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Properties of RE–W cathode and its application in electrostatic precipitation for high temperature gas clean-up

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ABSTRACT

In this paper, three rare earth tungsten (RE-W) electrostatic precipitation cathodes (cerium-tungsten (Ce-W), lanthanum-tungsten (La-W) and yttrium-tungsten (Y-W)) were prepared by powder metallurgy. The microstructure and the electron emission properties of the three cathodes were examined. The application of the Y-W cathode in high temperature electrostatic precipitation was investigated. Results show that the addition of the rare earth reduces the tungsten crystal size, with yttrium (Y) affecting the crystal size more significantly than lanthanum (La) and cerium (Ce). The rare earth (cerium, lanthanum or yttrium) exists, in the form of crystalline tungstates, at the tungsten (W) crystal boundary. The effective work function of the La-W cathode has the lowest value of 2.88 eV at vacuum condition, but the Y-W cathode shows the best emission properties at atmosphere pressure. The collection performance of an electrostatic precipitator (ESP) using the Y-W cathode was investigated by ash particles with various resistivity and size distribution at various temperatures. The particles with higher resistivity tend to be collected effectively at high ambient temperature, and the collection efficiency rise with the increase in temperature. If the resistivity of dusts is not high enough, corresponding to the highest collection efficiency, there exists an optimum temperature value. As in a conventional corona ESP, the larger particles are collected more effectively in an ESP with Y-W cathode.

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1. Introduction

High temperature precipitation technology is of great importance for particle collection in not only integrated gasification combined cycle (IGCC) and pressurized fluidized bed combustion (PFBC) technologies applied in power plants but also advanced technologies applied in gasification power plant. In addition, it is also widely applied in chemistry industry where many high temperature gases used as either desiccation or heating media must be purified.

Electrostatic precipitation is a technique to remove suspended particles in a gas using an electrostatic force [1]. It has advantages of lower pressure drop, no jam, high collection efficiency and low operation cost, and thus has been used widely in various industries such as utility boilers, cement kilns. A conventional electrostatic precipitator (ESP) contains sharp discharge electrodes and smooth collecting electrodes [1–3]. When a high voltage is applied to the discharge electrode, a corona discharge takes place. Ions and electrons are produced at the corona point, and ionic current flows

through the space. These ions attach to suspended particles. The charged particles are then moved towards the collecting electrode by a Coulomb force and finally collected on collecting electrodes. The conventional ESPs are usually used at temperatures lower than 673 K, because the range of corona voltage becomes narrow and it is hard to maintain stable corona discharge at high temperature [4,5].

To overcome the drawbacks of traditional ESPs, a new type of ESP based on thermionic emission cathodes was developed [6,7]. This type of ESP uses lower work function cathodes. When used at high temperature conditions, the thermal electrons are emitted from the cathode surface. It is these electrons colliding with gas molecules that produce gaseous ions. The dust charging and collecting mechanisms of this new ESP are basically similar to those in a conventional corona ESP. The stable and sufficient thermionic emission of cathodes is the key to it.

Some studies exist in the literature on finding effective lower work function cathodes that can offer sufficient electron density under high temperature flue gas conditions. Metal oxide composite materials, metal coated with alkaline earth oxide metal (such as BaO) and some other additives have been investigated [8]. However, since the melting points of these materials are low, the

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surface of cathode using this kind of materials may be polluted and corroded to certain extent at high temperature conditions, which will cause physical and chemical change of the materials. In addition, the emitter coating usually falls off easily from the metal substance because of the thermal load. These problems may cause the electron emission ability of an ESP to drop.

Due to its high melting point (3655 K), low vapor pressure and reasonable work function (4.52–4.55 ev), tungsten (W) may be an ideal choice for the cathodes of the above newly developed ESPs. Thoria tungsten (Th–W) alloys were considered as the best thermionic emission sources and have been widely used since 1913 [9]. However, thorium is a radioactive element. Its use may pollute environment and harm human's health. Therefore, many new kinds of materials were developed to replace Th–W. Of them, rare earth tungsten (RE–W) was considered as the best candidate [10,11]. So far, RE–W materials have been used as vacuum electron emitting source, ion source and hot cathode in metallurgy, welding, surface treatment, vacuum electron, etc. However, to our knowledge, RE–W materials have never been used in dust removal at temperatures higher than 1173 K, the operating temperature conditions of PFBC, IGCC and other advanced industrial units.

In this paper, three typical RE–W cathodes, cerium–tungsten (Ce–W), lanthanum–tungsten (La–W) and yttrium–tungsten (Y–W), were selected as the cathode for the newly developed ESP mentioned above. The comparative study on the microstructure and fundamental electron emission properties of the RE–W cathodes was conducted and the ESP dust removing characteristics at temperatures up to 1306 K were investigated.

2. Experimental setup and procedure

The materials for the cathodes were first prepared. Then the emission properties of the materials were measured at vacuum and atmosphere conditions. Finally, the performance of an ESP using the cathode made of one of the prepared materials was investigated.

2.1. Materials preparation

The cathode materials were first prepared by powder metallurgy. Blue tungsten (W_4O_{11}) was doped with three rare earth nitrate solutions $(Ce(NO_3)_3, La(NO_3)_3, Y(NO_3)_3)$, dried and reduced to mixed powder $(W+Ce_2O_3, W+La_2O_3)$ and $W+Y_2O_3)$, respectively. The mixed powder is composed of 97.8 wt% tungsten and 2.2 wt% rare earth oxides (Ce_2O_3, La_2O_3) or Y_2O_3 . The powder was then pressed in steel dies and sintered by electric resistance heating in hydrogen to form three kinds of cathodes (Ce-W, La-W) and Y-W. Their microstructures were examined by X-ray diffraction (XRD) and Optical Microscope (OM).

2.2. Emission property measurements

Since the effective work function is usually used to stand for the thermionic emission ability of a material, the thermionic emission properties of the three RE–W cathodes were first tested in a dynamic vacuum system [12]. The standard parallel-plate diode configuration with adjustable anode- cathode distance was adopted for emission property measurements. The vacuum system was evacuated by a turbine molecular pump and an ion pump. The pressure of the vacuum system was kept at lower than $5\times 10^{-5}\,\mathrm{Pa}$ during the measurements. The electron pulse emission performance at pulse width of 500 ms, repeated frequency of 250 Hz and the highest pulse voltage of 1000 V was tested by a self-designed computer-controlled automatic emission-testing instrument. The anode voltage and the emission current were re-

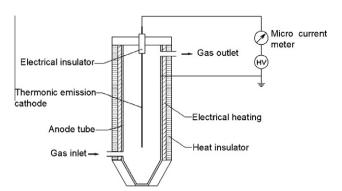


Fig. 1. Schematic diagram of RE–W cathode emission measurement. The thermionic emission cathode is a RE–W round bar.

corded automatically by the computer. Corresponding software was developed to extrapolate zero-field emission density. The effective work function was calculated by Richardson function [12].

Most industrial units work at atmosphere or higher pressure conditions. Therefore, the thermionic emission properties of RE-W cathodes were also investigated at atmosphere pressure in a self-designed computer-controlled automatic laboratory setup, as shown in Fig. 1, which includes thermionic emission cathode, circular stainless steel tube used as anode, gas inlet, gas outlet, emission current measurement system, electrical heating system and controlling parts. An electrical heating system was applied to maintain the ambient temperature at certain level. The round cathode bar (diameter of 2.5 mm) was hung right on the center of the anode tube (diameter of 12 mm) in order to emit electrons uniformly when heated. The cathode was loaded on negatively high voltage and the anode tube was connected to the ground. The emission current at different temperature, voltage and ambient gas environment could be measured by a micro-current meter and displayed on the user interface of the control system. The current density can be calculated from the measured current divided by the cathode surface area.

2.3. Collection performance of a RE–W cathode electrostatic precipitation

Finally, the performances of an electrostatic precipitation (ESP) using one of the RE–W cathodes, Y–W, were investigated. The Y–W was chosen due to its outstanding application characteristics (as will be discussed in Section 3.3). The influences of dust properties and ambient temperature on collection efficiency were investigated.

The experimental ESP setup is shown in Fig. 2. It is a two-stage-tubular ESP [2] in which charging section is separated from collection section. The RE-W cathode is hung in the cylinder center by a stainless steel suspender and connected to the negatively high voltage. The cylinder is connected to ground and used as ash collection anode. The main design parameters are shown in Table 1.

The ash samples were added to the flue gas of $\mathrm{CH_4/air}$ combustion to simulate the flue gas of a coal-fired boiler furnace. A dust feeder with constant dosage characteristics was employed so that the ESP efficiency was investigated at constant average values of fly ash concentration at the inlet of the ESP model. The particle concentration of inlet flue gas and that of outlet were measured by an optical particle counter (PBD5SPM4110, Beijing Big Dipper research institute of chemical industry), respectively. The overall collection efficiency was determined by simultaneous direct measurements of the fly ash mass concentration at the inlet and that in the outlet of the ESP:

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