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Assessing the feasibility of silica-based media for coal preparation operations: A novel method of carbon feedstock production for the silicon market

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ABSTRACT

Production of silicon for solar cells and semiconductors requires the use of ultra-clean coal as the main reducing agent in the silicon production process. Most coal preparation plants employ magnetite suspensions as the dense medium in coal cleaning processes. Although downstream operations are designed to remove and recover magnetite, the unavoidable presence of residual magnetite in the final coal product dramatically decreases the purity and suitability of the coal for the silicon market. Given that silicates are the main raw materials in the silicon production, a systematic study through laboratory dense-medium cyclone tests was used to evaluate fine silica-based alternative materials as a substitute for magnetite in coal cleaning operations. These alternative silica materials include byproducts generated during the silicon production process. The results indicate that laboratory tests with silica-based materials were able to achieve similar separation performance when compared to plant operations using magnetite. Following the outcomes of the experimental study, a novel flowsheet arrangement was designed using silica-based media rather than magnetite. If properly implemented, this new separation system can eliminate iron contamination and provide coal that can be marketed as a high-quality silicon feedstock.

1. Introduction

1.1. Background

Silicon is an important raw material for various markets and applications, including the chemical industries, alloy production, and electronics [1]. The end-use and the ultimate value of the silicon are largely driven by the elemental purity as well as the composition of the contaminants [2,3]. Silicon with over 95% purity is deemed metallurgical grade silicon and is produced by reacting silica-rich materials with carbon-rich materials at high temperatures [4,5]. Further processing is often required to produce solar grade and electronic grade silicon which requires over 99.9% Si purity. The cost of the final silicon product can be elevated by a factor of 10 by improving the purity specifications from 0.5–100 ppm to less than 1 ppb [2,3,5]. Over the last decade, metallurgical-grade silicon has doubled in price from \$0.77 per pound in 2005 to a high of \$3.48 per kilogram in 2011 [6,7]. Most recently, metallurgical-grade silicon was valued at \$2.67 per kilogram in 2014 [7].

The most economically viable route to produce metallurgical-grade and electronic grade silicon is by carbothermic reduction of silica-rich materials (e.g., quartz and quartzite) with carbonaceous reduction materials (e.g., coal, woodchips, charcoal, etc.) in a submerged arc furnace. While the industrialized production of silicon is quite complex, the idealized reaction for silicon production is expressed by [2,4,8]:

$$SiO_2(quartz) + 2C(coal, charcoal, woodchips) \rightarrow Si + 2CO. \tag{1}$$

During the silicon smelting process, silica and carbon raw materials are charged into the arc furnace, and the inside temperature is immediately increased to 700–1300 °C by high temperature gases rising in the arc furnace. In this environment, different chemical reactions can occur based upon the carbon coverage,[1] which result in the production of free molten silicon, silica fume and silica slag (Fig. 1). The production of the latter two materials is undesirable and results in a reduction of final silicon recovery [1,9].

The presence of metal contamination in the refined silicon product results in poor performances and efficiency degradation in devices and materials constructed from silicon. Of all the metal impurities, iron is one of the most troublesome and must be minimized throughout the

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[1] Carbon coverage may be defined by parameter x in the reaction between silica–rich materials and carbon reduction agents as follows: $SiO_2 + xC$.

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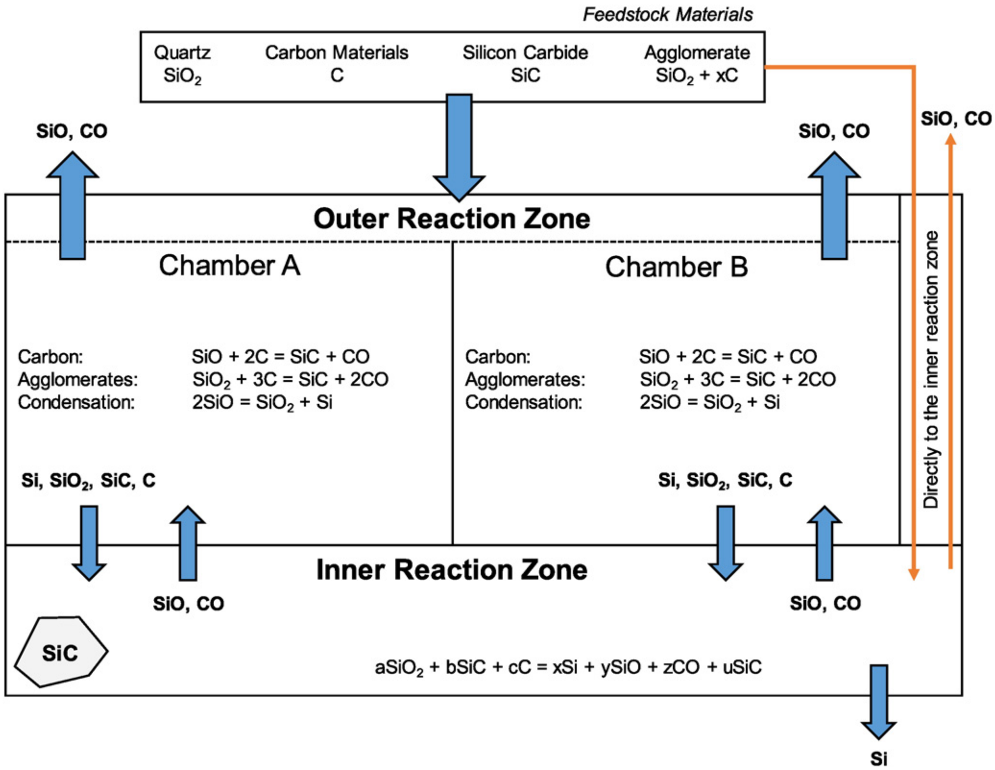

Fig. 1. Stoichiometric model illustrating chemical reactions and material flowing in the submerged arc furnace (after Schei and Larsen [23]).

production process [3,10,11]. Unfortunately, iron is one of the most abundant elements in nature and is difficult to entirely remove once it is in the silicon production line. Due to the high boiling point temperature of elemental iron, more than 99% of total iron in the raw feedstock material will ultimately condense into the final silicon product [12]. For electronic and semiconductor applications, iron is considered the most challenging impurity due to its high electrical activity. As a result, the maximum allowable level of iron contamination in electronic-grade silicon is decreasing annually as smaller electronic devices are increasingly sensitive to trace iron contamination [11,13]. Consequently, to ensure the highest purity of produced silicon, iron must be completely removed from the feedstock material.

1.2. Role of coal preparation

As shown in the idealized reaction (Eq. (1)), production of metallurgical-grade silicon requires the use of ultra-clean coal. Coal is the main reducing agent in the silicon production and accounts for 26% of raw material used in the silicon smelting process [1]. Coal preparation technology plays a significant role in the silicon market by upgrading run-of-mine (ROM) coal to satisfy the particle size and quality specifications. Modern coal preparation plants utilize a complicated arrangement of solid–solid and solid–liquid separation processes designed to improve the purity of coal thereby increasing the inherent value and suitability for downstream usages. Coal preparation plants can be designed and operated to serve a number of different downstream applications and markets, including power generation, coke production and silicon production [14–16].

The dense-medium cyclone (DMC) is one of the most widely applied gravity concentration units employed in the coal preparation process to clean a major portion of the plant feed mass. The geometry of the unit induces a centrifugal field which causes differential settling of particles with contrasting densities. High density particles settle quickly to the outside of the unit and are expelled through the underflow while low density particles are retained with the air core and expelled through the overflow (Fig. 2). Traditional DMCs use a suspension of finely ground magnetite to control the medium density and the separation cutpoint. In conventional applications, DMCs are used to remove high density impurities from ROM coal particles greater than 1 mm. The separation in a DMC strongly depends on the weight distribution in the coal as a function of particle density and size, as well as the stability and rheological properties of the medium [16,17].

After the cleaning operation, the purified coal material is passed over fine drain-and-rinse screens. In this stage, water sprays are used to remove and recover the magnetite, which is then recirculated through the plant [18]. Despite this additional cleaning stage, small quantities of ultrafine magnetite remain on the surface of final clean coal product.

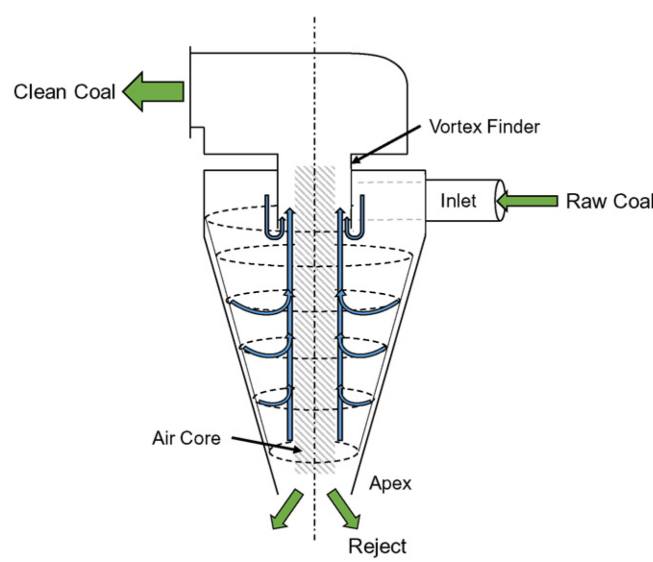

Fig. 2. Schematic of internal flow for a typical DMC (after Sanders [24]).