Study on plasma spraying graphite purification device

Jinmao Li$^{1,2}$, Chunlian Song$^2$, Dandan Lu$^2$, Rui Huang$^{1,2}$, Haobo Zhang$^{1,2}$

$^1$Department of electrical and information engineering, Heilongjiang university of technology, Jixi, China
$^2$Heilongjiang provincial key laboratory of plasma biomass materials research and testing, Heilongjiang university of technology, Jixi, China

*Corresponding author e-mail: songchunlian@163.com

Abstract. Given the problems in traditional graphite purification methods, such as low impurity removal efficiency; Using chemical purification method, the reagent is toxic and contaminated, which does not meet the environmental protection requirements; The physical purification method has the disadvantages of high energy consumption and high cost. The plasma spraying graphite purification method is proposed and its mechanism is studied. A prototype of the plasma spraying graphite purification device was assembled and developed. With the built purification device prototype as the research object, the state monitoring and comprehensive evaluation of the working gas flow, system working current, working voltage, carbon powder quantity, and other indicators are carried out. The change of impurity content in graphite was analyzed by the plasma spraying method under different index conditions. Experiments show that the graphite can be effectively purified by plasma spraying graphite purification method, and the carbon content is increased from 94.18% to 97.23%.

1. Introduction

Graphite, known as “black gold”, is a strategic resource$^{[1]}$. With its characteristics of high-temperature resistance, corrosion resistance, good thermal conductivity, and high chemical stability, its application scope covers the core fields of metallurgy, electronics, sealing, chemical industry, national defense, aerospace, military industry, people's livelihood, and so on, which play an important role in the development of national economy and modernization$^{[2]}$.

The main problems of the existing graphite purification technology are: acid-base corrosion reaction equipment, purification by-products or reaction reagents have great environmental pollution, large energy consumption in the purification process, and high cost of special equipment$^{[3, 4]}$. The above problems are determined by the characteristics of the purification method. To reasonably solve the problems in the existing graphite purification, one of the effective ways is to find a more ideal new method for graphite purification based on the existing technology. In this paper, a plasma spraying graphite purification method was proposed, and a prototype of the plasma spraying graphite purification device was assembled and developed.

2. The basic principle of purification

Plasma, also known as the “fourth state of matter”, is a collection of electrons, positive ions, negative ions, neutral atoms, or molecules in which the negative charges of particles are equal to the total number of positive charges. According to the plasma temperature, it can be divided into high-
temperature plasma and low-temperature plasma. Low-temperature plasma can also be divided into thermal plasma and cold plasma. Due to the harsh formation conditions of thermal plasma in a strict sense, it is difficult to obtain thermal plasma in the laboratory or in practical applications, while arc discharge plasma is relatively easy to obtain in industrial production. Arc discharge plasma has high-temperature characteristics, generally between $10^3 \sim 2\times10^4$ K, and has rich chemical properties, strong activity\cite{5}. Due to the above two properties, arc discharge plasma technology has been widely used in organic degradation, pollutant treatment, textile wastewater treatment, arc plasma metallurgy, and preparation of NH$_3$ liquid substances\cite{6, 7}.

The basic principle of the graphite purification method by arc discharge plasma is as follows: the raw material of the graphite to be purified is placed in the high-temperature environment generated by arc discharge plasma, and the impurities in the graphite are collided by using the high oxidizing active particles (O$_3$, ·OH) generated in the process of gas discharge. Under the action of strong oxidation, the impurities become impurity molecules that are easy to remove. In the industrial production process, the purification effect of graphite can also be enhanced by synergistic injection of active substances (such as organics, inorganics, or surfactants). This method takes into account the advantages of the high-temperature method for graphite purification, and at the same time has chemical characteristics, which puts forward a new idea for the graphite purification method. Figure 1 is the process diagram of plasma spraying graphite purification.

![Figure 1. The process diagram of plasma spraying graphite purification.](image-url)

### 3. Structure of Graphite Purification System by Plasma Spraying

#### 3.1. Overall design

The plasma spraying graphite purification system uses the plasma arc as the heat source, and the graphite powder to be purified is uniformly sprayed on the arc plasma flame. The active characteristics of the plasma flame are used to make it fully react with the impurities in the graphite to remove the impurities.

#### 3.1.1. Steps for purification of graphite by plasma spraying

When the graphite was purified by plasma spraying, a DC arc was generated between the cathode and the anode (nozzle), which heated and ionized the imported working gas into high-temperature plasma and sprayed from the nozzle to form plasma flame. Graphite powder is fed into flame by powder feeding gas, in which impurities are melted or reacted to become new impurities easily removed. Then high purity graphite was obtained by recycling device. The working gas can be argon, nitrogen, or hydrogen added from these gases or the mixture of argon and helium.

#### 3.1.2. Characteristics of Purification of Graphite by Plasma Spraying

The device has multiple powder feeding ports, and the active agent can be appropriately added as needed to achieve a better graphite purification effect. Researchers can reasonably select the working gas according to the
different impurity components in graphite. For example, the inert gas can be selected as a working medium to reduce the oxidation reaction of graphite powder during flight.

3.2. The main equipment of plasma spraying graphite purification system

Plasma spraying graphite purification device consists of main power source, transmitting box, liquid chiller, human-machine interface intelligent control system, gas safety center, powder feeder, plasma torch. The schematic diagram of the plasma spraying graphite purification device system is shown in Figure 2.

![Schematic diagram of plasma spraying graphite purification device system](image)

**Figure 2.** The schematic diagram of the plasma spraying graphite purification device system.

3.2.1. Main power source. The main power source of plasma spraying graphite purification system is controlled by the intelligent module, silicon controlled rectifier. The maximum input power is 85 kW and the transient load rate is 100%. The maximum output power is 80KW. Under the condition of ±15% grid fluctuation, the power supply can still work normally, and the current fluctuation range is controlled between ±1% and ±2%. The main power supply can provide DC power for the spray gun, and it can work continuously for 24 hours without fault.

3.2.2. Transmitting box. The transmitting box is the transfer center of the whole spraying system, which connects the power supply, control cabinet, heat exchanger, and spray gun. The box is equipped with high-frequency ignition, water, and gas control alarm devices. Box movable, water cable length 5m. The water and electricity of the plasma equipment are mixed and sent to the spray gun. At the same time, the high-frequency pulse voltage required for the arc of the spray gun and the return water temperature and cooling water pressure of the spray gun is provided. It is lightweight, flexible, and convenient to drag. The good arcing performance can effectively reduce the instantaneous current shock during the nozzle arcing.

3.2.3. Liquid chiller. The liquid chiller is composed of a high-pressure water pump, water chiller, and large stainless steel chilled water tank. The high-pressure pump has 12 kg of pressure. The power of the chiller is 5-10P. The storage capacity of the chilled water tank is 3 tons. The cooling device is used to ensure the stable operation of the thermal plasma generator.

3.2.4. Human-machine interface intelligent control system. The human-machine interface intelligent control system is the control center of the whole equipment, which can realize the automatic control of the whole spraying system. The control cabinet adopts PLC touch control terminal, which integrates operation, setting, help, display, process, and fault alarm monitoring. The device adopts a simplified panel design, which is more convenient to use and more simple to operate.

3.2.5. Gas safety center. Considering that H₂ gas is flammable and explosive, and the use of mass flowmeter (electrical components) to generate electrical sparks will have the risk of combustion and explosion, the “gas safety center” developed and manufactured has the following functions: equipped with gas leakage alarm device (which can detect H₂, N₂, Ar); fast cutting off the inlet pipe device; spray gun tempering prevention, extinguishing equipment; leakage protection, cut off and alarm device; current overcurrent protection device; voltage overvoltage protection device; water leakage...
pressure sensing device; water temperature warning system; dust concentration monitoring alarm system.

3.2.6. Powder feeder. The system is equipped with a double-tube powder feeder. It is a device for storing spray powder and conveying powder to a spray gun according to process requirements. The main gas of the spray gun is used as the powder feeding gas. The powder is transported to the outlet of the powder feeder through the rotation of the powder feeding scraper, and the powder is left in the powder feeding barrel through the gas pressure to enter the spray gun. The double-tube powder feeder is controlled by a PLC touch screen, which can freely program the parameters such as powder flow rate, frequency, and thickness, etc., and the system is highly automated. Powder feeder can send powder particle size range: 5 ~ 200 μm; Repeat error of powder feeding is less than ± 1 %; The two tubes can simultaneously supply powder with a feeding rate of 8 ~ 250 g/min.

3.2.7. Plasma torch. The plasma torch used in this device is a 100 kW multi-mode spray gun, which is widely used in thermal spraying. The plasma spraying gun has two kinds of powder feeding modes: internal powder feeding and external powder feeding. The two internal powder feeding ports can be used independently. If used simultaneously, a higher spraying speed can be obtained. Besides, the plasma spraying gun can also realize the mixed-use of different materials in the equal ion beam. Staff can formulate reasonable powder feeding mode according to the requirements of the graphite purification process. The schematic diagram of the plasma torch structure is illustrated in Figure 3.

Figure 3. The schematic diagram of the plasma torch structure.

4. Experiments and result analysis

With the designed plasma spraying graphite purification device, the influence of various parameters on graphite purity was studied. The experimental sample was 94.18 % flake graphite produced by Jixi Puchen Graphite Co., Ltd. Under the condition of airflow velocity of 20 and feeding velocity of 100, the influence of current and temperature changes on graphite purity was qualitatively analyzed. The temperature is measured by an infrared thermometer. The experimental data are shown in Table 1.

| Current/A | 200  | 200  | 300  | 300  | 400  | 400  | 500  | 500  |
|-----------|------|------|------|------|------|------|------|------|
| Temperature/℃ | 860  | 860  | 1820 | 1820 | 1910 | 1910 | 2070 | 2070 |
| Purity/%    | 95.21| 96.15| 96.99| 97.23| 97.01| 96.48| 97.05| 97.06|

According to Table 1, the trend diagram of graphite purity changing with current and temperature is plotted, as showed in Fig. 4. The experimental results demonstrate that the purity of graphite can be effectively improved by plasma spraying graphite purification device. With the increase of current, the temperature of plasma flame is increasing; In addition to the current and temperature, the purity of graphite may also be affected by other factors, such as air intake, voltage, etc. The mechanism of plasma spraying graphite purification method needs further study.
5. Conclusion

Plasma spraying technology was first applied to the graphite purification process in this paper. The fixed carbon content of the sample increased from 94.18 % to 97.23 %.

Using plasma spraying technology to produce plasma flame heat is highly concentrated. At the same time, the plasma flame contains a large number of strong oxidizing groups, which can effectively remove the impurity content in graphite. The method has the dual properties of physical and chemical purification of graphite and has a very good purification effect of graphite.

The plasma flame atmosphere is controllable. Reducing gas (such as H₂), inert gas (such as Ar₂), or a mixture of various gases can be used as the working gas. It is helpful to analyze the influence of different working gas and mixed working gas on the purification effect of graphite.

The purity of graphite in the purification process is affected by many factors, such as the current size, gas flow rate, powder feeding mode, plasma flame temperature, and so on. It is an important direction for future research to optimize the adjustment parameters and obtain the relationship between the parameters and the purity of graphite.

6. Acknowledgments

This work was financially supported by the National Natural Science Foundation of China (Grant No. 51877024), 2020 Special Foundation Project of Fundamental Scientific Research Professional Expenses for Undergraduate Universities in Heilongjiang Province, the Natural Science Foundation of Heilongjiang Province, China (Grant No. LH2020E111).

References

[1] D.jar Allah, Betemariam Amha, Woldetinsae Girma, et al. Purification, application and current market trend of natural graphite: A review. International Journal of Mining Science and Technology, 2019, 29(5): 671-689.
[2] Xiangwen Zhou, Yang Yang, Jingtao Ma, et al. Effects of purification on the properties and microstructures of natural flake and artificial graphite powders. 2020, 360.
[3] Qi Qi Zhang, Xian Zheng Gong, Xian Ce Meng. Environment Impact Analysis of Natural Graphite Anode Material Production. 2018, 4740:1011-1017.
[4] Allah D. Jara, Jung Yong Kim. Chemical purification processes of the natural crystalline flake graphite for Li-ion Battery anodes. 2020, 25(prepublish).
[5] M. F. Zhukov. Electric arc generators of thermal plasma (review). Plasma Devices and Operations, 2006, 5(1).
[6] Son Byung-Koo, Lee Kyu-Hang, Kim Tae-Hee, et al. Purification and Nitrogen Doping of Nanothin Exfoliated Graphite Through RF Thermal Plasma Treatment. 2019, 9(7).

[7] Shen Ke, Chen Xiaotong, Shen Wanci, et al. Thermal and gas purification of natural graphite for nuclear applications. 2021, 173:769-781.