



Review

Recycling of refractory bricks used in basic steelmaking: A review

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ABSTRACT

Refractories are indispensable for all high temperatures processes, such as the production of metals, cement, glass and ceramics. It is estimated that up to 28 million tons of spent refractories are generated every year. Despite these significant amounts, recycling of spent refractories has received little attention due to the abundance of low cost virgin raw materials and low disposal costs of the, largely inert, materials. In the last two decades, recycling of spent refractories has started to receive more attention due to environmental considerations and increasing costs for landfilling. However, recycling in applications such as road bed foundations or slag conditioners does not capture the full intrinsic value of the materials. Higher value recycling as refractory raw materials is much more limited, and estimated at only 7% of refractory raw material demand. Recently, rising prices and supply issues for high quality virgin raw materials have created a strong incentive for closed-loop refractory recycling. This review gives an overview of the history of refractory recycling and the main refractory recycling applications, with a particular focus on recycling in new refractories. Current spent refractory processing in view of raw material recycling is discussed, and an outlook is given to future trends and developments.

1. Introduction

Refractories are solid materials that can withstand high temperatures and maintain their mechanical function for a required period of time under all circumstances, even in contact with corrosive liquids or gases. Refractories are indispensable for all high temperatures processes, such as the production of metals, cement, glass and ceramics.

An enormous variety of refractories exists, designed to meet the temperature and process requirements of each application. Refractories can be classified in numerous ways, with the most common being based on method of installation (shaped or unshaped), type of bonding (tempered, fired) and chemical composition (acid, basic or neutral) (Fang et al., 1999).

Shaped refractories or bricks are pressed in a predefined geometry and installed as such, whereas unshaped refractories, commonly referred to as monolithics, are provided in powder form and shaped on-site during installation by pouring, troweling, gunning, ramming, vibrating and injecting (Fang et al., 1999). In the group of shaped refractories further distinction can be made between ceramic and carbon bonded bricks. Ceramic bonded or fired bricks are formed at high temperatures (1500 °C) using temporary binders and a sintering process, while carbon bonded or tempered bricks are formed at lower temperatures (300 °C) using hydrocarbon binders (resin, pitch, oil, ...)

and final strength is developed during in situ firing.

The classification into acid, basic or neutral is based on the interaction of the main raw material with water. Acid refractories such as alumina-silicate materials, silica and zircon are typically used for lower operating temperatures than other refractories and tend to be much less expensive to produce (Fang et al., 1999). Basic refractories, including magnesia, doloma and spinel, are often combined with carbon and graphite and used in highly basic environments. They can withstand the highest operating temperatures but are susceptible to hydration and therefore require appropriate handling. Neutral refractories, such as chromia and alumina refractories, are used extensively by the metal industries because of their high melting temperature, moderate price, and ability to be used in both acidic and basic environments (Fang et al., 1999). Since the production of chromia refractories has declined due to environmental concerns, alumina refractories are the most readily available neutral material.

Based on a recent overview (IMFORMED, 2016) (Table 1), the most used refractories worldwide are refractory clays (46%) and magnesia-based refractories (26%). Refractory clays are used in a wide variety of applications and industries, while magnesia refractories are very important for the steel industry. Doloma, although a minor player worldwide (3%), is strongly linked to the stainless steel industry, where doloma refractories have largely replaced magnesia-chrome bricks

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Table 1
World refractory raw materials consumption (Source: (IMFORMED, 2016)).

Raw material	% of world refractory raw material consumption	Primary source country
refractory clays	46%	China
magnesia	26%	China
recycled refractories	7%	
calcined bauxite	4%	China
brown fused alumina	3%	China
doloma	3%	USA
tabular alumina	2%	
calcined alumina	2%	China
graphite	1%	China
calcium aluminat cements	1%	
sillimanite minerals (incl. andalusite)	1%	South Africa
chromite	1%	South Africa
white fused alumina	1%	China
zircon	1%	Australia
silicon carbide	0.25%	China
silica	0.25%	USA
spinel	0.25%	China
olivine	0.25%	Norway

outside of China.

Worldwide refractory production is around 35–40 million tons per year, with annual fluctuations determined largely by the iron and steel industry that is responsible for up to 70% of the total demand. During the use phase, 30–40 wt.% of the refractory is consumed (Nakamura et al., 1999), indicating that up to 28 million tonnes of spent refractories are generated every year.

Despite these significant amounts, recycling of spent refractories has in the past received little attention due to the abundance of low cost virgin raw materials and low disposal costs of the, largely inert, materials. Historically, the problem of refractory waste generation was mainly dealt with by decreasing the refractory consumption per ton of product. In the steel industry, refractory consumption has declined from 25 to 30 kg of refractory per ton of steel in 1970 to about 8 kg/t today in the US and Japan (Domínguez et al., 2010; Koros, 2003). A similar trend is observed for other industries, such as the cement industry where consumption has decreased from more than 2 kg refractory/ton cement clinker to on average 0.9 kg /ton, and even to less than 0.2 kg/ton in the most modern kilns (Guéguen et al., 2014).

In the last two decades recycling of spent refractories has started to receive more attention due to environmental considerations and increasing costs for landfilling.

The waste hierarchy, as included in the latest version of the European Commission's Waste Framework Directive 2008/98/EC (EC, 2008), sets a priority order for waste management, namely 1) prevention 2) preparing for reuse 3) recycling 4) other recovery (e.g. energy recovery) 5) disposal. One of the shortcomings of the hierarchy is that it does not differentiate between different types of recycling to maximize the inherent value of waste. In closed-loop recycling, the inherent properties of the recycled material are not considerably different from those of the virgin material. The recycled material can thus substitute virgin material and be used in identical products as before. In open-loop recycling, the inherent properties of the recycled material differ from those of the virgin material in a way that it is only suitable for use in other product applications, mostly substituting other materials (Huysman et al., 2015). Closed-loop recycling has the potential to generate higher economic benefits, while contributing to the evolution towards a circular economy. In a circular economy, as defined among others by the Ellen MacArthur Foundation, the goal is to keep the functionality and therefore value of a material as high as possible over an as long as possible time period (EMF, 2015). Therefore, the waste hierarchy itself is not sufficient to achieve absolute reduction in material throughput in the economy (dematerialization). A value-based

concept of waste with a stricter specification regarding open- and closed-loop recycling is one of the suggested solutions to improve the use of the hierarchy (Van Ewijk and Stegemann, 2016).

Spent refractories have been used largely in open-loop recycling applications such as roadbed aggregates (Domínguez et al., 2010) and slag conditioners in the steel industry (Conejo et al., 2006; Lule et al., 2005; Nakamura et al., 1999). In such applications, the value of the recycled refractories is however limited to the cost of the replaced material, e.g. less than 20 USD/ton for roadbed aggregates and around 60 USD/ton for fluxes (Lule et al., 2005) while market prices for dead burnt magnesia may be 3–5 times higher.

Higher value recycling as refractory raw materials is nevertheless much more limited, and estimated at only 7% of refractory raw material demand (IMFORMED, 2016; Odreitz, 2016).

Recently, rising prices and supply issues for high quality virgin raw materials have created a strong incentive for closed-loop refractory recycling, and interest for recycling within the refractory producing industry is increasing.

This article gives an overview of the history of refractory recycling and the main refractory recycling applications, with a particular focus on recycling in new refractories. Current spent refractory processing in view of raw material recycling is discussed, and an outlook is given to future trends and developments.

2. Historical evolution

Because of concerns with chromium toxicity, spent-chrome containing refractories were the first to attract recycling attention, with research dating back to the early 1980s (Fang et al., 1999) and a first patent on reprocessing of spent magnesium-chrome bricks into refractory raw materials published in 1985 (Nazirizadeh et al., 1985). Increasing environmental awareness, increasing costs for waste disposal and decreasing availability of space for landfill apparently sparked an interest in recycling of also other refractory types a decade later, with numerous patent applications related to refractory regeneration published after the mid 90s (Table 2). In the US, a larger study to identify recycling options and reduce landfilling, with related publications reporting on the characterization of spent refractories and development of techniques to recycle them was initiated also at this time (Fang et al., 1999; Smith et al., 1999).

The interest in recycling varies widely between countries and regions in relation to the local stress on resources and landfilling options. In Japan, refractory recycling was already studied and put into widespread practice after the oil crisis in the 1970s (Sugita, 2008) with examples of refractory recycling into the production of refractories shown as early as 1999 (Nakamura et al., 1999). At this time, 99% of refractory waste was reported to be still landfilled in the US (Fang et al., 1999), where interest in refractory recycling was limited due to a lack of strong economic or environmental driving forces, given the low landfilling cost of most refractories. The abundance of natural raw materials was considered to justify only simple beneficiation of spent refractories for economic viability (Kwong and Bennett, 2002). Canadian and European experience in refractory recycling was claimed to be ahead of US in 2001 due to stronger environmental regulations and

Table 2
Number of patent filings for refractory recycling by 5 year interval and country.

	Canada	China	France	Germany	Japan	Korea	US	Total
1985–1989				1				1
1990–1994					2			2
1995–1999	1		2	3	3		1	10
2000–2004			1		11	1	1	14
2005–2009		6			11	1		18
2010–2014		20			8			28
2015–2016		10			2	2		14

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