

Integrated Control of Submicron Particles and Toxic Trace Elements by ESPs Combined with Chemical Agglomeration

LI Hailong, ZHANG Junying, ZHAO Yongchun, ZHANG Liqi, ZHENG Chuguang
(State Key Lab. of Coal Combustion, Huazhong University of Science and Technology,
Wuhan 430074, Hubei, PR China E-mail: jyzhang@hust.edu.cn)

Abstract: Enrichment of typical toxic trace elements As on $PM_{2.5}$ was investigated through bench scale experiments. Results showed that the vaporization of arsenic in pyrite was easier than in arsenate form. Along with increasing temperature from 1100°C to 1400°C, arsenic content in $PM_{2.5}$ increased from 0.09 mg/Nm³ to 0.35 mg/Nm³. Then, a novel Electrostatic precipitators (ESPs) combined with chemical agglomeration technique was developed to control the emission of $PM_{2.5}$ and toxic trace elements. $PM_{2.5}$ enriched toxic trace elements are agglomerated by some new chemical agents injected as an aqueous solution upstream of ESPs and come into being conglomerations which can be captured by ESPs easily. Systematic experiments of $PM_{2.5}$ agglomeration showed that chemical agglomeration was a useful method to promote growing up process of submicron particles, XTG was the most effective agglomerant. Resistivity tests revealed that agglomerants could make fly ash resistivity two orders lower at certain concentration, would improve the performance of ESPs with resistivity related performance problems.

Keywords: Submicron particles, Toxic trace elements, Electrostatic precipitators, Agglomeration, Resistivity

1 INTRODUCTION

China is the world's largest coal producer and consumer. The environmental problems related with coal combustion have become serious. In china, particle matter is the chief contamination in the urban atmosphere, and 90% of them come from coal combustion. Although the total efficiency of exist dedusting units are as high as 99%, there are still many submicron particles emit into the atmosphere, for submicron particles can escape the dedusting units more easily. Submicron particles escaped from dedusting units always enriched in some toxic trace elements (Hg, As, Se, Pb, Cd, and Cr) have done great harm to human health[1-5]. Because of their tiny size, light weight and enormous number, conventional ways used to control submicron particles exhibited little effect, set pretreatment stage before conventional dedusting units has become a hot topic. Many physical methods (acoustic agglomeration[6, 7], optic agglomeration[8], electric agglomeration [9, 10], magnetic agglomeration[11], thermal agglomeration [12], and phase transition agglomeration) have been used to promote fine particle agglomeration. These technologies all have some defects[13], it is difficult to apply them in large-scale. In chemical agglomeration process, chemical agglomeration agents are sprayed into the flue before ESPs to accelerate agglomeration between submicron particles. The chemical agglomeration technology whose cost is cheaper can control the emissions of submicron particles and remove toxic trace elements simultaneously without changing the operation parameters of the ESPs. This particle agglomeration technique is a useful and promising method to control the emission of submicron particles from coal combustion.

In this paper, the enrichment of As in $PM_{2.5}$ has been

investigated through bench scale experiments, influence of combustion temperature and occurrences of As in coal were analyzed. Then, a novel technique was developed to control the emission of $PM_{2.5}$ and toxic trace elements. Systematic experiments of $PM_{2.5}$ agglomeration were conducted on a homemade bench, the influencing factors include agent types, temperature were analyzed. Finally, DR electrical resistivity test instrument was used to evaluate regulatory effect of chemical agglomeration agents on fly ash resistivity.

2 ENRICHMENT AND EMISSION OF ARSENIC IN SUBMICRON PARTICLES

Coal combustion experiments were carried on a laboratory-scale drop tube furnace[14]. Coal samples were grinded mechanically to make sure the particle size is less than 74 μ m, and the samples were dried at 105 °C at least 2 hours to remove external water. The reaction temperature varied from 1100 °C to 1400 °C. The raw coal at a feeding rate about 0.2 g/min was entrained by air into the furnace, and a mixture of N₂ and O₂ gases was used as the combustion atmosphere. Oxygen content was 20% being balance. The total gas flow rate was 10L/min. The residence time of the particles in the tube was about 2 second. Under given conditions, all the coals burnt completely. The fly ash was collected by cyclones having a cut-off size around 10.0 μ m, and directed to a low pressure impactor (LPI) for a size-segregated collection. The LPI used here was consisted of thirteen collection stages designed to collect particles of decreasing size as follows: 9.8, 6.6, 3.95, 2.36, 1.58, 0.936, 0.605, 0.377, 0.258, 0.154, 0.0944, 0.0565 and 0.0281 (50% aerodynamic cutoff diameter – in μ m). Each stage was

composed of a filter above a substrate and a substrate holder. By the size-segregated collection using LPI, both the concentration of PM_{10} and its particle size distribution were obtained simultaneously. X-ray fluorescence spectrometry (XRF) was used to determine the major oxides in the PM_{10} .

To quantify the emission of arsenic, the emission concentration is calculated as follows:

$$E_m = \sum M_i \times C_i \quad (1)$$

$$E_c = E_m / V \quad (2)$$

where, E_m is the emitted amount of As; E_c is the As concentration in the emission; M_i is the mass in size fraction i ; C_i is the arsenic concentration in size fraction i ; V is the total volume of flue gas.

3 EFFECT OF COMBUSTION TEMPERATURE ON ARSENIC EMISSION

The emission of arsenic in different temperature is shown in Fig.1. Along with increasing temperature from 1100 °C to 1400 °C, the emission of arsenic in PM_1 increases from 0.07 mg/Nm³ to 0.25 mg/Nm³, the emission of arsenic in $PM_{2.5}$ increases from 0.09 mg/Nm³ to 0.35 mg/Nm³ and the emission of arsenic in PM_{10} increases from 0.18 mg/Nm³ to 1.03 mg/Nm³. This result is caused by two reasons, one is the amount of total PM increased by temperature increase, and the other is that much more arsenic vapor condensed and reacted on the PM surface at high temperature.

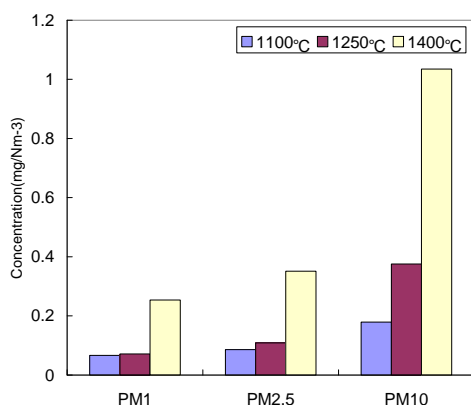


Fig. 1 The emission of arsenic within PM at different temperatures

4 EFFECT OF OCCURRENCE MODE ON ARSENIC EMISSION

As shown in Fig. 2, although the arsenic content in two coals is equal, but the emission during coal combustion varies greatly. Arsenic in LT-K2 lignite mainly occurs as a substitution element for sulfur in pyrite, while present in oxidized form as arsenate minerals in mixed PDS bituminous coal[15]. Some laboratory studies have suggested that the behavior of arsenic during combustion will be determined by the occurrence mode of arsenic in coal[16-18]. The former mode might experience explosive volatilization, in which most of the arsenic will be volatilized, either as arsenic vapor or as fine-particle arsenic oxide, and the latter mode will fuse,

react, and be assimilated as arsenate in the aluminosilicate fly ash particles [19]. This is the dominant reason for the emission difference.

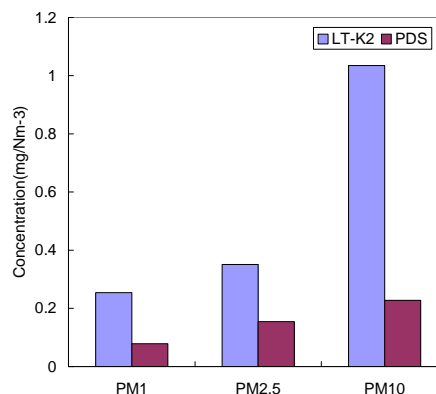


Fig. 2 The emission of arsenic within PM from different coals

5 AGGLOMERATION OF SUBMICRON PARTICLES

Pretreatment stage before ESPs can help ESPs to remove $PM_{2.5}$. Hence, it has become a hot topic in dedust researches. Some physical or chemical means are adopted in pretreatment stage to promote submicron particles growing up. As acoustic, optic, electric, magnetic, thermal, and phase transition means are defective, we developed a new chemical agglomeration method to promote agglomeration between submicron particles[20,21]. The schematic diagram of this new method is shown in Fig. 3. $PM_{2.5}$ are agglomerated by agglomerants sprayed into the flue before ESPs. The resistivity of particles is regulated by agglomerants simultaneously. In this way, the operational conditions of ESPs can be optimized, the efficiency of ESPs removing submicron particles can equal to bag filter, and even exceed bag filter. The ESPs combined with chemical agglomeration way whose cost is cheaper can control the emissions of submicron particles and can remove toxic trace elements simultaneously without changing running operation of boilers and dedusting units. It is a useful and promising method to control the emission of submicron particles from coal combustion.

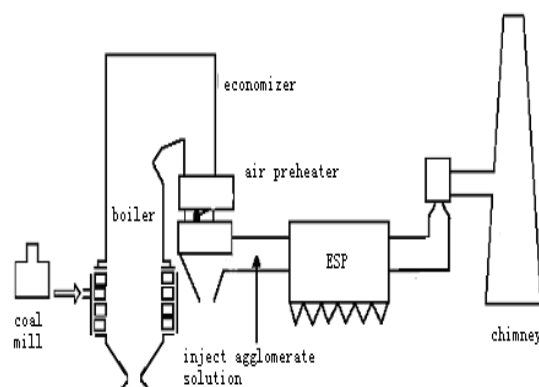


Fig. 3 Schematic diagram of ESPs combined with chemical agglomeration method

Systematic experiments of $PM_{2.5}$ agglomeration are conducted on a homemade bench, the influencing factors

include agent types and temperature are analyzed. The experimental bench and detailed operational conditions are the same as described in literature 20.

6 AGGLOMERATION EFFECT OF DIFFERENT AGGLOMERANTS

Four different agglomerants solutions and water were injected into the simulated flue gas to evaluate the agglomeration efficiency. The different molecular chain structures of agglomerants result the significant difference in agglomeration rates (Fig. 4). The particles concentration at the outlet of dust collecting equipment was about 480.8 mg/m³ without the addition of agglomerants. With the addition of agglomerant solution, the particle emission concentration in flue gas decreased significantly. The agglomeration efficiency of XTG agglomerant was the highest, after the injection of XTG solution, the outlet particle concentration decreased to 262.3 mg/m³. This is because XTG's molecular weight is moderate, molecular chain is linear structure, it has higher flexibility and can agglomerate submicron particles more effectively.

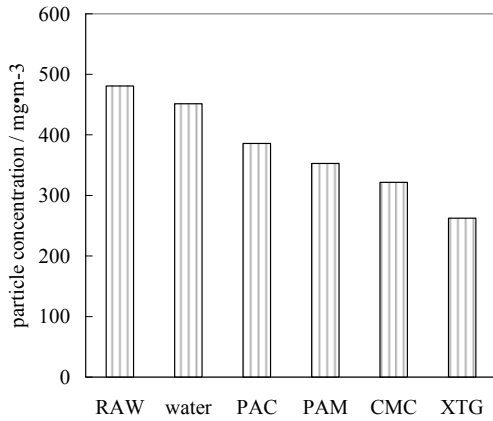


Fig. 4 The agglomeration effect of different agglomerants (RAW - without agglomerants)

7 INFLUENCE OF TEMPERATURE ON AGGLOMERATION

Temperature in agglomeration chamber showed significant influence on submicron particles agglomeration. Changing temperature in agglomeration chamber, maintaining other operation conditions of agglomeration such as agglomerants types, flow rate, concentration stable, the influence of temperature was studied. As shown in Fig.5, along with increasing temperature from 180□ to 230 □, dust concentration after

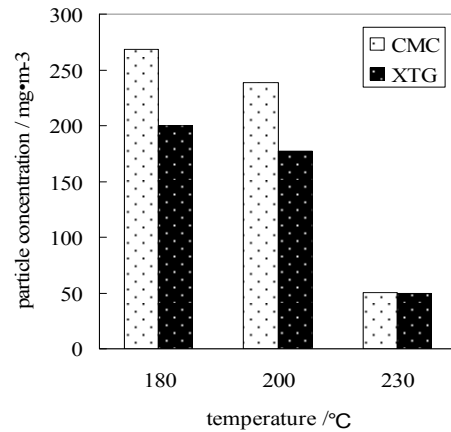


Fig. 5 The influence of temperature on the agglomeration dedusting unit reduces. Especially, when temperature increase from 200□ to 230□, dust concentration decreases obviously. This result may caused by two reasons, one is the wetting ability and adsorption activity increased with temperature increase, and the other is that agglomerants are more flexible at higher temperature.

8 INFLUENCE OF AGGLOMERANTS ON FLY ASH RESISTIVITY

Four different agglomerant additives were blended with fly ash into a mixture to evaluate their regulatory effect on fly ash resistivity. A DR dust electrical resistivity test instrument was used to test the resistivity of the mixture. The results are shown in Fig. 6, the ordinate is logarithm of resistivity, and the abscissa is concentration of additives. As shown in Fig.6, fly ash resistivity decreased swiftly with increasing additive concentration, while changed slightly once additive concentration up to 1%. Additive 3 can decrease fly ash resistivity effectively when additive concentration is less than 1%, it can decrease fly ash resistivity from 10^{11.25} Ω·cm to 10^{9.18} Ω·cm with the concentration increasing from 0 to 1%. But when additive concentration is more than 2% additive 1 shows the best effect, it can decrease fly ash resistivity from 10^{11.25} Ω·cm to 10^{8.77} Ω·cm with the concentration increasing from 0 to 4%.

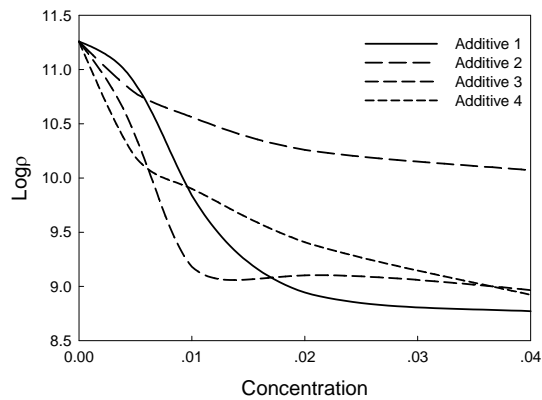


Fig. 6 Resistivity of fly ash mixed with different agglomerants additives

Fig.6

9 CONCLUSIONS

(1) The emission of arsenic in submicron particles is influenced by the occurrence mode of As in coal and the temperature during coal combustion. The vaporization of arsenic in pyrite is easier than in arsenate form. Along with increasing temperature from 1100 °C to 1400 °C, arsenic content in PM_{2.5} increased from 0.09 mg/Nm³ to 0.35mg/Nm³. The emission of arsenic within PM_{2.5} was high without any control devices, which would cause great harm to human health.

(2) Chemical agglomeration is a useful method to promote growing up process of submicron particles. Submicron particles can be agglomerated by XTG agglomerant effectively. Along with increasing temperature from 180 °C to 230 °C, dust concentration after dedusting unit reduces.

(3) Agglomerants can make fly ash resistivity two orders lower at certain concentration. These agglomerants will improve the performance of ESPs with resistivity related performance problems.

(4) The ESPs combined with chemical agglomeration way whose cost is cheaper can control the emissions of submicron particles and can remove various pollutants simultaneously without changing running operation of boilers and dedusting units. It is a useful and promising method to control the emission of submicron particles and toxic trace elements from coal combustion.

ACKNOWLEDGEMENTS

This research was supported by the High-tech Research and Development Program of China (2006AA05Z303), National Basic Research Program of China (2006CB200304), National Natural Science Foundation of China (NSFC) (40672098, 90410017), and Graduates' Innovation Fund of Huazhong University of Science and Technology (HF0504407121).

REFERENCES

- Sui Jianca, Xu Minghou, Qiu Jihua et al. Physio-chemical characteristics and formation mechanism of inhalable particulate matter in coal combustion process. *Journal of Chemical Industry and Engineering*, 2006, 57(7): 1664-1670.
- Neas M L. Fine particulate matter and cardiovascular disease. *Fuel Processing Technology*, 2000, 65-66: 55-67.
- Freitas, M. Carmo; Almeida, S. Marta; Reis, Miguel A.; Oliveira, Orlando R. Monitoring trace elements by nuclear techniques in PM₁₀ and PM_{2.5}. *Nuclear Instruments and Methods in Physics Research A*, 2003, 505(1-2): 430-434.
- Meij, R, Te Winkel H. The emissions and environmental impact of PM₁₀ and trace elements from a modern coal-fired power plant equipped with ESP and wet FGD. *Fuel Processing Technology*, 2004, 85(6-7), 641- 656.
- Linak, W P; Miller, C. Andrew; Shoji, T, Huggins, F.E, Huffman, G.P.. XAFS spectroscopy analysis of selected elements in fine particulate matter derived from coal combustion. *Energy and Fuels*, 2002, 16(2): 325-329.
- Rodriguez-maroto J J, Gomez-moreno F J, Martin- espigares M et al. Acoustic Agglomeration For Electrostatic Retention Of Fly-Ashes At Pilot Scale: Influence of Intensity of Sound Field at Different Conditions. *Journal of Aerosol Science*, 1996, 127: 621-622.
- Changdong SHENG. Modelling of acoustic agglomeration processes using the direct simulation Monte Carlo method. *Journal of Aerosol Science*, 2006, 37(1): 16-36.
- Di Stasio S. Observation of Restructuring of Nano-particle Soot Aggregates In a Diffusion Flame By Static Light Scattering. *Journal of Aerosol Science*, 2001, 132(4): 509-524.
- Watanabe T. Submicron Particle Agglomeration by an Electrostatic Agglomerator. *Journal of Electrostatics*, 1995, 34: 367-383.
- Hautanen J et al. Electrical Agglomeration of Aerosol Particles in an Alternating Electric Field. *Aerosol Sci and Tech*, 1995, 22: 181-189.
- He Yeqing, Zhou Zengshou, Qi Song, et al. Magnetic agglomeration interaction among particles of Nd-Fe-B powders and its finite element calculation. *Functional Materials*, 2002, 33(2):154-157.
- Shisen XU. Experimental Research on the Performance of Cyclone at High Temperature Improved by Agglomerating Micro-Particles. *Power Engineering*, 1999, 19(4): 309-314.
- Wei Feng, Zhang Junying, Wang Chunmei, et al. Review of submicron particles agglomeration in coal combustion process. *Coal Conversion*, 2003, 26(3): 27-31.
- Zhao Yongchun, Zhang Junying, Huang Wenchun et al. Arsenic Emission during Combustion of High-Arsenic Coals from Southwestern Guizhou, China. *Energy conversion & management*, 2008,49: 615-624.
- Wei Fang. Study on sub-micron particle formation and agglomeration mechanism from coal combustion. PhD Dissertation, Huazhong University of Science and Technology, Wuhan, China, 2005.
- Zielinski RA, Foster AL, Meeker GP, Brownfield IK. Mode of occurrence of arsenic in feed coal and its derivative fly ash, Black Warrior Basin, Alabama. *Fuel*, 2007, 86: 560-72.
- Querol X, Juan R, Lopez-Soler A, Fernandez-Turiel JL, Ruiz CR. Mobility of trace elements from coal and combustion wastes. *Fuel*, 1996, 75: 821-38.
- Kolker A, Huggins FE, Palmer CA, Shah N, Crowley SS, Huffman GP, et al. Mode of occurrence of arsenic in four US coals. *Fuel Processing Technology*, 2000, 63: 167-78.
- Zeng T, Sarofim AF, Senior CL. Vaporization of arsenic, selenium and antimony during coal combustion. *Combustion and Flame*, 2001, 126: 1714-24.
- Wei Feng, Zhang Junying, Zheng Chuguang. Agglomeration rate and effect forces between atomized particles of agglomerator and inhaled-particles from coal combustion. *Journal of Environmental Science*, 2005, 17(2): 335-339.

21. Zhao Yongchun, Zhang Junying, Wei Feng, et al. Experimental Study on Agglomeration of Submicron Particles from Coal Combustion. Journal of Chemical Industry and Engineering (china), 2007, 58(11): 2876-2881.