ton of steel has decreased from 27 kg in 1970 to 17 in 1992[7]. At one major Canadian integrated steel producer, the amount of refractories purchased has decreased from 25,000 tons in 1993 to 16,000 tons in 1995. The refractory wastes decreased from 10,000 tons to 5,430 tons over the same time period. In another example, LTV's Indiana Harbor Works has increased their BOF furnace availability from 78 percent in 1984 to 97 percent in 1994[8]. One of the factors contributing to these consumption reductions is improved operating practice through the use of slag splashing. This procedure involves rapid blowing of nitrogen through the lance and into the BOF. The slag is 'splashed' onto the sides of the vessel, forming a protective solid layer which reduces the erosion. Similarly, in the non-ferrous sector, copper coolers are employed to reduce the temperature of the refractory wall such that the protective slag layer forms and the life of the refractory lining is extended.

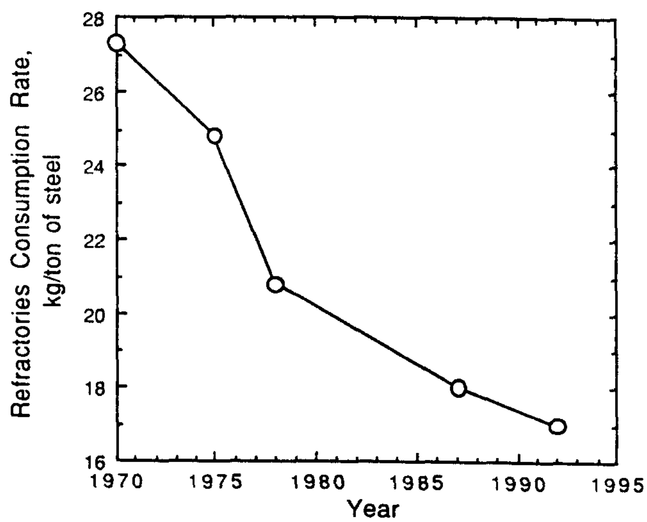

Figure 1. Refractory consumption rate in the U.S. Steel industry from 1970 to 1992[7].

In another operating practice which extends refractory life, producers are attempting to saturate the slag with magnesia (MgO) to reduce the basic refractory dissolution into the slag. This magnesia can either come from cleaned spent refractories or from raw materials. At Inco's Port Colborne refinery, the furnace life was extended by about 25 percent after a practice of adjusting the flux to produce a benign slag[9]. The magnesia content of the slags must be balanced with other metal making demands such as viscosity and liquidus temperature. The goal is to maintain the highest MgO content possible while still meeting the operational requirements of the vessel.

The longevity of refractory linings has also been extended by improvements to the bricks and materials themselves. Mechanical, thermal and chemical properties of the bricks are now becoming better understood and can be optimized for each application. Finite Element Analysis (FEA) is emerging as a powerful tool in the design of refractory linings, leading to extended lining life. This trend is expected to continue as new physical property data becomes available and increasingly sophisticated computer tools are developed.

Reduction/Elimination of Chromite Ore Use

The annual consumption of refractory grade chromite ore over the last 40 years is presented in Figure 2. The consumption has decreased from highs of over 400,000 metric tonnes per year in the mid-1950's and 60's to the present day usage of about 25,000 tonnes[1]. This reduction in the chromite ore consumption is due to the virtually universal change in the steelmaking industry from employing magnesite-chromite (Mag-Chrome) bricks to magnesite-carbon (MgO-C) bricks.

Although the consumption of Mag-Chrome bricks in the non-ferrous industries has decreased, the bricks are still predominately used in smelting and converting operations. Currently, an alternative brick which does not contain chromite, but performs as well as the standard Mag-Chrome is being sought. One possibility is the magnesite-alumina ($MgO-Al_2O_3$) spinel brick. Since the chromite spinel is added to the magnesite to improve the alter the thermal expansion properties, an alternative spinel such as $MgAl_2O_4$ should suit the same purpose while eliminating any hazardous waste concern. The spinel brick is currently being employed in some applications, but has yet to replace the Mag-Chrome brick to any substantial degree. Two problems exist with spinel as a refractory material. First, spinel is relatively expensive in comparison to chromite and secondly, metallurgical slags have a much higher capacity to dissolve alumina than chromium oxide.

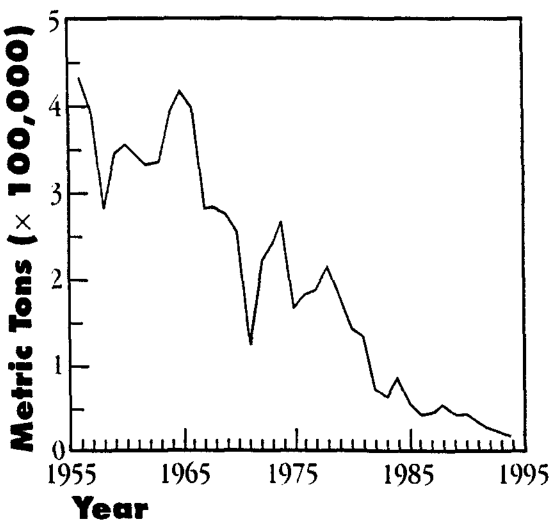

Figure 2. Refractory grade chromite ore consumption in the United States from 1955 to 1995[1].

<u>**Extending Potliner Life**</u>

Unlike iron and steel refractories which can be designed to eliminate the chromium hazard by substituting new materials, sustainable design investigations for potliners must focus on methods to eliminate infiltration of nitrogen (which reacts to form cyanide) and fluoride into the potliner.

The longevity of potlinings can vary over a wide range. Pots may last as little as two years, or as long as ten years, with an average life of 6 years[10]. In the late 1980's and early 1990's, literature reported that potlining waste was generated at a rate of approximately 39.6 kg/tonne of aluminum produced (range of 30 to 50 kg/tonne)[5], whereas reports from 1993 onwards suggest a range of 13 to 35 kg/tonne[3,10,11]. Figure 3 predicts the reduction in generation of SPL in Europe for the next 15 years[11].

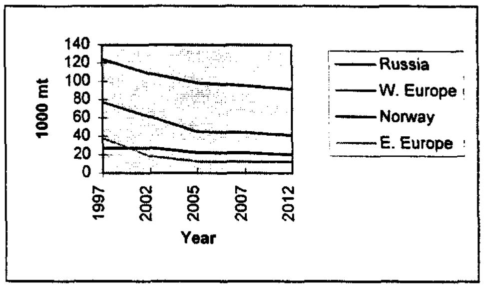

Figure 3. Predicted generation of Spent Potlining in Europe between 1997 and 2012[11].

Efforts to extend pot life have focused on making the potliner more resistant to chemical infiltrations by the electrolyte and molten solution. Use of diffusion barriers, which aim to inhibit and postpone the inevitable penetration of fluorides, nitrogen and other elements into the pot lining, is one possibility. Using effective barriers at appropriate depths in the potliner may result in only one layer (i.e.- the carbon layer) being infiltrated, allowing effective reuse/recycle of the lower layers, such as thermal insulation. Barrier materials investigated include grafoil with or without metal sheet, mild steel sheet, lapped or continuous, pyrex glass cullet, alumina containing powders and glasses, etc[5]. Mathematical models to optimize chemical and mechanical design as well as operation conditions are also used to extend potlife.

Other design suggestions for extending potliner life include the use of graphitized cathode blocks instead of amorphous carbon blocks. Investigations have shown the improved electrical conductivity and higher resistance against the relevant wear mechanisms of graphitic and graphitized cathode blocks versus amorphous blocks[5]. This design option might ideally be used in conjunction with a suggested recycle option of heating the SPL at very high temperatures (2000°C) to obtain a graphitic pure material[12].

RESOURCE RECOVERY AND RECYCLING

Almost every output stream can be seen as an input to another process, instead of a costly waste disposal issue. In some cases, the by-product may require reprocessing prior to input to another process; other times, the by-product can be reused as is. Frequently, the components of a waste stream (metals, salts, etc.) which are the cause of costly hazardous disposal requirements are in fact valuable resources worth recovering. The existence of companies devoted solely to resource recovery and recycling, such as Laidlaw, Philip Environmental and others indicate the profitability of this pollution prevention approach.

In this paper, recycling is defined as the act of re-processing the material such that it can be reused, avoiding its disposal. Recovery refers to the retrieval of valuable components of the refractory and potlining material.

Recycling and Reuse of Refractories

Refractory recycling has been limited by a number of complex materials issues. A significant amount of research and development work is required to design the recycling process and to find applications for the renewed refractory materials. The use of the recycled product for refractories is not usually a viable option due to high shipping and processing costs and changed material chemistry. In 1990, less than 45,000 tonnes of refractories were recycled by metals producers, representing less than 2 percent of total production[13].

There are some recycled refractory materials which have potential as a raw materials. Treated high-alumina brick can be employed to enrich the alumina content of firebrick; lower grades of alumina can be included in monolithics such as gunning mixes and castables; and firebrick can be recycled into grogs for new brick and insulating powders[14,15]. Refractories with high raw materials costs such as silicon carbide, zirconium oxide and chrome oxide can be economically recycled.

A typical beneficiation flow sheet is presented in Figure 4. One of the challenges facing refractory recyclers is that each product must undergo different processing, some more complex and expensive than others. To recycle Mag-Chrome bricks the product is separated into two products after the crushing and grinding stages. The first product is the high iron containing (and thus magnetic) chromite, which is separated by a magnetic separation line. The remaining material is MgO-rich and is then employed as a sand substitute in concrete. Both streams are washed to remove impurities. The separated chromite is recycled for use in the Mag-Chrome brick.

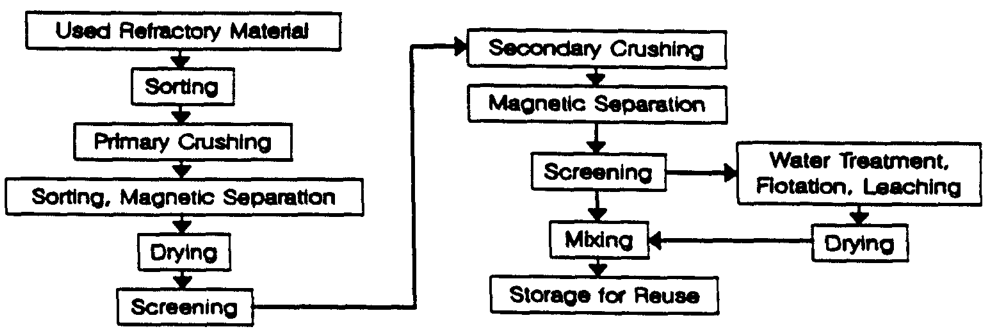

Figure 4. A typical refractory recycling flowsheet[1].

Contamination of the bricks is the largest impediment for large scale reuse. Bricks are contaminated during their lifetime by the melts with which they are in constant contact. Any elements within the melts can be considered to be within the spent refractories, in various minor concentrations.

Refractory reuse is increasingly prevalent as manufacturers realize that it is an economically and environmentally sound alternative to landfilling. In 1993, a major integrated steelmaker reused 32 percent of its refractory waste. This had increased to over 80 percent in 1995, leaving only about 1000 tons for transfer to a waste disposal facility. In this process, alumina bricks are cleaned to remove impurities, particularly metallics, crushed and screened. The coarse material is then sent to the blast furnace while the fines are employed in sludge thickening. Similarly, magnesia brick are cleaned, crushed and added to the slag in the steelmaking process to enhance the lining coating operation.

Design for Efficient Removal and Uniformity of Potlining

SPL is a challenging material to reuse and recycle. The composition of SPL is highly variable, requiring treatment options to be versatile enough to handle a wide variety of compositions. Table 1 lists the range of spent potliner constituents. Once failure of the pot occurs, the spent liner is removed from the cell in several layers by mechanical digging. The first cut consists primarily of a carbon rich fraction which has absorbed the majority of the cyanide and carbide contamination. The second cut consists primarily of thermal insulation weakly contaminated with fluorides. The material size ranges from slabs of several square metres down to sub-micron dusts. The difficulty of treating this material is enhanced by the presence of immeasurable amounts of cathode bar and aluminum[16].

Material	1st Cut Carbon Material	2nd Cut Refractory Material	Range
Carbon	60	2	13 - 69
Fluorides	14	20	7 - 22
Sodium	12	15	9 - 22
Aluminum	10	19	7 - 22
Silica	2	7	1 - 11
Calcium Oxide	1	2	1 - 2.6
Cyanides	0.2	.002	0.002 - 0.6
Iron	2	3	0.3 - 2.8

Table 1. Composition of Spent Potlining (mass percent) [5].

Although not widely reported in the literature, efforts to enhance separation of the layers (carbon rich and thermal insulation) would benefit all aluminum producers. Easily separated potliner layers could lead to treatment streams which independently eliminate cyanide from the carbon layer, recover fluorides, and reuse slightly contaminated insulation. Recycling costs would be reduced if treatment methods could concentrate on smaller ranges of material compositions.

Fluoride Recovery from Spent Potliners

Various SPL treatment methods attempt to recover fluoride. The recovery processes are essentially stabilization techniques; however, aluminum producers are attempting to find uses for the stabilized products. Alcan International Ltd.[3] and Comalco Aluminum Ltd.[16] have developed a caustic leach and lime hydrometallurgical process to recover fluoride in the form of CaF_2 from the crushed SPL. Options for recycle and reuse of CaF_2 (commonly known as fluorspar) include:

- conversion to AlF_3 for direct recycle back to the electrolytic cells [16],
- addition to cement kiln charges to reduce the clinkering temperature,
- substitute for fluorspar in fluxing applications.

New technology in the recovery of fluoride compounds from low grade calcium fluoride waste has been developed in both the nuclear waste[6] and petro-chemical waste industry, and may have applications for SPL. As mentioned above, SPL can be used in the iron and steel industry as a replacement for fluorspar (CaF_2) as flux in steelmaking[17]. Although demonstrated to be possible, this option is likely not viable as it is very difficult to maintain stable operating conditions due to the variability of SPL compositions.

Energy Recovery from Spent Potliners

Spent potliner has substantial energy and material value which could be utilized. The carbon portion can contain over 3800 kJ/kg of potential energy. Cyanide is destroyed in a variety of heat recovery processes, and is not considered a concern if SPL is added as a fuel supplement for cement kilns and mineral wool smelters[18]. However, other industries are reluctant to burn a material classified as hazardous because of permitting and liability issues, which has substantially reduced untreated SPL's use as a fuel supplement. There may be opportunities to recover energy from treated SPL which has obtained a non-hazardous designation.

ABATEMENT

Abatement refers to the elimination of negative impacts of a waste material, so that it can be safely, or more safely be disposed of. It does not aim to recover any of the material or energy value of a process by-product, and accordingly is of least value to the industry producing the waste.

Destruction of Cyanides in Spent Potliners

Virtually all treatment processes for SPL begin with the destruction of cyanides that have infiltrated the material. Cyanides are known to decompose at high temperatures, and rotary kiln, fluidized beds, cement kiln combustion, and calciners have been investigated for their thermal destruction capabilities.

The Spent Potlining Insolubilization Technology (SPLIT) process is strictly a stabilization process[10]. Ground SPL is thrown into a hot air flow vortex rotating at supersonic speed, along with an additive of calcium sulphate or gypsum. The rapid temperature change is sufficient to destroy cyanides and the fluorides react with the additive to produce an insoluble compound. The treated spent potliner is then safe for disposal. Reynolds adds lime and additive to SPL and thermally treats it in a rotary kiln[6]. Cyanides are oxidized and destroyed, and fluorides react with lime to form fluorspar (CaF_2). Reynolds reports that the product of the process has been de-listed by the EPA and thus, can be landfilled, although it is approximately two times the original volume of SPL waste. Reynolds is also pursuing opportunities to reuse the non-hazardous material as aggregate for asphalt paving, brick and block manufacturing, steel desulphurization, and use as a flux in the production of some types of glass-based specialty products.

Comalco Aluminum Ltd.'s COMTOR process employs calcination, in which leachable and complexed cyanides are thermally oxidized[16]. Its advantage over other thermal processes is that it requires no additives (to eliminate agglomeration) in the cyanide destruction process, thus avoiding the large volume increase of treated material, which subsequently occupies more landfill space if not reused. Essentially, Comalco's, Alcan's and Reynolds' processes are stabilization techniques, and until a useful market is found for the treated SPL or fluorspar, the material is sent to landfill.

ZERO WASTE IMPLEMENTATION

The "zero waste" approach is being adopted by many of our clients in the ferrous, non-ferrous and mineral processing industries[19]. It acknowledges that the reduction, recycling and recovery of wastes discharged by a production process is good for a business' bottom line as well as the surrounding community's well being. A successful zero waste program has five key factors:

- Total commitment from the highest levels of management,
- Cross discipline teamwork,
- Clear-sighted identification of areas which provide environmental and economic opportunities,
- Objective process evaluation,
- A continuous improvement outlook.

Identify Pollution Prevention Opportunities

Identifying opportunities can be the most difficult portion of the pollution prevention program since it involves challenging basic assumptions about current production processes. The processes, materials and operating practices in all plants have usually evolved over the years without any formal review. Process areas that currently pose a significant problem with respect to environmental compliance and/or costs are good places to start.

Prepare Process Flow Diagrams: Process flow diagrams and energy balances for the chosen processes should be prepared, indicating all raw material, products and waste streams. The information required is already available within the company's Title V, Clean Water Act and Emergency Response and Community Right to Know Act documents. This information can also be used to determine current emissions, identify major costs, and track progress from year to year.

Assign Costs to Waste Streams: It is crucial to identify all costs associated with a waste stream. Costs for waste disposal, regulatory compliance, and an allowance for future liability where applicable should be included. An attempt must be made to evaluate those unquantifiable costs associated with some process wastes; estimates and clearly articulated assumptions are a reasonable approach.

Review Process to Identify Causes of Waste: Often the causes of waste are poor housekeeping, operational negligence, poor maintenance, or choice of materials.

Identify Opportunities: Pollution prevention opportunities may require operational controls, recycling programs, replacement of hazardous materials, e.g. solvents, reuse of waste or creation of a salable by-product and improved housekeeping.

SUMMARY

Unfortunately, abatement and subsequent landfilling continues to be the pollution prevention policy of choice for many companies. Efforts to stabilize SPL and spent refractories are useful and necessary as they reduce (or preferably eliminate) the impacts of the material to the environment.

However, an effective pollution prevention strategy for refractories and spent potlinings eliminates the wastes, or converts them to a profit-bearing material resource. Beginning at the design phase, processes can be developed or modified such that refractory materials employed are not hazardous nor become hazardous during normal use. The elimination of chromite, design for an inert liner composition and ease of component separation should continue to be explored. Developments in computer modeling tools (Finite Element Analysis) and improvements in the resistance of liner materials are continuing to extend refractory and potliner life. As the zero waste implementation methodology indicates, identifying pollution prevention opportunities involves challenging basic assumptions about current production methods that have evolved over many years.

Effective environmental policies will also involve techniques which include material reclamation, recycling and reuse of the refractory and SPL materials. Reclamation of chromium and fluoride from refractories and SPL are positive examples.

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