



## Full Length Article

# Research on characteristics of heavy metals (As, Cd, Zn) in coal from Southwest China and prevention method by using modified calcium-based materials



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## H I G H L I G H T S

- Content, characteristics, transformation of heavy metals in coal and ash are discussed before and after combustion.
- Modified calcium carbonate (CaCO<sub>3</sub>) is used as an additive to treat heavy metals.
- The migration and release of heavy metals in coal is studied and tested in the experiments.

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## A B S T R A C T

Coal plays an important role in the Chinese energy source. The heavy metals released from combustion process stay in the environment and are not easily degraded. This paper examines 8 coal samples from China. It explores the content, characteristics, transformation and control of heavy metals through the microwave digestion, sequential chemical extraction, static burning test and X-ray diffraction (XRD) methods. The zinc (Zn) and cadmium (Cd) in coal mainly exist in exchangeable, sulfide, and residual forms. The arsenic (As) content is mainly present in the sulfide binding state. For these three elements, degrees of organic speciation are not considered to be important and the residue speciation mainly occurs in coal ash. After coal combustion, the heavy metals in coal ash are relatively enriched, and the harm to the environment cannot be ignored. Most of the Cd, As and Zn residues are released into the atmosphere and their volatility rates can be ordered as follows: As > Zn > Cd. There are significant differences in mineral composition between the main phases and mineral carriers of heavy metals in coal and ash. Based on these differences, modified calcium carbonate (CaCO<sub>3</sub>) is used as an additive. In orthogonal experiment, the optimal parameters for treating heavy metals are the temperature (1000 °C), metal salt (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>), and ion ratio ( $\gamma = 15$ ). Under this condition, the volatilization rates of heavy metals (As, Cd and Zn) decreased remarkably.

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## 1. Introduction

Coal plays an important role in the energy structure of China, which will not change quickly. However, coal combustion causes serious pollution problems, and air pollution is very harmful and as such extremely difficult to control.

The contents and distribution characteristics of heavy metals in coal are studied by a series of methods. The migration and

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transformation of heavy metals in the process of combustion are analyzed through relative experiments. Then, a method based on the use of a solid adsorbent is modified and improved to control heavy metal emission.

In the process of coal combustion, the concentration of heavy metals is relatively lower than the gaseous/particulate pollutants released ( $\text{CO}_2$ ,  $\text{SO}_2$ ,  $\text{NO}_x$ , acid gas and soot). However, the ecological damage to the environment caused by these pollutants is very serious and difficult to control [1–5]. The Clean Air Act Amendments (CAAA) noted that coal combustion has become the main source of heavy metals in the air and that the emissions of arsenic (As), cadmium (Cd), manganese (Mn), cobalt (Co), lead (Pb), mercury (Hg), and selenium (Se) should be controlled with the total emissions of each heavy metal to bring down less than 10 tons/year. The US Environmental Protection Agency has reported that atmospheric pollutants released from coal include not only  $\text{SO}_2$ ,  $\text{NO}_x$  and some organic compounds but also dust and heavy metals, which are responsible for causing cancer, especially particulates at the submicron level. By analyzing the atmospheric pollution emissions data from the US, Canada, Western Europe and the former Soviet Union, Nraigu estimated the amounts of heavy metals from coal-fired plants (2–6% As, 10–14% Sb, 6–13% Se, 9–17% Hg, 2–3% Cd) [6].

The 26 trace elements in coal shown in Table 1 are all important for the environment. The environmental impact of the first class is the greatest, and the impact decreases from first to third.

The forms of heavy metals in coal sometimes determine the products of combustion and the damage to the environment. It is important to study the characteristics of those processes. Huffman searched XAFS and found that arsenic mainly occurred in the pyrite and arsenate forms [7]. Riley used the sequential chemical extraction method to study the forms of heavy metals in Australian coal and divided them into exchangeable, carbonate and sulfide bound, pyrite and residual fractions [8].

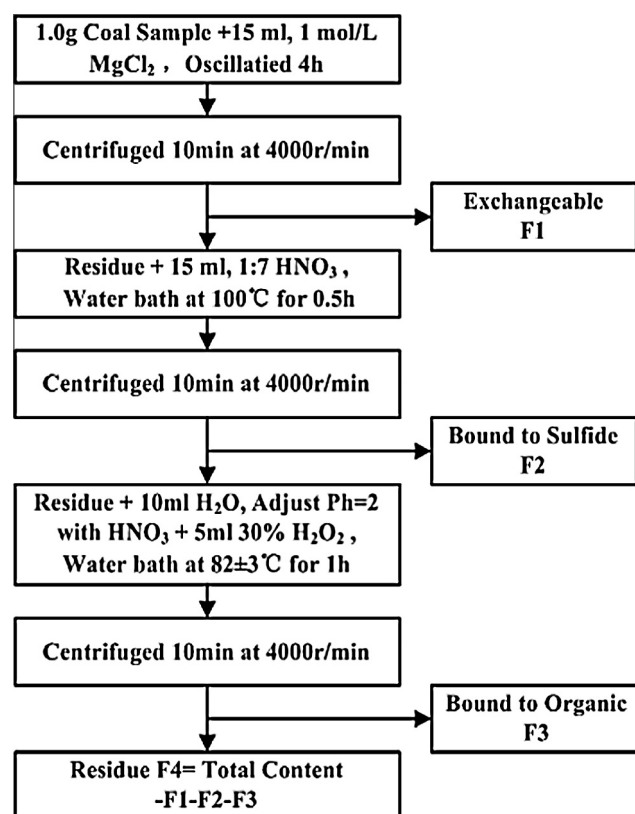
Coal undergoes a series of physical and chemical reactions in the process of combustion. Coal particles are pyrolyzed, and the compounds they contain are volatilized. Under high-temperature conditions, heavy metals can also react with the surrounding gases, and some may become gasified. After combustion, some refractory metal oxides form a series of aerosol-based particles and are then transformed into fly ash particles. Most heavy metals (such as As, Zn, Se) can transition from the gas to the solid phase [9–13]. When the gas temperature drops, the heavy metal elements may become part of the gas phase, the ultra micron and submicron aerosol phase or the ash particle phase [14–16]. The volatility of heavy metals was analyzed, and it was found that Cd, As, Pb, Zn, Cu mainly existed in submicron fly ash particles and were volatile at a temperature of 600–1400 K. In comparison, cobalt (Co), chromium (Cr) and Mn mainly existed in bottom ash and fly ash and were nonvolatile below 1400 K [17].

The methods of controlling heavy metals in coal are divided into three categories: pre-control (coal preparation, coal washing, and coal water slurry technology); mid-control (fluidized bed, the use of additives and bag filter technology); and post-control (dust collectors, flue gas treatment device, and multi-stage purification device) [18–23]. Gullett added limestone, hydrated lime and kaolin at 1000–1300 °C and found a great absorption effect for arsenic, cadmium and lead. Mahuili determined that hydrated lime,  $\text{Al}_2\text{O}_3$  and kaolin had the effect of adsorbing As in smoke. They reported that the adsorption process and mechanism led to the production of calcium arsenate, which was generated by the reaction of calcium oxide and arsenic oxide. The use of calcium carbonate ( $\text{CaCO}_3$ ) as an adsorbent has some effect on various metal elements and this effect can be improved by new research methods [24–30].

**Table 1**

Trace elements in the concerns of environment from coal.

| First class | Second class | Third class |
|-------------|--------------|-------------|
| As          | B            | Ba          |
| Cd          | Cl           | Co          |
| Cr          | F            | I           |
| Hg          | Mn           | Ra          |
| Se          | Mo           | Sb          |
|             | Ni           | Sn          |
|             | Be           | Te          |
|             | Cu           |             |
|             | P            |             |
|             | Th           |             |
|             | U            |             |
|             | V            |             |
|             | Zn           |             |



**Fig. 1.** Flow chart of a sequential extraction method.

**Table 2**

Levels of factors.

| Levels | Factors          |                              |           |
|--------|------------------|------------------------------|-----------|
|        | Temperature (°C) | Metal salt                   | Ion ratio |
| 1      | 900              | $\text{Na}_2\text{CO}_3$     | 15        |
| 2      | 1000             | $\text{K}_2\text{CO}_3$      | 10        |
| 3      | 1100             | $\text{Al}_2(\text{SO}_4)_3$ | 20        |

## 2. Experimental

### 2.1. Total quantity analysis

Before analyzing, the process of combustion is simulated by a static test. The steps of this test are as follows: the coal samples are ground to pass through a 200 mesh sieve. One gram is placed

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