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Influence of carbothermic reduction on submerged arc furnace energy efficiency during silicon production

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ABSTRACT

In this study, we investigated the exergetic efficiency and thermal energy source of the off-gas system of a submerged arc furnace, which varied from 27% to 35% and 47% to 55%, respectively, of the total energy supply. We used a case study to evaluate the thermal exergy in the off-gas of a real furnace, which exhibited an additional power capacity up to 2.7 MW amounting to 10% of the total energy supplied to the process (or 23% of the electrical power fed to the furnace.) We also determined the perfect negative correlation coefficients (*r* as the standard) between the exergetic efficiency and raw material consumption via linear regression and observed moderately positive relevance between power consumption and raw material consumption in the furnace. We attributed this correlation to increased graphitization and reduced resistivity of carbonaceous materials as the charging began sink slowly into the reaction zone and the charging temperature increased. Compared to coal, petroleum coke showed a significant impact on total power consumption according to the linear regression results; especially in regards to the fact that petroleum coke underwent graphitization more easily than coal as charging temperature increased.

1. Introduction

With the rapid economic development and the continuous improvement of living standards, there has been substantial and rapidly increasing demand for energy. The primary global energy demand is now and can be expected to remain overwhelmingly dominated by fossil fuels. To this effect, the effective utilization of alternative energy sources such as solar and wind has become crucial.

Solar energy is one of many sustainable and clean energy sources [1–3], but it can be challenging to harness. Industrial silicon is necessary as initial material to produce pure silicon for photovoltaic and semiconductor materials. Currently more than 80% of commercial solar cells are made of silicon, particularly multi-crystalline silicon [4–6]. It is commercially produced through the reduction of silicon oxide (silica) with carbon materials in submerged arc furnaces [7]. The electrical energy consumed by silicon production represents about 45% of the total energy supplied to the industry, while the raw materials account for the remainder. Exergy analyses of industrial silicon furnaces have shown overall exergy efficiencies of 33% [8], where the remaining energy leaves the process as thermal energy in the cooling water, i.e., hot off-gas, by radiation and convection from the furnace and from the liquid silicon cooling process [9]. Improving energy efficiency in silicon production, particularly methods that recover energy effectively, is a major issue that remains to be addressed.

There are methods of exergy analysis that have been well established [10,11] and applied to a wide range of industrial processes [12–21]; research has even examined the effects on national economies [22,23] and the human body [24] via exergy analysis. Within the steel industry, to further decrease electrical energy

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consumption, researchers have explored a wide array of techniques for optimizing processes: measuring various melting stages and electrode positions [25], for example, as well as understanding the influence of gas burners [26] and raw materials [27,28] on energy efficiency.

Despite the many valuable contributions to the literature (only in terms of industrial steel production) there have been relatively few reports on the production process of silicon though it is also an energy-intensive sector. Hjartarson [29] published an interesting thesis on the exergy analysis of silicon production, and other researchers [8] examined the recovery of thermal energy in the off-gas in both an ideal, theoretical and actual industrial silicon production process, to establish numerical standards for high exergy efficiency. They found that exergetic efficiency during real-world production is 0.33 ± 0.02 without power recovery and 0.41 ± 0.03 when waste heat is utilized; in the ideal process, exergetic efficiency of 0.51 and utilization of thermal exergy in an 800 °C off-gas increased the exergetic efficiency to 0.71 [8]. Other researchers have evaluated the exergy of the silicon furnace using a set of two performance indicators: overall exergy efficiency and silicon yield exergy [30]. The exergy efficiency they identified (0.30) coincides with real-world reported values for exergy efficiencies, indicating that an exergy loss of about 70% is typical for today's standard furnace operation. In terms of making the silicon production process as eco-friendly and cost-effective as possible, it is vital to effectively understand the process parameters that can decrease the specific power energy consumption of the furnace.

In this paper, we examined SAF energy balance values calculated from actual collected plant data as well as energy balance values complied from the literature. Our primary objective was to evaluate the thermal exergy in furnace off-gas (using previously reported methodology [8]) to establish workable energy and exergy analyses for real-world furnaces. The work extends preliminary reports on exergy [8,31] and energy analyzes [30,32,33], especially the source of chemical energy of the off-gas system which is often neglected. Several other important factors that influence energy consumption and exergy are related to raw materials, which we also took into account when developing our proposed technique. Raw materials can affect exergy and decrease the specific power energy consumption of a silicon furnace, or can increase the maximum overall exergy efficiency, but researchers tend to neglect them. We analyzed multiple periods of operation with a mixture of petroleum coke and coal as carbonaceous raw materials and determined the correlation coefficients between exergetic efficiency and raw material consumption according to the production process of an actual silicon furnace.

2. Internal energy balances of the submerged arc furnace

The theoretical silicon production process is generally considered to run on pure silicon dioxide and carbon (graphite) in the furnace, plus electrical energy. The carbothermic reduction reaction of silicon dioxide is as follows:

$$SiO_2 + C = Si + CO_2 \quad (1)$$

A previous paper [30] contains a short description of the actual production process focusing on material and energy flows. The furnace reaction in the industrial silicon production process can be written in a simplified form as follows:

$$SiO_2 + (1+x)\,C = x\,Si + (1-x)\,SiO\,(g) + (1+x)\,CO\,(g) \quad (2)$$

where x is the silicon yield, $0 < x < 1$. Electric power is required to achieve and sustain the high temperatures (above 1800 °C) necessary for the reduction reaction. The main inputs are silica or quartz (SiO_2) and carbonaceous reduction (a mixture of coke, coal, charcoal, and woodchips.) Carbon material is added to the process through the consumption of carbon electrodes and moisture, as well as the introduction of volatile and ash material.

The main features of the theoretical silicon production process, first simplified and described by Schei et al. [34], is a suitable reference for the real-world industrial process. (Please refer to Fig. 1.). The two main inputs of carbon, electric energy, and silicon oxide are charged from the top of furnace. The temperatures above 1800 °C necessary for silicon production are achieved by inputting large amounts of electric energy. Carbonaceous materials play two roles during production: As a partial source of energy by burning the carbon materials, and (mainly) as reductants involved in the carbothermic reduction of silicon oxide.

The two main outputs of materials, liquid silicon tapped from the bottom of the furnace and off-gas (SiO + CO), are burned with excess air from the furnace hood, which is captured into the gas-cleaning system and filtered. The temperature of liquid silicon at tapping is 1600 °C initially, then as it is poured into molds it cools to 25 °C and is crushed into desired particle sizes. The off-gas temperature is controlled by the amount of excess air in the system. Off-gas particles (also called “condensed silica fume”) consist mainly of SiO_2. Because the off-gas is mixed and burned with excess air before escaping the furnace hood at high temperatures (200–700 °C or higher) it is the key to generating electric energy through waste heat recovery/utilization with a steam turbine and generator system.

It is also inevitable that thermal energy is lost as the liquid silicon cools, both in the cooling water and by radiation and convection from the furnace body. Previous researchers [9] have estimated the heat lost by radiation and convection to be 5% of the total energy input to the process. The enthalpy and exergy in the slag and amorphous SiO_2 has been estimated to be less than 0.5% of the total enthalpy and exergy leaving the furnace.

Submerged arc furnace energy sources are comprised of electrical energy and energy generated from oxidation-reduction reactions of carbonaceous reductants during the carbothermic silicon reduction process. The submerged arc furnace energy balance is:

$$\begin{aligned} E_{Total} &= \int_{Charging}^{Tapping} P_{Electric}dt + \Delta H_{Carbon} + \Delta H_{Oxidation} + \Delta H_{Volatiles} \\ &= \Delta H_{Silicon} + \Delta H_{Slag} + E_{off-gas} + \int \Delta\dot{Q}_{Cooling}dt + \int \Delta\dot{Q}_{Radiation,\ other\ losses}dt \end{aligned} \quad (3)$$

Carbon-based fuels are numerous, with complex respective