Applying Industrial Ecology to Address Environmental Concerns Associated with Aluminum Smelter Spent Potlining

Bernie Cooper, Regain Technologies Pty Ltd, Australia

Abstract

Production of primary aluminium metal with the Hall-Heroult process involves electrolytic reduction of alumina in cells or pots. The electrolyte is made up of molten sodium aluminium fluoride (cryolite) and other additives and is contained in a carbon and refractory lining in a steel potshell. Over time, the effectiveness of the lining deteriorates and the lining of the pot is removed and replaced with a new lining. The lining material that is removed from the pots is known as spent potlining (SPL).

When SPL is first recovered from the pot it contains substances that are toxic and that present environmental hazards. With changing environmental regulatory requirements, primary aluminium smelters are faced with increasing expectations that they will find alternatives to landfilling and/or long-term storage of SPL. SPL has become a major environmental concern.

Separately, manufacturers of energy intensive products such as cement and clay bricks are faced with increasing energy costs and societal expectation of reduction in carbon dioxide emissions. SPL is rich in substances that have beneficial energy saving and carbon dioxide emission reduction properties when used in cement and clay brick manufacture. However, the hazards associated with SPL and its highly variable nature have prevented realisation of the potential energy savings and CO_2 emission reduction. A new level of industrial sustainability emerges when:

1. The hazards in SPL are addressed, and
2. The SPL materials are reprocessed into forms that enable predictable and reliable beneficial effects in the complex chemical processes at the heart of cement and clay brick making.

This paper sets out a solution to the problems associated with SPL developed using the principles of industrial ecology. Industrial ecology uses the natural environment as a model for solving environmental problems. In the natural environment, the waste from one species is food for other species. Industrial ecosystems are based on reconceptualising waste as products. Industrial ecology fosters broad stakeholder engagement with a synthesis of environmental imperatives, technological innovation and business economics.

This SPL solution has been proven over ten years with more then 90,000 tonnes of SPL reprocessed from four aluminium smelters in Australia. All of the SPL is incorporated into products with genuine value in energy intensive cement and clay brick manufacture such that there is economic gain, reduced energy usage and CO_2 emission reduction. Regulatory acceptance has been gained with recognition that the SPL is being managed as a byproduct of aluminium smelting rather than a waste with all of the SPL transformed into manufactured products with genuine downstream industrial use and with no residual waste material.

Introduction

A schematic view of a Hall-Heroult process aluminium reduction cell is shown in Figure 1.

The cathode for the electrolytic process is a carbon lining made up of pre-formed carbon cathode blocks. Other carbon materials line the sides of the pot. Refractory materials generally sit between the carbon cathode and the steel potshell. The lining materials removed from the steel potshell are known as SPL.

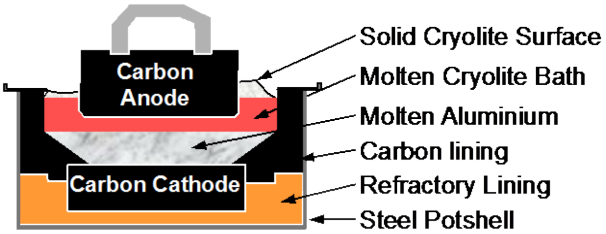

Figure 1 – Aluminium Reduction Cell Simplified Cross Section

Primary aluminium smelters are faced with increasing expectations that they will find alternatives to landfilling and/or long-term storage of hazardous waste materials. Sørlie and Øye[1] refer to environmental agencies in an increasing number of countries defining SPL as hazardous material. Certain waste materials such as SPL are hazardous because of the presence of cyanide compounds, soluble fluoride and a propensity to combine with water and generate explosive gases. These materials are subject to close regulatory control including the Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and their Disposal (the Basel Convention).[2]

Pawlek[3] noted that about 25kg of SPL results from each tonne of aluminium metal produced and that, while in the past, most of the SPL has gone to landfill, "*this practice must change if the industry wants to claim a reasonable degree of sustainability and environmentally tolerable emissions.*"

Separately, manufacturers of energy intensive products such as cement and clay bricks are faced with increasing energy costs and societal expectation of reduction in carbon dioxide emissions.[4,5] SPL is rich in substances that have beneficial energy saving and carbon dioxide emission reduction properties when used in cement and clay brick manufacture. However, the hazards associated with SPL and the highly variable nature of the material have thwarted realisation of the potential energy savings and CO_2 emission reduction.[6,7]

A further obstacle to the sustainable realisation of the potential value in SPL lies in the regulatory and broader community perceptions regarding SPL. Referring to the regulatory situation in the United States of America, the Cement Industry Environmental Consortium referred to a “Regulatory conundrum”.[7] To the mind of a concerned stakeholder, who may not be fully informed on SPL, an Internet search of terms such as spent potlining, spent potliner and SPL presents a story of legacy pollution, heavy regulatory control and an apparently intractable problem.

This paper sets out a solution for SPL based on the following key elements.

1. Technology to eliminate the cyanide and explosive gas hazards in the SPL materials making them safe to transport and use.
2. Utilisation of the chemicals and minerals in SPL to deliver energy savings and CO_2 emission reductions in cement and clay brick industries. This requires refining the recovered SPL material and using the refined material to manufacture products that bring predictable and reliable beneficial effects in the complex chemical processes that are at the core of processes such cement and brick making.
3. Application of the principles of industrial ecology to guide the technical development, business viability, engagement with regulatory authorities and engagement with other stakeholders.
4. Regulatory acceptance and recognition that the SPL has been transformed into inherently valuable products such that there is no longer a hazardous waste that requires waste regulation.

The Nature of Aluminium Smelter SPL

SPL is particularly hazardous due to:

- Its propensity to combine with water and generate flammable gases[1,6,8]
- Presence of cyanide at toxic levels [1,3,6,9]
- Presence of leachable fluorides[3,6,9]

In its raw form, SPL varies in size from fine dust to lumps of up to one metre (**Figure 2**). It typically presents a wide range in mineral and chemical composition as different materials in the pot lining are mixed together.

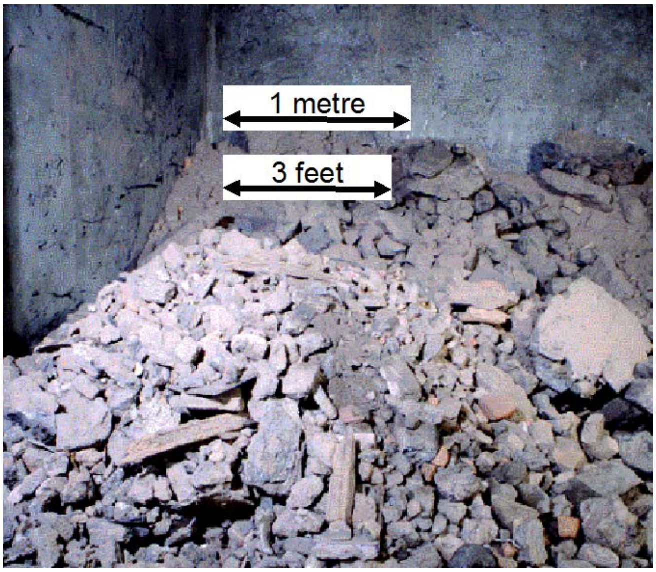

Figure 2 – Raw SPL

Over the typical life of a pot (three to ten years), materials such as aluminium metal, calcium, fluorides and sodium infiltrate the cathode lining and cause it to deteriorate.

Øye[6] makes the observation that there is complex chemistry in the pores in the carbon. Sørlie and Øye[1] and, also, Hop et al[10] provide an appreciation of the type and complexity of the chemical reactions that take place in the linings of an aluminium reduction cell over the life of the linings and describe the mechanism by which the lining is progressively penetrated by sodium metal, the sodium aluminium fluoride/sodium fluoride electrolyte and aluminium metal.

Table 1 sets out analysis of SPL based on progressive sampling of 90,000 tonnes of SPL from four smelters in Australia between 2002 and 2012.

Table 1 – Range of Chemical Composition of Recovered SPL

Description			Carbon Lining	Refractory Lining
Carbon		%	30 to 50	2 to 7
Silicon	as SiO_2	%	2 to 15	10 to 40
Iron	as Fe_2O_3	%	2 to 4	1 to 6
Aluminium	as Al_2O_3	%	10 to 30	15 to 30
Calcium	as CaO	%	1 to 5	1 to 5
Magnesium	as MgO	%	0 to 2	0.5 to 2.5
Sodium	as Na_2O	%	3 to 25	5 to 30
Potassium	as K_2O	%	0.2 to 1	0.5 to 1.5
Fluorine	Total	%	5 to 15	5 to 15
Cyanide		mg/kg	100 to 4,000	100 to 1,500

Chemical reactions result in the formation of various carbides, nitrides and cyanide within the pot linings (e.g. refractory lining, carbon cathode, carbon sidewalls). Indicative examples of the chemical reactions are:

- metals such as aluminium react with nitrogen to form nitrides e.g. $2Al + N_2 \rightarrow Al_2N_2$
- metal oxides also react with carbon to form carbides e.g. $2Al_2O_3 + 9C \rightarrow Al_4C_3 + 6CO$
- various carbon-nitrogen compounds are also produced in the forms of cyanides, in both simple and complex forms.

When the linings are removed from the pot, the resultant SPL contains aluminium metal, sodium metal, carbides, nitrides and cyanides. SPL readily absorbs atmospheric water (humidity), which reacts with these components. Typical chemical reactions of metals or chemical compounds with water that produce explosive gases are:[1]

- aluminium metal to hydrogen $2Al + 3H_2O \rightarrow 3H_2 + Al_2O_3$
- aluminium carbide to methane $Al_4C_3 + 12H_2O \rightarrow 4Al(OH)_3 + 3CH_4$

Regulatory Perspective

Regulation and regulatory attitudes regarding SPL vary between regulatory jurisdictions. The general trend is away from disposal and towards recovery and recycling. An example of this trend is in the 2008 European Union Directive on waste,[11] which in Article 5 sets out the following conditions for developing criteria for a substance resulting from a production process being regarded as a byproduct rather than a waste:

- Further use of the substance or object is certain
- The substance can be used directly without any further processing other than normal industrial practice
- The substance is produced as an integral part of a production process
- Further use is lawful, i.e. the substance fulfills all relevant product, environmental and health protection requirements for the specific use and will not lead to overall adverse environmental or human health impacts.

Article 6 of the EU Directive sets out the conditions for development of criteria for certain specified waste to reach end of waste status:

- The substance or object is commonly used for specific purposes
- A market or demand exists for such a substance or object
- The substance or object fulfills the technical requirements for the specific purposes and meets the existing legislation and standards applicable to products
- The use of the substance or object will not lead to overall adverse environmental or human health impacts.

The input from regulatory authorities in Australia to an organisation formulating an SPL solution is summarised as follows:

- Reluctance to support any SPL solution that has residual material that must be landfilled or diluted with other substances
- Sensitivity to potential community reaction associated with SPL reprocessing facilities and associated with cement plants and brick plants that use products derived from SPL
- Reluctance to issue approvals for transboundary shipment of SPL under Basel Convention protocols unless no other option is available.

Value from SPL for Cement and Clay Brick Manufacturers

The key constituents in SPL that can provide value for cement and clay brick manufacturers are:

- carbon as a source of thermal energy
- sodium that acts as a flux
- fluorine that acts as a mineraliser for cement
- alumina and silica that are useful raw materials.

A flux is a substance that lowers the temperature at which solid materials enter a liquid phase and/or increases the quantity of liquid at a given temperature. A mineraliser is a substance that accelerates reaction rates and promotes the formation of desired materials in the liquid phase of cement clinker or similar materials. A mineraliser results in a higher quality product with reduced residence time and less energy needs for processes such as cement clinkering.[12] [13]

Cement Industry

Cement manufacture involves the decomposition of limestone in a high temperature manufacturing process, typically 1,400 degrees Celsius.[14] Each tonne of cement results in between 0.8 and 1.0 tonne of carbon dioxide (CO_2) emission. Cement production, using current technology, generates in the order of 5% of CO_2 emission induced by human activity.[15] This presents a significant challenge for the cement industry in terms of society's expectations for an urgent reduction in the rate of CO_2 emission.

Manufacturers of cement are facing increasing energy costs along with increasing expectation that they reduce the embodied energy in their product and reduce CO_2 emission. Cement is typically made of energy intensive cement clinker and a relatively small proportion of other less energy intensive minerals. Increasing the proportion of low energy intensity minerals in cement delivers a double benefit in CO_2 emission reduction:

- less CO_2 from decomposition of limestone in the cement raw feed
- less CO_2 as a result of less energy required per tonne of final cement product.[16]

This practice is known as clinker substitution and is measured in terms of the clinker to cement ratio or the clinker factor. Clinker substitution is a key initiative in cement industry CO_2 emission reduction.[4] Many cement companies are now setting reduced clinker factor targets to help focus on reducing energy costs and carbon dioxide (CO_2) emissions. Clinker substitution allows for increased cement production for low marginal cost with significant financial gains in many cases.

Fluxes and mineralisers increase the rate of the reaction of the formation of tricalcium silicate, the key active ingredient in cement clinker. This means that a more reactive clinker can be produced with higher tricalcium silicate levels allowing increased substitution of clinker with lower cost materials in composite cements. Fluorine is a widely used and effective mineraliser. Sodium fluoride and cryolite have been used to achieve fluoride mineralisation of cement clinker.[12]

Clay Brick Industry

Clay brick production requires heating material to temperatures in the order of 1,000 degrees Celsius. This requires large amounts of energy and significant associated CO_2 emissions. This presents a significant challenge for the clay brick industry in terms of society's expectations for an urgent adjustment to the rate of CO_2 emissions.[5]

Sodium is an effective flux, lowering the firing temperature required to achieve a given quality of clay brick product and delivering energy savings and CO_2 emission reductions.[17] [18] Research in Australia in the early 1990s showed that the sodium in the refractory in SPL is an effective flux.[19]

Strategy for an SPL Solution

The inherent hazards, highly variable nature and complex chemistry of SPL mean that developing viable and practical solutions to the SPL problem has proved to be difficult. The primary aluminium industry and its support industries have invested significant effort into the research and development of SPL solutions with limited success. The success of potential SPL solutions has hampered by one or more of the following aspects:

- Complex process chemistry
- Technical risk in process technology
- Capital intensive process equipment and high operating costs
- Quantities of residual waste material well in excess of the original quantity of SPL processed
- Unknown fate of the fluorine when disposed of by other parties.

The strategy for the SPL solution that is the subject of this paper was formulated with three key inputs:

- Lessons learned from prior industry experience with SPL solutions[20] [21] [22] [23] [24] [25]
- Support of three aluminium smelting organisations with smelters in Australia
- Increasing interest in energy reduction and CO_2 emission reduction in the cement and clay brick industries and knowledge that the chemicals and minerals in SPL and other residual materials from primary aluminium smelting could enable substantial gains in energy and CO_2 emission reduction for these industries.

The following objectives were identified:

- 100% beneficial use of SPL with no residue
- Low cost of SPL re-processing
- Lower energy consumption and greenhouse gas emissions for manufacturing products such as cement and clay bricks
- Positive impact on the environment

The following strategy was formulated to achieve the above objectives and to manage environmental, regulatory and associated financial risks.

1. Develop and apply a chemical process to eliminate the explosive gas and cyanide hazards at the smelter, i.e. detoxify hazardous or contaminated minerals on the smelter site so that only products are shipped from the smelter, not hazardous waste materials.
2. Secure support of regulatory agencies, environmental groups and community stakeholder groups through:
    - Effective communication to ensure stakeholders are informed and understand recovery and recycling initiatives and to create an objective understanding of SPL
    - Understanding of the environmental gains (energy saving & greenhouse gas reduction) that flow from the beneficial fluxing effects of sodium and fluorine in energy intensive industries.
3. Use industrial mineral trading and marketing to identify markets, develop processes to use the products in target industries and to promote and distribute products. Encourage the cement and clay brick industry to see the economic value in energy saving and to move away from seeking rent to process hazardous waste with the attendant regulatory and community issues.
4. Gain classification of the manufactured products derived from SPL and other smelter by-products as products and not as hazardous waste. Thus the products are not subject to onerous regulatory requirements. In this way, the SPL solution is set up to avoid the generation of a hazardous waste and meet the by-product requirements of the EU directive on waste.[11]
5. Establish and maintain integrity of the overall system that supports the SPL solution by ensuring all materials are accounted for by involving only reputable and trustworthy service providers and end-user customers.

SPL Processing

The strategy for the SPL solution required chemical processes and technology that can handle the highly variable properties of SPL. An overview of the process for refining SPL and manufacturing new products that are not classified as waste materials is shown in Figure 2.

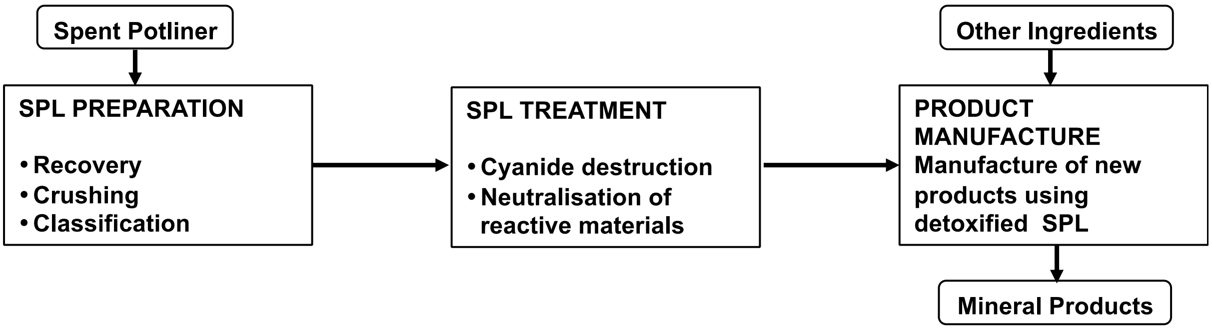

Figure 2 – SPL Processing Flow Diagram

SPL Preparation involves (a) recovery of material from storage or directly from pots, (b) primary segregation of aluminium metal, carbon materials and refractory materials, (c) sorting into like material streams and (d) crushing and size classification in preparation for further processing.

The cyanide and explosion hazards in the SPL are eliminated through the **SPL Treatment** process. Cyanide destruction is achieved by thermal oxidation i.e. heating the material in the presence of oxygen. Neutralisation of the reactive compounds and aluminium metal is achieved by using water to bring on the reactions that generate the explosive gases in a controlled environment in special apparatus such that no more gas can be generated.

Indicative chemical reactions that take place in SPL treatment process are:

- Cyanide to carbon dioxide & nitrogen $C_2N_2 + 2O_2 \rightarrow 2\ CO_2 + N_2$
- Reaction with water, generating flammable gases as described earlier
- Combustion of flammable and other gases - e.g.
    - methane to carbon dioxide and water $CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$
    - ammonia to nitrogen and water $4NH_3 + 3O_2 \rightarrow 2N_2 + 6H_2O$
    - hydrogen to water $2H_2 + O_2 \rightarrow 2H_20$

Mineral products with fluxing and mineralising properties are **manufactured** by processing the detoxified SPL material and other ingredients.

Valuable Mineral Products

Small amounts of fluorine and sodium can have a significant effect in cement clinker manufacture. Mineralisation of cement clinker can be achieved with addition rates of fluorine of 0.2% of clinker. Similarly, effective fluxing of clay in brick making is achieved by adding sodium (as SiO_2) at the rate of 0.2% of raw material. The savings in energy and greenhouse gas emissions are substantial. However, variation in dosing rates of these chemicals can cause process upsets.

Realisation of the potential value in the chemicals and minerals in SPL requires high quality products with homogeneous properties manufactured to tight specifications. These products can then be dosed into cement clinker plants and clay brick plants in a way that delivers predictable and reliable beneficial effects on process chemistry and reliable production outcomes.

Industrial Ecology

Implementation of the proposed strategy required constructive engagement with a wide range of disparate stakeholders. A common theme was needed where each stakeholder group could see their particular interests and concerns being addressed.

The concepts of industrial ecology were used to set the framework and tone for engagement with aluminium smelters, regulatory agencies, community stakeholders and potential users in the cement and clay brick industries. Industrial ecology provides the basis for informed, objective and balanced consideration of the issues around SPL. The industrial ecology approach was instrumental in gaining the necessary support for the initiative. This approach enabled a progressive shift in mindset from SPL as a hazardous waste to a mindset of SPL as a potentially valuable resource to be mined for economic and environmental benefit. It provides the basis for maximising the value in SPL and minimising the costs of meeting environmental requirements regarding SPL.

Industrial ecology enables a synthesis of environmental imperatives, technological innovation and business economics. In a natural ecosystem, the waste of one species is food for other species. Tibbs[26] explains that industrial ecology uses the natural environment as a model for solving environmental problems and that "..*the key to creating industrial ecosystems is to reconceptualize waste as products*." Tibbs states that industrial ecology is the subject of active study and promotion by reputable research organisations and proposes the idea that industrial ecology provides a coherent framework for shaping and testing thinking around environmental issues for industry.

Fiksel[27] describes the concept of industrial ecology as providing *"…a useful systems perspective to support sustainable development while assuring shareholder value creation*." Fiksel suggests that industrial ecology provides a framework to address the scope and complexity of industrial sustainability issues with a set of design principles that generally lead to positive outcomes. Erkmann[28] provides a more comprehensive statement of the scope of industrial ecology.

"Industrial ecology does not just address issues of pollution and environment, but considers as equally important, technologies, process economics, the inter-relationships of business, financing, overall government policy and the entire spectrum of issues that are involved in the management of commercial enterprises."

Van Berkel[29] puts the argument that industry is a primary agent for environmental improvement as it has the technological, managerial and other resources necessary for environmentally informed design of products and processes. Jelinski et al[30] emphasised that industrial ecology is proactive, not reactive, in that it is initiated and promoted by industry.

A particular boost to the emergence of industrial ecology came with a 1989 article in Scientific American by Frosch and Gallopoulos,[31] who put forward the concept of an industrial ecosystem as a more integrated concept than the traditional industrial model of raw materials being used to make products for sale and waste to be disposed of. Frosch and Gallopolous made the further observation:

"Corporate and public attitudes must change to favor the ecosystem approach, and government regulations must become more flexible so as not to unduly hinder recycling and other strategies for waste minimization."

An Industrial Ecosystem

The SPL solution is based on a multi-party system (Figure 3) comprised of:

- Participating primary aluminium smelters
- SPL process plants provided to the smelters as part of the solution
- Manufacture, marketing and distribution of products derived from SPL that are valuable in energy intensive processes such as brick and cement manufacture
- Cement manufacturers
- Clay brick manufacturers
- Enabling structure that integrates:
  - technology to detoxify SPL
  - technology to use the products in cement and clay brick manufacture
  - regulatory approvals for technology and products
  - trading of manufactured products
  - optimised marketing and logistics of products
  - access to research and a body of knowledge to support use of the products

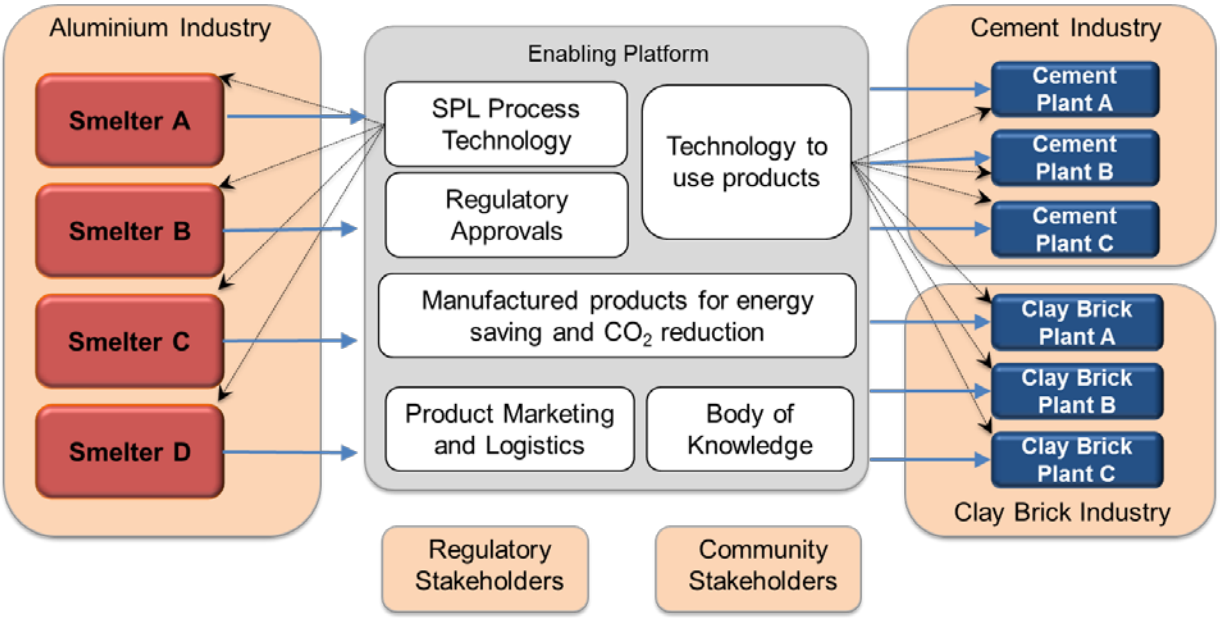

Figure 3 – Industrial Ecosystem for SPL

The system described above has been developed over a period of ten years. It provides SPL handling services for four aluminium smelters in Australia. More than 90,000 tonnes of SPL have been processed through this system. Access to this system is provided as a service to the participating aluminium smelters. In this way, the SPL problem is "taken care of" and the smelter organisations do not have to manage the complexity of capital investment in SPL process plant, plant operations and marketing final products. With SPL processing plants provided at the smelter site SPL is refined on site and there is no risk to the smelter associated with hazardous waste outside its immediate control.

The innovation and uniqueness of the SPL detoxification technology has been recognised with patents on the technology granted in Australia, Canada, New Zealand, South Africa and the United States of America.

Manufacturing and marketing quality, branded products with inherent energy saving and CO_2 reduction value for the cement and clay brick industries has seen:

- Regulatory acceptance that the products derived from SPL are not hazardous waste and do not need to be classified and regulated as hazardous waste
- Market acceptance that the products are valuable, with the market not seeking rent for processing hazardous waste.

The products derived from SPL are not classified as hazardous waste by:

- Australian Federal Government environmental regulator and Basel Convention Consenting Authority
- Australian states of New South Wales, South Australian and Victoria environmental regulators
- Chinese Government environmental regulator
- Philippines Government environmental regulators
- Government of Thailand pollution control and industrial works regulators

Conclusion

Given the changing regulatory conditions facing the aluminium smelting industry whereby the past practice of land filling SPL must cease in an increasing number of regulatory jurisdictions, there is a strategic opportunity to establish a best practice economic and environmental solution for the aluminium smelting industry that is attractive to environmental regulators and offers significant economic gains to the cement and clay brick industries.

The strategy of refining the SPL and using it with other aluminium smelter by-products to manufacture safe, high quality products that are not classified as hazardous waste and that deliver energy saving and greenhouse gas reduction to other industries and has seen the development of a sustainable industrial ecosystem around SPL. With the appropriate formal regulatory recognition in place, these products can be traded and transported freely and safely across the globe as non-dangerous goods.

Applying the principles of industrial ecology enabled informed, objective and balanced consideration of the issues around SPL and resulted in constructive engagement with a range of stakeholders. It provides the ongoing basis for maximising the value in SPL and minimising the costs to the primary aluminium industry of meeting environmental requirements regarding SPL.

This solution for SPL is an innovative model of environmental sustainability with gains for the aluminium industry, for end-users of products and for the environment. This solution was established over a period of ten years with four aluminium smelters in Australia and the transformation of more than 90,000 tonnes of SPL into valuable products. It provides smelters with a comprehensive, low risk and economic solution for SPL.

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