



Co-treatment of spent cathode carbon in caustic and acid leaching process under ultrasonic assisted for preparation of SiC



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ABSTRACT

Spent cathode carbon (SCC) from aluminum electrolysis has been treated in ultrasonic-assisted caustic leaching and acid leaching process, and purified SCC used as carbon source to synthesize silicon carbide (SiC) was investigated. Chemical and mineralogical properties have been characterized by X-ray diffraction (XRD), X-ray fluorescence (XRF), scanning electron microscopy (SEM), and thermogravimetry and differential scanning calorimetry (TGA-DSC). Various experimental factors temperature, time, liquid-solid ratio, ultrasonic power, and initial concentration of alkali or acid affecting on SCC leaching result were studied. After co-treatment with ultrasonic-assisted caustic leaching and acid leaching, carbon content of leaching residue was 97.53%. SiC power was synthesized by carbothermal reduction at 1600 °C, as a result of yield of 76.43%, and specific surface area of 4378 cm²/g. This is the first report of using purified SCC and gangue to prepare SiC. The two industrial wastes have been used newly as secondary sources. Furthermore, ultrasonic showed significant effect in SCC leaching process.

1. Introduction

Spent Cathode Carbon (SCC) is a hazardous byproduct waste of electrolytic aluminum industry discharge from shut-down cells [1]. Cathode carbon blocks are subject to corrode by high-temperature electrolyte, molten aluminum, metal sodium and other substances, continuously and inevitably, during operation of the cells [2]. Now, life of most aluminum electrolytic cell is about five to eight years [3,4]. SCC is made of carbon, fluorides, and other inorganic impurities. Carbon and fluorides produce the SCC high recovery potential, and cyanides and soluble fluoride salts are the components of greatest environmental concern [5]. SCC seriously affects the health of animals and plants as well as ecological balance [6,7], and many countries have classified it as a hazardous solid waste.

Numerous efforts on SCC have been explored because of its toxicity [8] and recyclability [9]. SCC was used as raw material or additive in some industries, such as cement industry [10], iron and steel industry [11] and copper recovery industry [12]. There are a variety of hydro-metallurgical processes for SCC treatment and recovery, including acid leaching [13], flotation [14], soluble aluminum salt solution leaching [15–17], caustic leaching [18], and co-treatment with acid and alkali [19]. However, the purity of carbon powder obtained in existing SCC treated processes was not ideal due to complex impurities and the lack

of efficient methods. It is imperative to find a new treatment process or a perfect auxiliary method.

The use of ultrasound may improve reaction efficiency because ultrasonic-assisted reactions are often more efficient than traditional methods [20–22]. Ultrasonic assisted leaching is an efficient and economical method and has been widely used in separation and purification processes. Saterlay [23] removed and destroyed cyanide and cryolite in used carbon cathode from aluminum smelting using ultrasound assistance, discovered a faster leaching speed and higher leaching rate of cryolite than traditional leaching. Xiao [24] treated SCC by ultrasound-assisted and traditional caustic leaching methods, resulting in shorter leaching time and higher leaching efficiency in the ultrasound assisted leaching process. Chemat [25] studied the mechanism of ultrasound assisted extraction and investigated ultrasonic effect on higher yielding and purity of artemisinin crystals from *Artemisia annua* L. leaves. Zhang [26] contrasted ultrasonic-assisted and regular leaching of germanium from by-product of zinc metallurgy, found some advantages in ultrasonic-assisted leaching process.

As an industrial waste, coal gangue can be regarded as potential silicon source because it is rich in silicon. With advantages of stable chemical character, high thermal conductivity, low-thermal expansion coefficient, and good abrasion resistance, SiC power has been used widely. Some papers reported that coal gangue was used as raw

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material to synthesize SiC power. Xiao [27] studied the treatment of coal gangue by elevated temperature acid leaching for the synthesis of SiC and the specific surface area of SiC power sintered at 1550 °C was 4000 cm²·g⁻¹. Jia [28] investigated SiC synthesis from coal gangue by carbothermal reduction in N₂ atmosphere at 1400 °C. Han [29] investigated the preparation of β-SiC/Al₂O₃ composite from kaolinite gangue by carbothermal reduction.

At present, there are few studies and reports about the purification of SCC under ultrasonic assisted, and the purity of carbon power obtained from the current SCC treatment processes is unsatisfactory. In this study we try to recycle carbon from SCC by co-treatment with caustic leaching and acid leaching in the presence of ultrasonic, aim at high purity carbon power. Although coal gangue has been used to prepare SiC, there is a blank area of SiC synthesis used the treated SCC and gangue. In this work the feasibility of SiC preparation has been explored by carbothermal reduction with raw materials of purified SCC and gangue.

2. Experimental

2.1. Materials

In this study, SCC was supplied by an aluminum smelter (Sichuan Province, China). The sample was smashed with a jaw crusher and a ball grinder, and then sifted with a 100-mesh sieve. Afterward, sample powder was dried in an oven and then purified in caustic leaching and acid leaching processes. The caustic leaching solution was prepared from a mixture of sodium hydroxide NaOH (analytical grade) and homemade deionized water. The acid was hydrochloric acid (analytical grade).

Coal gangue used in this study was obtained from the Ordos coal mine (Inner Mongolia, China). The purification process has been investigated by the team numbers [27].

2.2. Experimental procedure

2.2.1. Purification of SCC

A 20-g dried sample powder was collected and mixed with alkali and deionized water in a 500-mL Teflon beaker. The obtained mixture was subjected to caustic leaching at different temperatures, times, liquid-solid ratios, initial alkali concentrations, and ultrasonic powers. After the powder was leached, filtered, and washed with water multiple times, filter cake was dried in a vacuum oven at 105 °C for 12 h. The dried cake was purified further in hydrochloric acid solution with identical experimental procedure and apparatuses as in the SCC caustic leaching process. SCC leaching experiments were performed in the presence of ultrasonic wave.

A variety of complex impurities were removed in caustic leaching process and it was difficult to calculate the removal rate of one or two kinds of impurities. Thus, to simplify the analysis course, SCC caustic-leaching results were expressed with F leaching rate and carbon content of leaching residue. Dried leaching residues were incinerated in a muffle stove at 800 °C for 4 h in air. Afterward, the carbon content of leaching residue was calculated using the following Eq. (1):

$$\eta_c = \left(1 - \frac{m_a}{m_s}\right) \times 100\% \quad (1)$$

where η_c is carbon content of leaching residue (%), m_a is ash weight of leaching residue incinerated at 800 °C for 4 h in air (g), m_s is weight of leaching residue incinerated at 800 °C for 4 h in air (g).

F leaching rate was calculated by the following Eq. (2):

$$X = \left(1 - \frac{cv}{m\eta}\right) \times 100\% \quad (2)$$

where X is F leaching rate (%), c is filtrate (g/mL), V is filtrate volume

(mL), m is SCC sample mass (g), η is F content of SCC sample (%).

The acid leaching result of SCC was calculated by Eq. (1), too.

2.2.2. Synthesis of SiC

SiC power was prepared by carbothermal reduction. Purified SCC and gangue were collected and mixed by ball mill for 4 h with moderate absolute ethanol as surface modifying additive. The homogeneous mixture pressed into a cylinder under 25 KN was sealed into a corundum crucible and then sintered in muffle furnace. Following high-temperature treatment, the calcined sample was incinerated at 800 °C for 4 h in air. Afterwards, the ash was purified in hydrofluoric acid solution and multiple washing, dried in the oven. Finally, SiC power was analyzed and characterized.

2.3. Characterization and apparatus

The characterization methods and experimental apparatus used in the work are enumerated.

XRD: The crystallite structures of powders were characterized by XRD with Cu K α radiation in a Rigaku D/Max 2500 diffractometer at a scan speed of 5°/min.

XRF: XRF spectrometer type is XRF-1800, produced by Shimadzu Corporation, Japan, and used for material ultimate analysis.

SEM: The SEM image was characterized using JSW-6360LV scanning electron microscope, produced by JEOL. Ltd.

TGA-DSC: Thermal properties of SCC powder were studied using SDTQ600 thermogravimetric analysis apparatus (TA Co., Ltd., USA). Samples were heated from room temperature to 1000 °C in air and to 1200 °C in Nitrogen, with a heating rate of 10°/min and a flow rate of 100 mL/min.

Ball mill: Sample was milled in ball grinder (QXQM-4, Changsha tencan powder Co., Ltd., China) at the speed of 400 r/min.

Muffle stove: SX2-10-12 muffle, rated temperature 1600 °C (Changsha electric oven company, China).

Ion meter: PXSJ-216 ionmeter (Shanghai hengci Co., Ltd, China).

Ultrasonic equipment: It was an ultrasonic cleaner (KQ-400KDE, Kunshan Ultrasonic Equipment Company, China) with ultrasonic power of 0–400 W and constant frequency of 40 kHz.

3. Results and discussion

3.1. Characterization of SCC sample

Tables 1 and 2 summarize the proximate and ultimate analyses of SCC sample, respectively. The sample XRD pattern and SEM image are showed in Figs. 1 and 2, respectively. XRD pattern reveals that graphite is the main carbon material, about 60% of the total mass of raw material. Inorganic impurities (bright fractions in Fig. 2) include NaF, Al₂O₃, Na₃AlF₆, and aluminosilicate. Clear borders are observed between carbon and impurities. As shown in Fig. 3(a), TGA curve changed slightly when the sample was heated from room temperature to 500 °C in air, which means volatile and water lost in this process. Between 500 and 800 °C, TGA curve declined with increased temperature sharply, an obvious exothermic peak was found in DSC curve and the peak value corresponds to 597.34 °C. It means that in this temperature interval there may be one or more exothermic reactions. The sample mass lost with temperature increase and the mass loss rate was 63.64%, approximately carbon content in sample ultimate analysis. Therefore, the

Table 1
Proximate analysis of the SCC.

Sample	W	FC	V	A
Content/%	1.62	61.37	1.14	35.87

W: water; FC: fixed carbon; V: volatile matter; A: ash.

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