Effect of Phosphorus Transformation on the Reduction of Particulate Matter Formation during Co-combustion of Coal and Sewage Sludge

DATASET · OCTOBER 2014

5 AUTHORS, INCLUDING:

J.-K. Zhuo
Tsinghua University
18 PUBLICATIONS **93** CITATIONS
SEE PROFILE

Lei Duan
Tsinghua University
87 PUBLICATIONS **2,143** CITATIONS
SEE PROFILE

Qiang Song
Tsinghua University
98 PUBLICATIONS **368** CITATIONS
SEE PROFILE

Qiang Yao
Tsinghua University
270 PUBLICATIONS **1,253** CITATIONS
SEE PROFILE

energy&fuels

Article

pubs.acs.org/EF

Effect of Phosphorus Transformation on the Reduction of Particulate Matter Formation during Co-combustion of Coal and Sewage Sludge

Jiankun Zhuo,[*,†] Shuiqing Li,[†] Lei Duan,[‡] and Qiang Yao[†]

[†]Key Laboratory of Thermal Science and Power Engineering, Department of Thermal Engineering, and [‡]Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084, People's Republic of China

ABSTRACT: Co-combustion of municipal sewage sludge with coal receives renewed attention, for its advantage of low pollutant emissions other than the high thermal efficiency and possibly optimal operation. In this paper, the effect of phosphorus on particulate matter (PM_{10}) formation during co-combustion of coal and sewage sludge was intensively investigated in a 25 kW one-dimensional, down-fired pulverized coal combustor. PM_{10} was collected from the furnace centerline in the outlet of the flue gas cooler using a two-stage nitrogen-aspirated, water-cooling isokinetic sampling probe, followed by an electric low-pressure impactor (ELPI). The particle size distributions (PSDs) of the PM_{10} mass concentration during co-combustion exhibit a high sub-micrometer peak and a relatively low super-micrometer peak compared to those in pure coal combustion. Considering the high ash content of sewage sludge, at a 15% thermal blending ratio, the mass concentration of the total fly ash and PM_{10+} (dp > 10 μm) increases from 1088 to 5059 mg N^{-1} m^{-3} and from 547 to 4403 mg N^{-1} m^{-3}, respectively, with respect to pure coal combustion. However, the mass concentrations of fine particulates, including PM_1, $PM_{2.5}$, and PM_{10}, still maintain the similar emission level with coal combustion. The phosphonation of inorganic matters during the cooling of flue gas obviously reduces the mass concentration of fine particles. When the fraction of sewage sludge was 15% (on a thermal basis), the phosphate and P_2O_5 were appreciably rich in the ultrafine mode. The PSDs of phosphorus indicate the intermediate mode, knowing as a fact that the formed phosphates of inorganic metals cause finer particles to form larger agglomerates.

1. INTRODUCTION

Municipal sewage sludge is the largest byproduct of the wastewater disposal system. With the growth of the city population, the treatment of increasing sewage sludge with a good techno-economic feasibility becomes challenging. Recently, co-combustion of municipal sewage sludge with coal received increasing attention as a prospective disposal with high thermal efficiency, low investment and operating costs, and low emissions. However, for the different fuel properties of coal and sewage sludge, co-combustion of coal with sewage sludge leads to a number of potential disadvantages of combustion, fouling and slagging, and fine particle emission. For instance, the presence and behavior of phosphorus in produced fine particles brings the environmental adverse effect and deactivation of selective catalytic reduction (SCR) $deNO_x$ catalysts.

The formation of fine particles is affected by the difference of the contents of mineral elements between coal and sewage sludge. Some key elements, such as K, Na, Ca, Mg, and Fe, reduce the melting point of the major aluminosilicate matrix, which then enhances the collision−coalescence process of fine particles to form the larger particles or agglomerates. Wolski et al. and Seames et al. found that the increasing blending ratio of sewage sludge in the mixture inhibits the formation of sub-micrometer particles (dp < 1 μm), although the total amount of fly ashes is increasing.[1,2] It was reported that the interaction between the inherent inorganic matter of sewage sludge and the external inorganic matter of coal may be a main reason.[3−6] In addition, the sulfation and phosphonation of inorganic matters significantly change the particle size distribution (PSD) of fine particles during the period of flue gas cooling,[7] especially the interaction of phosphorus compounds with coal ash during the combustion of phosphate-doped coal.[8,9] Particularly, considering the abundance of the phosphorus in sewage sludge, the interaction between phosphorus and inorganic elements during co-combustion of coal and sewage sludge is essential to study the formation of fine particles.

In this paper, the effect of phosphorus transformation on the reduction of particulate matter (PM_{10}) formation during co-combustion of coal and sewage sludge was intensively investigated using a 25 kW down-fired pulverized coal combustor.

2. EXPERIMENTAL SECTION

Experiments was conducted in a 3.2 m high, 0.15 m inner diameter vertical down-fired combustor (Figure 1) that had been described in detail in other literature.[7] Pulverized coal and sewage sludge were transported by primary air by a screw feeder and a micropowder table feeder, respectively. Liquefied petroleum gas (LPG) was used to heat the furnace to ensure the requested temperature profile. A distributed control system (DCS) was used for the automatic operation of the unit, in which the temperature/time histories were recorded.

The blending ratio of sewage sludge is 0, 5, 10, and 15% on a thermal basis (signed as SBR0, SBR5, SBR10, and SBR15), and the input energy is kept at 23.4 ± 0.4 kW (Figure 2), where the input energy from hot air occupies 10.3% and the rest is from coal and sewage sludge. The secondary air was heated up to 400 °C, while the oxygen contents in the flue gas were kept varying between 6 and 7%. The fuels used here are Shenhua lignite coal and the municipal sewage

Special Issue: 7th International Symposium on Coal Combustion

Received: November 16, 2011
Revised: February 14, 2012
Published: February 15, 2012

   dx.doi.org/10.1021/ef201796j | *Energy Fuels* 2012, 26, 3162−3166

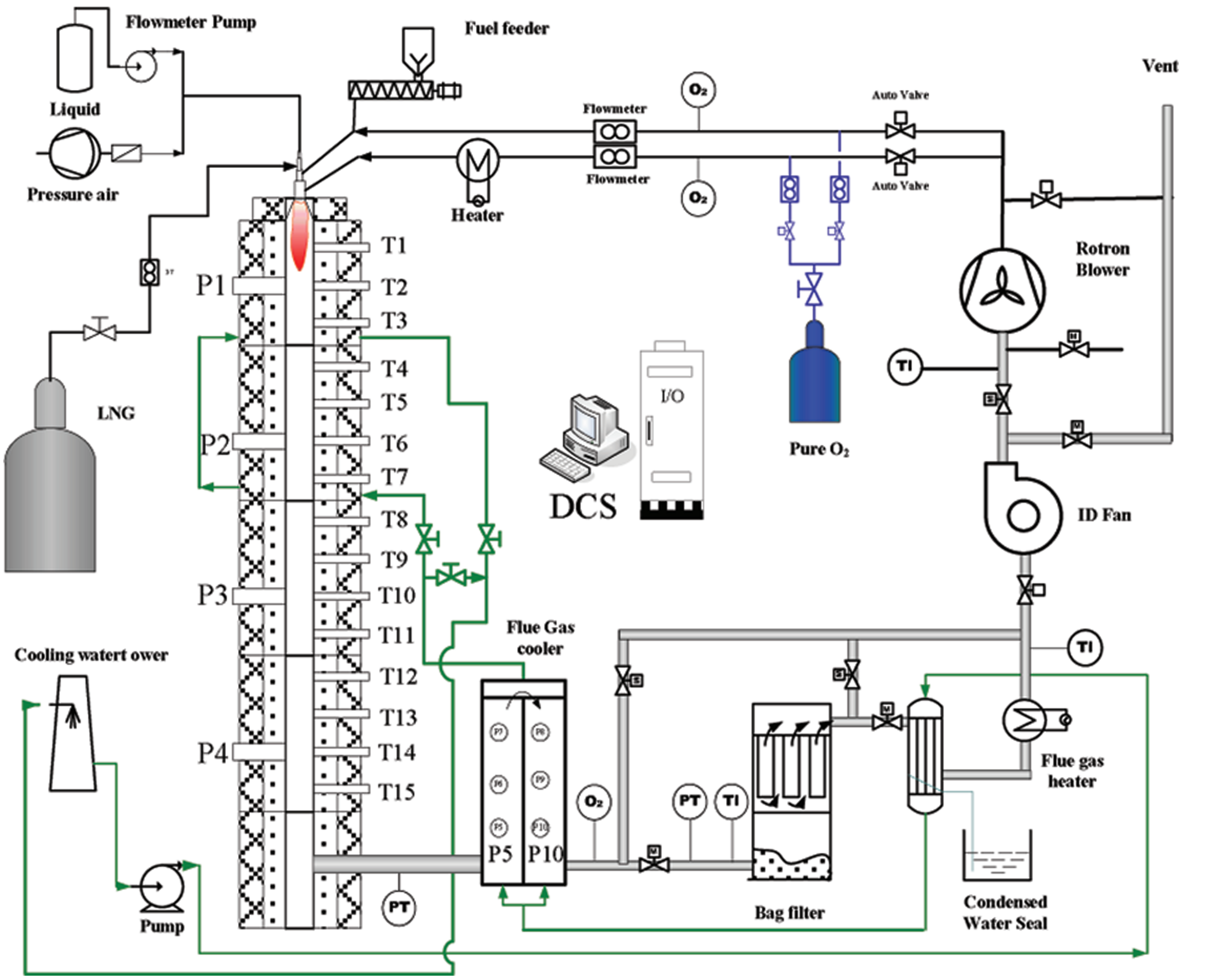

Figure 1. One-dimensional down-fired pulverized coal combustor.

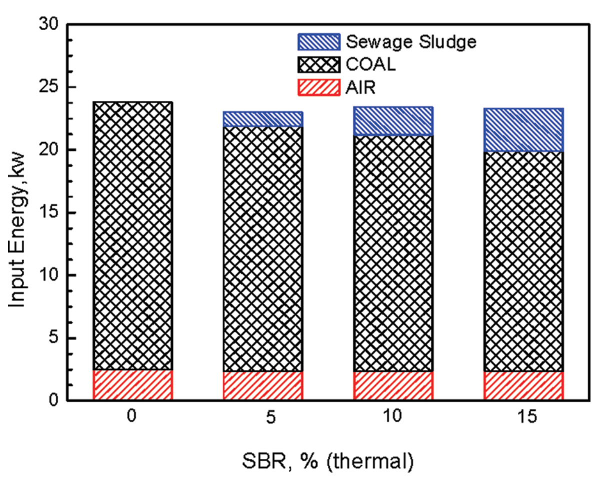

Figure 2. Distribution of input energy of the down-fired combustion in the four blending ratios of sewage sludge: 0, 5, 10, and 15% on a thermal basis.

sludge, whose properties are shown in Table 1. There are distinct differences in fixed carbon, volatile matter, ash, and even P_2O_5 contents of coal and sewage sludge. The sizes of coal and sewage sludge are 14.6 and 177 μm, respectively. Because of the high ratio of volatile matter/fixed carbon with increasing the sewage sludge content in the feedstock, the carbon burnout ratio increased from 95.42 to 99.32% at the P4 sampling port of 2660 mm from the burner exit after mixing with 5% of sewage sludge in the coal (Table 2), while in the outlet of the flue gas cooler (P10 sampling port), the carbon burnout ratios are all larger than 99%.

The temperature profile is kept steady naturally in a self-sustaining combustion state (Figure 3) in all cases at four blending ratios. For instance, the temperature at the distance of 350 mm away from the burner exit is kept at 1145 ± 5 °C, while that at the distant of 5880 mm from the burner exit is only about at 312^{+15}_{-8} °C because flue gas was cooling (also at the sampling port P10).

The particle-laden flue gas was sampled from the furnace central of the flue gas cooler outlet located at the sampling port P10. The sampling probe is nitrogen-aspirated, isokinetic sampling probe cooled by water. To use the electric low-pressure impactor (ELPI), two-stage N_2 dilution is adopted and the total dilution ratio is held at about 70, which can be monitored by the flue gas composition analyzer of MGA 5. The sample out of the probe first passes through the PM_{10} cyclone and then is further separated into 13 stages by the ELPI. The sizes of 13 stages are correspondingly 0.0284, 0.0563, 0.0945, 0.157, 0.263, 0.382, 0.613, 0.948, 1.60, 2.39, 4.00, 6.68, and 9.92 μm. The fine particles of each stage were collected by the polycarbonate in the ELPI for PSD. The samples were collected by a Teflon filter from repeated experiments separated in 3 days for every case, and the morphologies and compositions of samples were analyzed by scanning electron microscopy−energy-dispersive X-ray (SEM/EDX, JSM-6301F SEM), employed at an accelerating voltage of 15 kV and a working distance of 15 mm.

dx.doi.org/10.1021/ef201796j | Energy Fuels 2012, 26, 3162−3166

Table 1. Properties of Coal and Sewage Sludge

proximate analysis	lignite coal (wt %, ad)	sewage sludge (wt %, ad)	LTA ash	lignite coal (wt %, in ash)	sewage sludge (wt %, in ash)
fixed carbon	51.59	1.68	SiO_2	50.86	43.05
volatile matter	32.94	60	Al_2O_3	18.25	12.75
ash	10.83	34.43	Fe_2O_3	10.14	6.08
moisture	4.65	3.89	CaO	9.1	9.22
lower heating value (LHV) (MJ/kg)	26.911	14.118	MgO	2.49	5.27
ultimate analysis	lignite coal (wt %, daf)	sewage sludge (wt %, daf)	TiO_2	0.8	0.68
C	65.05	33.60	SO_3	0.95	0.55
H	4.06	4.81	P_2O_5	0.24	12.42
O	14.01	55.23	K_2O	1.58	3.72
N	1.15	5.84	Na_2O	1.56	1.34
S	0.27	0.88	MnO_2	0.23	0.06

Table 2. Carbon Burnout Ratio of the Four Sewage Sludge Blending Ratios

sewage sludge blending ratio with coal (%)	0	5	10	15
volatile matter/fixed carbon	0.64	0.76	0.89	1.06
carbon burnout ratio at P4 (distance from the burner exit is 2660 mm) (%)	95.42	99.05	99.26	99.28
carbon burnout ratio at P4 (distance from the burner exit is 5880 mm) (%)	99.04	99.32	99.59	99.45

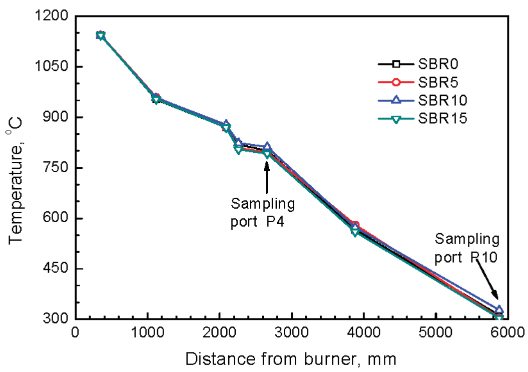

Figure 3. Temperature histories of the down-fired furnace for the four thermal blending ratios of sewage sludge with coal.

3. RESULTS AND DISCUSSION

3.1. PSD of PM_{10} during Co-combustion of Coal and Sewage Sludge. As for co-combustion of coal with sewage sludge with all four blending ratios, the mass PSDs of produced PM_{10} exhibit a similar trend. As seen in Figure 4, separated by a fraction of 0.157−0.263 μm, there are two distinct particle formation modes identified by two prominent peaks. The ultrafine mode, with the particle size less than 0.157 μm, has a peak around sizes of 56.3−94.5 nm. The mass concentration of this mode from pure coal combustion is about 175 mg N^{-1} m^{-3}, which is greatly less than those of the other three blending cases. Then, above the separation fraction of 0.157−0.263 μm, the PSDs of the mass concentration increase with the particle size and approach the second peak in the fraction of 2.39−4.0 μm in the super-micrometer coarse mode, where the mass concentration of this mode from pure coal combustion is about 550 mg N^{-1} m^{-3}, which is slightly larger than the other three blending cases. It should be noticed that PSDs of intermediate fractions ranging from 0.157 to 1.0 μm in blending cases are obviously higher than that in the pure coal combustion case. Is there an intermediate mode besides these two modes because

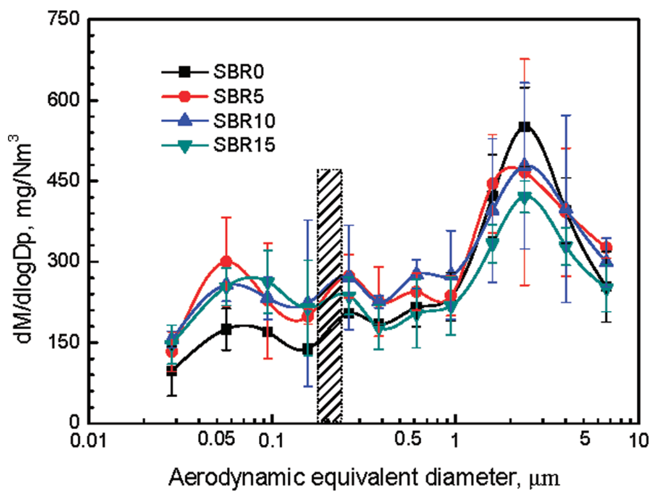

Figure 4. PSD of PM_{10} from four blending ratios of sewage sludge with coal.

of the changes of PSDs in this region? We also note, in comparison to pure coal combustion, the sub-micrometer peak of co-combustion is obviously high, while the super-micrometer peak is unconventionally lower, particularly considering high contents of total ash as well as volatile metal in the sewage sludge. All of these drive us to conduct the PSD of the detailed components, e.g., the phosphorus concentration in fine particles in the later section 3.3.

3.2. Reduction of PM_{10} during Co-combustion. The mass concentration distributions also change in the different sizes of ash particles, such as PM_1 (dp < 1 μm), $PM_{2.5}$ (dp < 2.5 μm), PM_{10} (dp < 10 μm), and PM_{10+} (dp > 10 μm) by different blending ratios of sewage sludge with coal. Because of the higher ash content in the sewage sludge than coal, the mass concentration of total fly ash increases from 1090 mg N^{-1} m^{-3} of the pure coal combustion case to 4634, 4930, and 5060 mg N^{-1} m^{-3} of the co-combustion cases, as the thermal blending ratio of sewage sludge increases, as shown in Figure 5. For the larger ashes, the mass concentration of coarse ash (PM_{10+}) increases with the increasing blending ratio of sewage sludge, from 547.29 mg N^{-1} m^{-3} of the pure coal case to 3885 mg N^{-1} m^{-3} of the 5% blending case. Then, for the fine particles, i.e., PM_{10}, $PM_{2.5}$, and PM_1, as the blending ratio of sewage sludge increases from 0 to 5% (on a thermal basis), the mass concentrations of PM_{10}, $PM_{2.5}$, and PM_1 only slightly increase, which can be attributed to the formation of P_2O_5 in the finer particles. As the blending ratio of sewage sludge further increases to 10%, the concentrations of PM_{10}, $PM_{2.5}$, and PM_1

dx.doi.org/10.1021/ef201796j | Energy Fuels 2012, 26, 3162−3166

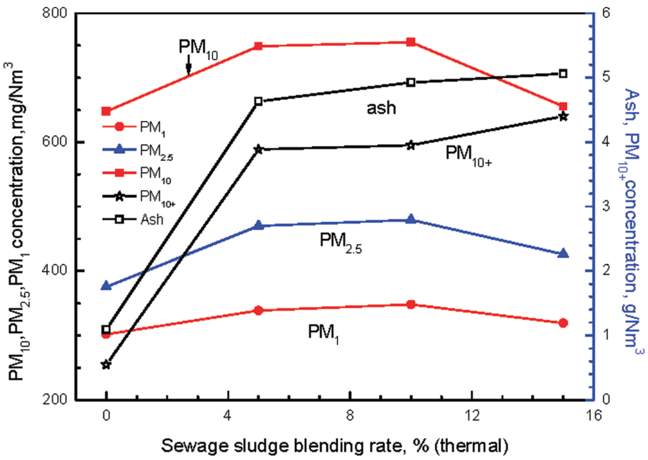

Figure 5. Mass concentrations of the four particle sizes of ash from four blending ratios of sewage sludge with coal.

achieve the highest values within our experimental ranges, which are higher than those of the 5% blending case by 3.6, 7.9, and 4.8%, respectively. However, at a blending ratio of 15%, the mass concentrations further decrease to the emission levels that are comparable to pure coal combustion.

3.3. Effect of Phosphorus Transformation on PM_{10} Formation. The results of the mass concentration of PM_{10} indicate the two distinct formation pathways during co-combustion of coal and sewage sludge that are similar to those that occurred in the coal combustion, in which the ultrafine mode is through vapor condensation of inherent matter and organically bound metals in the ash. Interestingly, these two distinct formation modes, ultrafine or coarse, can be identified by the PSDs of the phosphorus and sulfur in PM_{10} for all four blending ratios, as shown in Figure 6, where the profiles are separated by the transition fraction of 0.157−0.263 μm, which is similar to PSDs of PM_{10}. For three blending cases, such as SBR5, SBR10, and SBR15, the phosphorus concentrations are dramatically higher than those of pure coal combustion, particularly in a finer fraction, because of the high content of P_2O_5 in the sewage sludge ash, while the sulfur concentrations almost keep constant in the ash for all blending and unblending cases. For the ultrafine fraction below 0.157 μm, the phosphorus mainly exists as P_2O_5 and some phosphates together with Ca, Fe, Mg, K, and Na. When the size fraction increases, the phosphates of Ca, Fe, Mg, K, and Na are formed on the surface of the aluminosilicate particles, which makes the particles very sticky and then causes them to form large-size particle agglomerates.

The effect of the phosphorus on PM_{10} formation can be analyzed from the element progressive changes with the blending ratio of sewage sludge, shown in Figure 7. In this work, three typical particle sizes, 0.0563, 0.263, and 4 μm, that are located in different formation modes are specially selected. The elemental compositions of three sizes are illustrated in panels a−d of Figure 7. The prevalent elements of the large size fraction are silica and alumina, while P, S, Fe, Ca, K, and Na together with Al and Si constitute the ultrafine size fraction. With the increasing blending ratio of sewage sludge, although the content of phosphorus in the 4 μm fraction also increases, the dramatic increment or enrichment of phosphorus occurs in much finer fractions at 0.0563 or 0.263 μm. It further affects the content of Fe, Ca, K, and Na in the related fractions. For example, the contents of calcium in the finer (0.0563 μm), intermediate (0.263 μm), and coarse (4.0 μm) fractions are

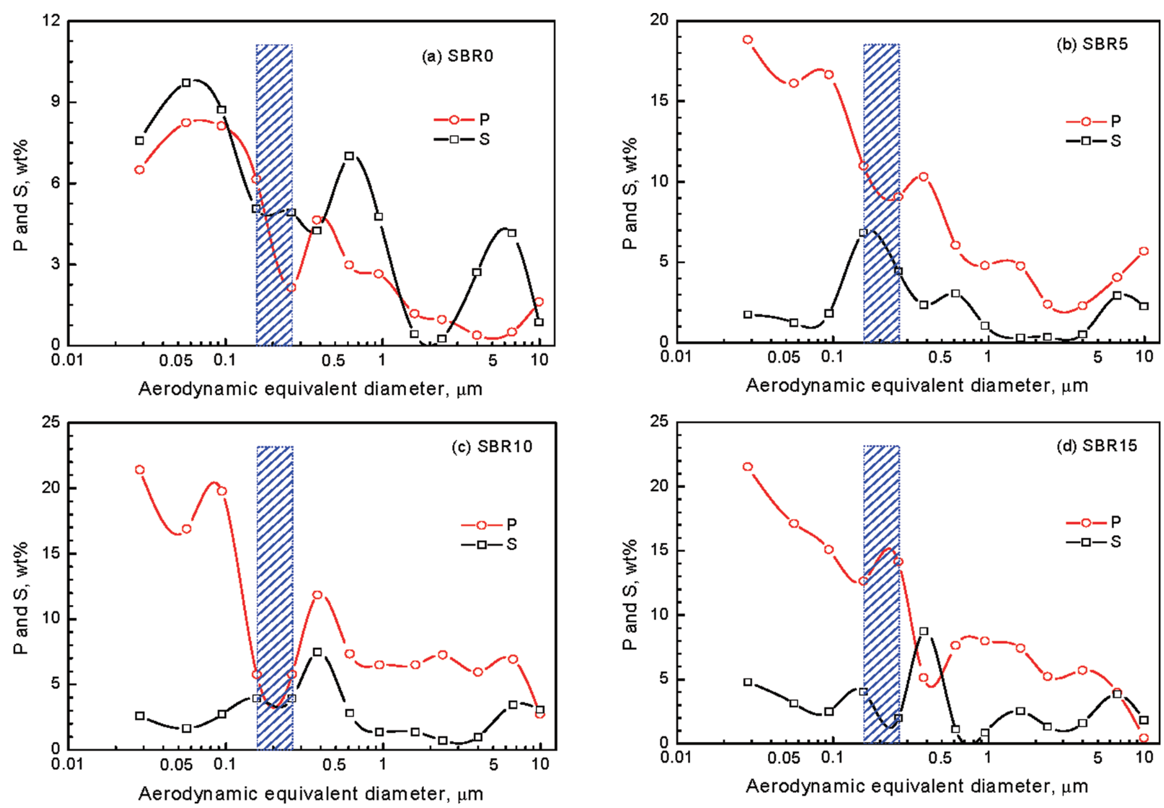

Figure 6. PSD of sulfur and phosphorus in PM_{10}: (a) SBR0, (b) SBR5, (c) SBR10, and (d) SBR15.

dx.doi.org/10.1021/ef201796j | Energy Fuels 2012, 26, 3162−3166

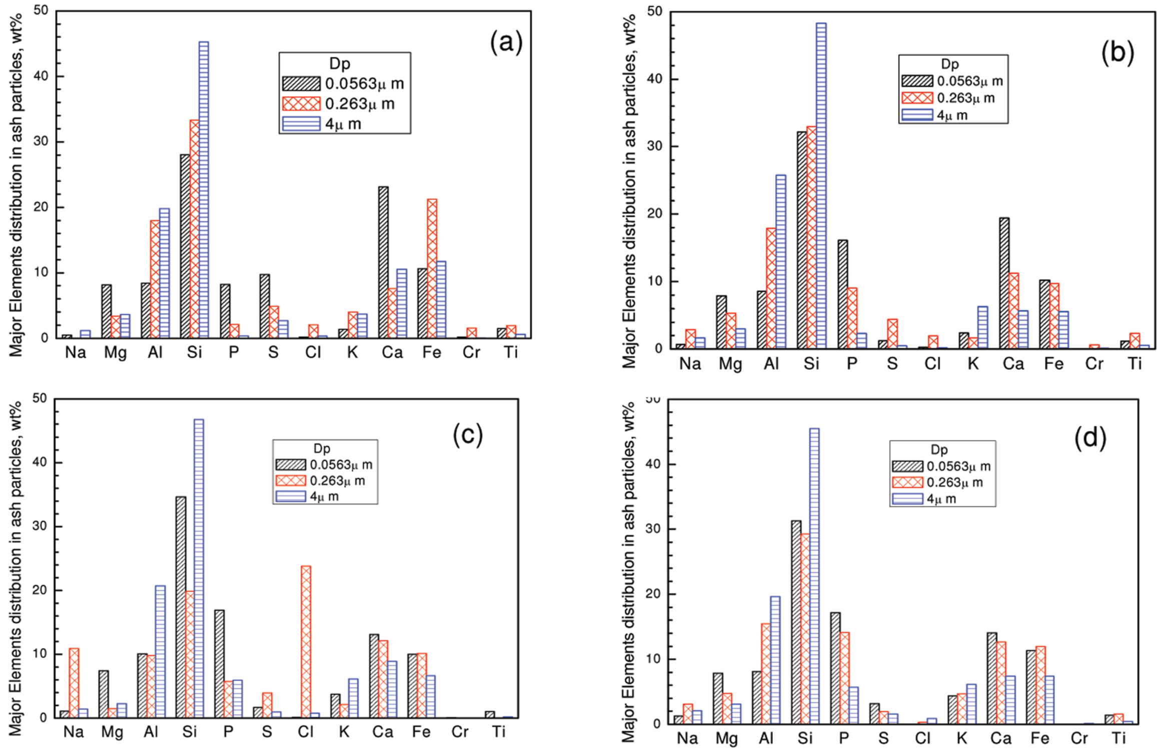

Figure 7. Mass content of major inorganic elements in different particle sizes during co-combustion of sewage sludge with coal: (a) SBR0, (b) SBR5, (c) SBR10, and (d) SBR15.

23.1, 7.6, and 10.5%, respectively. However, as the sewage sludge blending ratio increases to 15%, the contents of calcium in these three fractions are 14.1, 12.7, and 7.4%, respectively. Similar effects of phosphorus can be found for Fe and Na in the intermediate fraction. It implies the agglomeration of ultrafine particles to form a relatively larger fraction, which is reported as the intermediate model between the ultrafine and coarse modes.[7] The existing phosphate of inorganic metals significantly affects the PSD of the mass concentration of fine particles.

4. CONCLUSION

In this paper, the effect of phosphorus on the PM_{10} formation during the co-combustion of coal and sewage sludge is intensively investigated in a 25 kW, self-sustained pulverized coal combustor under strict nearly identical temperature profiles in all cases. The conclusions are drawn as follows: (1) As the blending ratio of sewage sludge increases, the submicrometer peak of PSDs becomes higher, while the supermicrometer peak becomes unconventionally lower, irrespective of the high ash content in sewage sludge. It is noted that the PSDs in intermediate fractions (0.157−1.0 μm) increase. (2) At a thermal blending ratio of 15%, the mass concentrations of total fly ash and PM_{10+} are 5059 and 4403 mg N^{-1} m^{-3}, which are greatly higher than those in pure coal combustion (1088 and 547 mg N^{-1} m^{-3}). However, the mass concentration of fine particulates, such as PM_1, $PM_{2.5}$, and PM_{10}, was maintained at the emission level of coal combustion. (3) The phosphorus concentrations in a finer fraction of blending combustion are higher than those of pure coal combustion. The PSDs of phosphorus indicate that the formed phosphate of inorganic metals causes finer particles to form larger agglomerates, which is known as the intermediate mode.

AUTHOR INFORMATION

Corresponding Author

*Telephone: +86-10-62773384. Fax: +86-10-62794844. E-mail: zhuojk@mail.tsinghua.edu.cn.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the Key Project of the Chinese Ministry of Education (309005), the National Program for New Century Excellent Talents in University, and the Tsinghua University Award for Young Talents on Fundamental Studies. Special thanks are due to Prof. Richard Axelbaum at Washington University in St. Louis (WUSTL), St. Louis, MO, and Dr. Yi Guo at the National Institute of Clean-and-Low-Carbon Energy (NICE), Beijing, China.

REFERENCES

(1) Wolski, N.; Maier, J.; Hein, K. R. G. *Fuel Process. Technol.* **2004**, *85*, 673−686.

(2) Seames, W. S.; Fernandez, A.; Wendt, J. O. L. *Environ. Sci. Technol.* **2002**, *36*, 2772−2776.

(3) Ninomiya, Y.; Zhang, L.; Sakano, T.; Kanaoka, C.; Masui, M. *Fuel* **2004**, *83*, 751−764.

(4) Wang, Q.; Zhang, L.; Sato, A.; Ninomiya, Y.; Yamashita, T.; Dong, Z. *Proc. Combust. Inst.* **2009**, *32*, 2701−2708.

(5) Zhang, L.; Ito, M.; Sato, A.; Ninomiya, Y.; Sakano, T.; Kanaoka, C.; Megumi, M. *Fuel Process. Technol.* **2004**, *85*, 983−1011.

(6) Zhang, L.; Ninomiya, Y. *Proc. Combust. Inst.* **2007**, *31*, 2847−2857.

(7) Zhuo, J.-k.; Li, S.-Q.; Yao, Q.; Song, Q. *Proc. Combust. Inst.* **2009**, *32*, 2059−2066.

(8) Beck, J.; Unterberger, S. *Fuel* **2006**, *85*, 1541−1549.

(9) Beck, J.; Unterberger, S. *Fuel* **2007**, *86*, 632−640.