



PSO Project 10085

Final Report – Co-Firing of Coal and RDF in Suspension

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A global equilibrium analysis of co-firing coal and solid recovered fuel

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Abstract

Global equilibrium calculations have been performed to study the behavior of ash forming species in co-combustion of a bituminous coal and a solid recovered fuel (SRF). It revealed that co-combustion of coal and 25% SRF (weight basis) could significantly reduce the formation of NaCl (g) and KCl (g) in moderate temperature (800–1000 °C), as compared with that in 100% SRF combustion. The simulation results were compared with the experiment results obtained at the same fuel condition, and qualitative agreements were achieved. Both results suggest that co-combustion of coal and SRF would be a feasible method to increase the electrical efficiency of utilizing waste, as the aluminosilicates content in the coal could mitigate the ash related problems caused by the relative high chlorine and alkali content in the SRF. The influence of different SRF quality on the behavior of co-combustion was evaluated through adding PVC and NaCl to the fuel mixture. Both the simulation and experimental results indicate that formation of NaCl (g) and KCl (g) at temperatures above 1000 °C is significantly increased with the addition of PVC and NaCl. And a large fraction of the formed NaCl (g) and KCl (g) is not converted to aluminosilicates during the flue gas cooling in the experiments, probably due to kinetic limitations. The results suggest that it is important to control the chlorine and alkali content in SRF, in order to facilitate co-combustion of coal and SRF.

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1. Introduction

Incineration is a widely applied method to produce heat and power from waste and at the same time reduce the volume of waste for landfill [1,2]. However, the electrical efficiency of a dedicated waste incineration plant is usually limited to 20–25%, mainly due to the relatively high alkali, chlorine and trace element contents in the waste. These elements may induce severe ash deposition and corrosion problems in the boiler, thus the steam temperature of a dedicated waste incineration plant usually cannot exceed 450 °C [1,3]. To improve the electrical efficiency of utilizing waste, one promising approach is to co-combust a certain fraction of waste together with coal in a pulverized coal-fired power plant [4]. The main advantage of this technique is that the synergy effects among the ash forming species in the two fuels may allow the coal-fired power plant operating at a steam temperature similar to that in dedicated coal combustion, thus the electrical efficiency of utilizing the waste can be greatly increased. The presence of such synergy effects has been demonstrated in co-combustion of coal and straw, showing that the sulphur and aluminosilicates in coal could effectively mitigate the ash related problems caused by the alkali and chlorine in straw [5-9]. Besides the advances in waste energy efficiency, another potential benefit of co-firing coal and waste is that the CO₂ emission from the coal-fired power plant can be reduced, as the majority of the combustible waste would be biogenic materials.

Solid recovered fuel (SRF) is an upgraded waste derived from high calorific fractions of nonhazardous waste streams such as municipal solid waste (MSW), commercial or bulky waste [10]. The main purpose of producing SRF from these waste streams is to facilitate the utilization of waste in coal-fired power plants or industrial furnaces such as cement kilns. The standardization of SRF in terms of the calorific value, chlorine and mercury contents has been set by the European Committee for Standardization (CEN/TS 15359, 2006). According to the standardization, the upper limit for the chlorine content in the SRF is in a range of 0.2–3 wt% (dry basis) [11]. This indicates that the chlorine content in SRF may be greatly higher than that of coal/biomass, and it could become a limiting factor for the utilization of SRF in coal-fired power plants. Therefore, a systematical assessment of the ash behaviors in co-combustion of coal and SRF at different SRF share and property conditions is desired.

The transformations of ash forming species in solid fuel combustion are complicated, which involve processes such as release of inorganic elements from burning particles, reactions among the inorganic and organic elements, and physical transitions such as coalescence, nucleation and condensation. Modeling work have been conducted to characterize part of the chemical and/or physical processes of ash transformations, and reasonably good agreements were achieved with some experimental results based on simplified combustion system [12-15]. However, the application of these models to a real combustion system is confined by the number of inorganic elements considered in these models which is usually much less than that present in the actual fuel. In this context, global equilibrium model, which takes into account the interactions among multiple inorganic and organic elements, is recognized as a promising modeling tool to study the ash transformation in a complicated combustion system [16-21]. Although global equilibrium modeling has its own drawbacks such as neglecting the kinetic and mixing limitations, it is still an advantageous approach to model the ash behaviors, particularly in capturing the general tendency of species speciation and indentifying the possible ash transformation pathways.

In the present work, global equilibrium calculation is used to investigate the ash behaviors in co-combustion of coal and SRF. The simulation is carried out at different SRF share and property conditions. The main emphasis of the work is on the behaviors of K, Na, S and Cl in co-combustion, as these elements are of key importance for ash deposition and corrosion in the boilers. The simulation results from the present work are compared with experiments carried out at the same fuel conditions in an entrained flow reactor. The primary objective of the present work is to provide a theoretical analysis

of the interactions among the ash forming species in co-combustion of coal and SRF, as well as the possible influences of different SRF share and properties on co-combustion. In addition, the comparison between the simulation and experimental results may shed light on how far a real combustion system is from the prediction of global equilibrium calculation. The discrepancies between the results from global equilibrium calculation and experiments are discussed.

2. Thermodynamic modeling

2.1 Software and database

Global equilibrium calculation was performed by using the software FactSage 6.0. The calculation was carried out with the equilibrium module of FactSage 6.0, which minimizes the Gibbs free energy of a given system and predicts the equilibrium distribution of the species. The database used in this study was selected from the FACT53, FToxid, and FTsalt databases. The principle of the database selection was that the species with thermodynamic data significantly deviated from the simulation temperature was excluded, and for the species which appeared in multiple databases, the thermodynamic data in the FToxide/FTsalt database was chosen. The elements involved in the calculation were C, H, O, N, S, Cl, Si, Al, Ti, P, Fe, Ca, Mg, Na and K. This means that the interactions among these elements are taken into account in the simulation. A total number of 741 species (259 gaseous, 91 liquids, and 391 solids) was considered in the calculation. Among these species, the species related with K, Na, S and Cl are the main emphasis of the present work.

2.2 Modeling approach

To assess the behavior of major inorganic elements in co-combustion of coal and SRF, two serials of equilibrium calculation were carried out. In the first serial, calculations were performed on co-firing a bituminous coal and a SRF which have been applied in the experiments in an entrained flow reactor (EFR). The simulation was carried out at different SRF mass share condition (0%, 7.9%, 14.8 %, 25%, and 100%), with the properties of the fuel mixture given in Table 1. The fuel condition used in the simulation is the same as that in EFR experiments, except for the case with pure SRF.

Although the production of SRF would follow the standardization of CEN/TS 15359, the properties of SRF may still deviate significantly according to the source of SRF and the processing techniques. Therefore the second serial of equilibrium calculation was carried out to evaluate the influence of different SRF properties on co-combustion of coal and SRF. This was achieved by mixing the coal and SRF with PVC/NaCl. In the mixture, the mass share of coal was fixed at 85.2%, and the total mass share of the secondary fuel (SRF+PVC/NaCl) was maintained as 14.8 %. As an example, the addition of 1 % NaCl means that the mass share in the fuel mixture is 85.2% coal, 13.8% SRF and 1% NaCl. In this way, the change of additive type/share can be regarded as a significant variation of the SRF property. The addition of PVC simulates a SRF with significantly high chlorine content, whereas the addition of NaCl simulates a SRF with high alkali and chlorine content. It should be noted that in practical the SRF may not contain such high PVC/NaCl contents. However, performing simulations at these extreme conditions would reveal the possible effects of the alkali and chlorine on the ash transformations in co-combustion. The properties of the fuel mixture used in the second serial of equilibrium calculation are given in Table 1. Experiments at the same fuel conditions have also been performed in the EFR.

Table 1 Fuel properties used in the equilibrium calculation.

Properties	Coal+SRF					Coal+SRF+Additive			
	Coal	7.9% SRF	14.8% SRF	25% SRF	SRF	2% PVC	4% PVC	1% NaCl	2% NaCl
Moisture (wt% wet)	5.25	5.25	5.24	5.24	5.20	5.16	5.08	5.19	5.14
C (wt% dry)	71.00	69.97	69.08	67.75	58.00	68.68	68.29	68.50	67.92
H (wt% dry)	4.90	5.03	5.15	5.33	6.60	5.12	5.08	5.09	5.02
O (wt% dry)	16.31	17.47	18.49	19.99	31.03	17.87	17.25	18.18	17.87
N (wt% dry)	1.50	1.46	1.43	1.38	1.00	1.41	1.39	1.42	1.41
S (wt% dry)	0.70	0.68	0.66	0.63	0.42	0.65	0.64	0.65	0.65
Cl (wt% dry)	0.03	0.05	0.07	0.09	0.28	1.20	2.33	0.67	1.28
Si (wt% dry)	3.06	2.90	2.77	2.57	1.08	2.75	2.72	2.76	2.75
Al (wt% dry)	1.21	1.13	1.06	0.96	0.21	1.06	1.05	1.06	1.06
Mg(wt% dry)	0.15	0.14	0.14	0.13	0.09	0.14	0.13	0.14	0.14
P (wt% dry)	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
Ca (wt% dry)	0.18	0.22	0.26	0.31	0.67	0.24	0.23	0.25	0.24
Na (wt% dry)	0.07	0.08	0.08	0.08	0.11	0.08	0.07	0.47	0.86
K (wt% dry)	0.21	0.20	0.19	0.18	0.09	0.19	0.19	0.19	0.19
Ti (wt% dry)	0.06	0.06	0.07	0.08	0.12	0.07	0.06	0.07	0.07
Fe (wt% dry)	0.60	0.57	0.55	0.52	0.28	0.55	0.54	0.55	0.55

*The mass share of coal is fixed at 85.2%.

To perform the equilibrium calculation, it was assumed that 1 kg of fuel (wet basis) was combusted at an excess air ratio of 1.43. This excess air ratio was chosen based on the average value applied in the co-combustion experiments in EFR. The temperature range selected in the calculation is 600–1400 °C. The lower limit was chosen as the 600 °C, in order to provide a comparison with the fly ashes from the EFR experiments. The fly ashes in the EFR were collected at a location with flue gas temperature about 600 °C. During the collection process, the fly ash and flue gas were cooled instantaneously by a water-cooled probe. Therefore, it is considered that equilibrium is difficult to reach at temperature below 600 °C, because of the insufficient reaction time and relatively slow reaction rate at this temperature. The upper limit was chosen as 1400 °C based on the achievable fuel combustion temperature in the entrained flow reactor. The temperature step used in the simulation is 10 °C, and the pressure is 1 atm.

2.3 Comparison with experiments

In our previous work, co-combustion experiments on coal and SRF have been carried out in an entrained flow reactor (EFR), with the same fuel conditions as given in Table 1 (except for the case with pure SRF). This provides an opportunity to directly compare the experimental results with the equilibrium calculations. It is well-known that global equilibrium calculation ignores a number of factors which would present and influence a practical combustion system, such as residence time, reaction kinetics, concentration/temperature gradients (mixing), and the association of elements in the fuels. Therefore, by comparing the simulation results with the experimental results, it is possible to identify whether the tendency observed in the EFR experiments is in agreement with the equilibrium calculation. This might provide theoretical explanations to the observed experimental results. Moreover, the comparison could reveal the discrepancies between the experimental and simulation results, which would reflect the limitations of equilibrium calculations.

The experimental data selected for comparison is concerned with the partitioning of K, Na and S in combustion. In the EFR experiments, the total K content and the water soluble K content in the fly ash were analyzed, thus the percentage of K present as water soluble form in fly ash could be calculated.

The parameter “percentage of K present as water soluble form” gives a rough measure on how much potassium in the fuel is converted to potassium chloride/sulphate during combustion. However, in the equilibrium calculation, such a parameter cannot be derived directly. To facilitate the comparison of the experimental and simulation results on K partitioning, it is assumed that K chloride and sulphate are the only source of water soluble K in ash. With this assumption, “the percentage of K present as water soluble form” in equilibrium calculation can be approximated as the percentage of K present as chloride and sulphate at different temperature. In this way, a comparison between the experimental and simulation results on K partitioning is possible. Similar approach is also used to compare the partitioning of Na in EFR experiments and equilibrium calculations. For the partitioning of S, “the percentage of S retention in ash” in the EFR experiments was obtained by performing mass balance calculation on the sulphur in ash and that in the original fuel. In the equilibrium calculation, “the percentage of S retention in ash” was derived from the percentage of sulphur found in solid/liquid phase at different temperature.

3. Results and discussion

3.1 Influence of SRF share

3.1.1 Distribution of K species

Fig. 1 shows the equilibrium distribution of the K, Na, S and Cl species in dedicated coal combustion, co-combustion of coal and 25% SRF (weight basis), and 100% SRF combustion. It can be seen that the distribution of K in co-combustion of coal and 25% SRF is similar as that in dedicated coal combustion. In both cases, the majority of K (>98%) is present as KAlSi_2O_6 (s2) in the temperature range of 690–1400 °C. The KAlSi_2O_6 (s2) is converted to KAlSi_2O_6 (s) in the temperature range of 690–680 °C, and a transition of KAlSi_2O_6 (s) to K_2SO_4 (s2) occurs at temperature below 620 °C. The presence of KCl (g) is generally negligible at temperature below 1000 °C. However, in the temperature range of 1000–1400 °C, the presence of small percentage of KCl (g) is observed in both cases. Compared with dedicated coal combustion, the formation of KCl (g) is slightly enhanced by co-firing coal with 25% SRF. At 1400 °C, approximately 0.3% of K is present as KCl (g) in dedicated coal combustion, whereas in co-combustion of coal and 25% SRF it is about 2.0%. The distribution of K species in 100% SRF combustion is significantly different from that in co-combustion of coal and 25% SRF. In the temperature range of 900–1400 °C, it can be seen that approximately 20% of K is present as KCl (g) in 100% SRF combustion, with the remaining K being KAlSi_2O_6 (s2). In the temperature range of 800–900 °C, part of the KCl (g) and KAlSi_2O_6 (s2) is converted to $\text{K}_2\text{Ca}_2(\text{SO}_4)_3$ (s). The formed $\text{K}_2\text{Ca}_2(\text{SO}_4)_3$ (s) is completely decomposed to K_2SO_4 (s) and CaSO_4 (s) in a narrow temperature range around 800 °C, and the remaining KCl (g) is converted to K_2SO_4 (s) in the temperature range of 700–800 °C.

The distribution of K species at different SRF share conditions clearly illustrates the benefits of co-combustion of coal and SRF. In 100% SRF combustion, a significant percentage of K would be present as KCl (g) at temperatures higher than 700 °C, which may induce ash deposition and corrosion problems in the convective part of the boilers. By co-combustion of coal and 25% SRF, the K species at temperature higher than 700 °C is dominated by KAlSi_2O_6 (s2), thus the ash related problems caused by KCl (g) may be prevented significantly. Compared with dedicated coal combustion, the percentage of K present as KCl (g) is only slightly increased in co-combustion of coal and 25% SRF, and the effect is particularly not obvious at temperatures lower than 1100 °C. This suggests that co-firing coal and 25% SRF would not significantly influence the K partitioning in a coal-fired power plant, especially in the convective part of the boiler with flue gas temperature about 800–1000 °C.

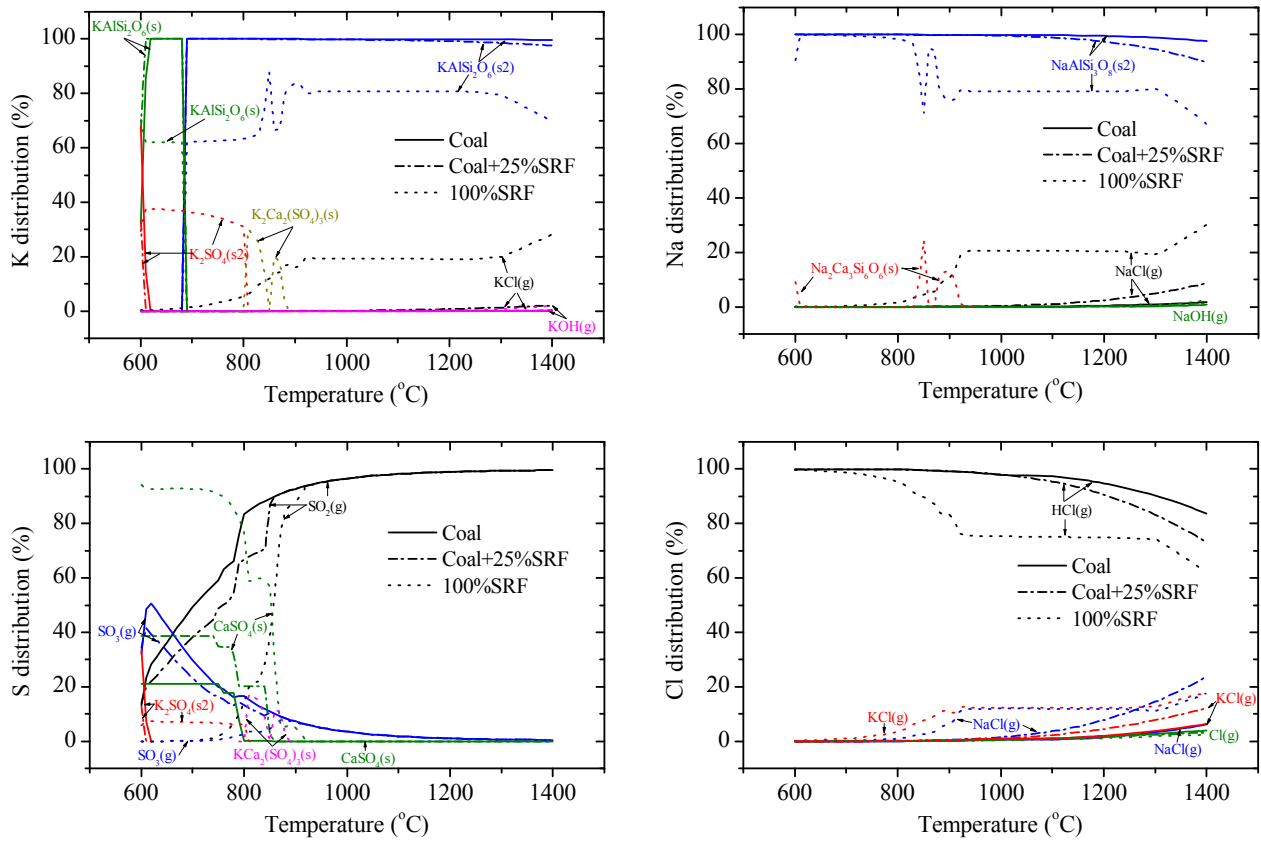


Fig. 1 Equilibrium distributions of K, Na, S and Cl species in dedicated coal combustion, co-combustion of coal and 25 wt% SRF, and 100% SRF combustion.

3.1.2 Distribution of Na species

As shown in Fig. 1, the partitioning of Na species in dedicated coal combustion and co-combustion of coal with 25% SRF is also similar. In both cases, $\text{NaAlSi}_3\text{O}_8$ (s2) is the dominant Na species in the temperature range of 600–1400 °C. The presence of NaCl (g) is only observed at temperatures higher than 1000 °C, and the percentage of Na present as NaCl (g) increases with increasing temperature. Compared with dedicated coal combustion, the formation of NaCl (g) is increased considerably in co-combustion of coal and 25% SRF, especially at high temperatures. At 1400 °C, approximately 1.7% of Na is present as NaCl (g) in dedicated coal combustion, whereas the percentage of NaCl (g) in co-combustion of coal and 25% SRF is about 8.7%. The behavior of Na in 100% SRF combustion is quite different from that in co-combustion of coal and 25%. It can be seen that, about 20% of the Na is present as NaCl (g) at temperatures higher than 920 °C, and the NaCl (g) is converted to $\text{NaAlSi}_3\text{O}_8$ (s2) or $\text{Na}_2\text{Ca}_3\text{Si}_6\text{O}_6$ (s) in the temperature range of 920–700 °C.

From the results of Na distribution, it can be seen that a significant fraction of Na is present as NaCl (g) in moderate temperature (800–1000 °C) during 100% SRF combustion. By co-firing of coal and 25% SRF, the presence of NaCl (g) in moderate temperature (800–1000 °C) can be significantly reduced, at least from thermodynamic point of view. This illustrates the advantages of co-combustion on reducing the corrosive species NaCl (g). Compared with dedicated coal combustion, the percentage of Na present

as NaCl (g) in the temperature range of 1000–1400 °C is considerably increased by co-firing of coal and 25% SRF, whereas the percentage of NaCl (g) below 1000 °C is not significantly different.

3.1.3 Distribution of S species

The distribution of S species at temperatures higher than 910 °C is almost identical in dedicated coal combustion, co-combustion of coal and 25% SRF, and 100% SRF combustion. Above 910 °C, all of the sulphur is present in the gas phase, with the majority (>90%) being SO₂ (g) and the remaining being SO₃ (g). The S distribution below 910 °C is considerably different for different cases. In dedicated coal combustion, it can be seen that all of the S is remaining in gas phase above 800 °C. In the temperature range of 800–750 °C, a transition from gaseous sulphur to CaSO₄ (s) occurs, resulting in about 20% of sulphur being converted to CaSO₄ (s). When the temperature is decreased to 620 °C, there is a further transformation of gaseous sulphur to solid sulphur, via the formation of K₂SO₄ (s₂). Compared with dedicated coal combustion, the initial temperature for CaSO₄ (s) formation is advanced to 850 °C in co-combustion of coal and 25% SRF, and about 40% of sulphur is finally converted to CaSO₄ (s). A transition of gaseous sulphur to K₂SO₄ (s) is also observed in co-combustion at temperatures below 610 °C. For 100% SRF combustion, it can be seen that the formation of CaSO₄ (s) mainly happens in the temperature range of 910–750 °C, resulting in about 90% of sulphur being converted to CaSO₄ (s). The formation of K₂Ca₂(SO₄)₃ (s) is observed in the temperature range of 900–800 °C, and it is further transformed to K₂SO₄ (s₂) and CaSO₄ (s) at temperatures below 800 °C. At 750 °C, the presence of gaseous sulphur is almost negligible in 100% SRF combustion, as the majority of sulphur (>99%) would be existing as K₂SO₄ (s₂) and CaSO₄ (s). The results of sulphur distribution reveal that the retention of sulphur in solid phase is mainly achieved through the formation of CaSO₄ (s). By co-firing coal with SRF, both the initial temperature of CaSO₄ formation and the percentage of sulphur present in solid phase can be increased.

3.1.4 Distribution of Cl species

As shown in Fig. 1, the major chlorine species present in dedicated coal combustion, co-combustion of coal and SRF, or 100% SRF combustion are HCl (g), NaCl (g), KCl (g), and Cl (g). In dedicated coal combustion and co-combustion of coal with 25% SRF, it can be seen that HCl (g) is almost the only stable chlorine species at temperatures below 1000 °C. At temperatures above 1000 °C, part of the HCl (g) is converted to KCl (g) and NaCl (g), and percentage of chlorine present as KCl (g) or NaCl (g) increases with increasing temperature. In dedicated coal combustion, KCl (g) and NaCl (g) represents for about 6.3% and 6.2% of the chlorine at 1400 °C. However, in co-combustion of coal and SRF, the percentage of chlorine present as KCl (g) and NaCl (g) at 1400 °C is about 23.7% and 12.0% respectively. The behavior of chlorine in 100% SRF combustion is somewhat different. It can be seen that at temperatures higher than 910 °C, the contributions of NaCl (g) and KCl (g) are both about 10% of the total chlorine. In the temperature range of 910–700 °C, the NaCl (g) and KCl (g) are transformed to HCl (g). Below 700 °C, HCl (g) is the dominant chlorine species in 100% SRF combustion.

The partitioning of chlorine species indicates that co-combustion of coal and 25% SRF would greatly shift the distribution of chlorine towards NaCl (g) and KCl (g) at temperatures higher than 1000 °C, as compared with that in dedicated coal combustion. However, due to the relatively low chlorine content in the fuel mixture (see Table 1), such a transition in chlorine distribution would not significantly affect the formation of NaCl (g) and KCl (g), which is reflected by the distribution of Na and K species in these condition. For 100% SRF combustion, the formation of NaCl (g) and KCl (g) would be greatly affected by the distribution of chlorine species because of the relatively high chlorine content in the SRF (see Table 1). Such an effect is illustrated by the distribution of Na and K species in 100% SRF combustion.

3.1.5 Comparison with experiments

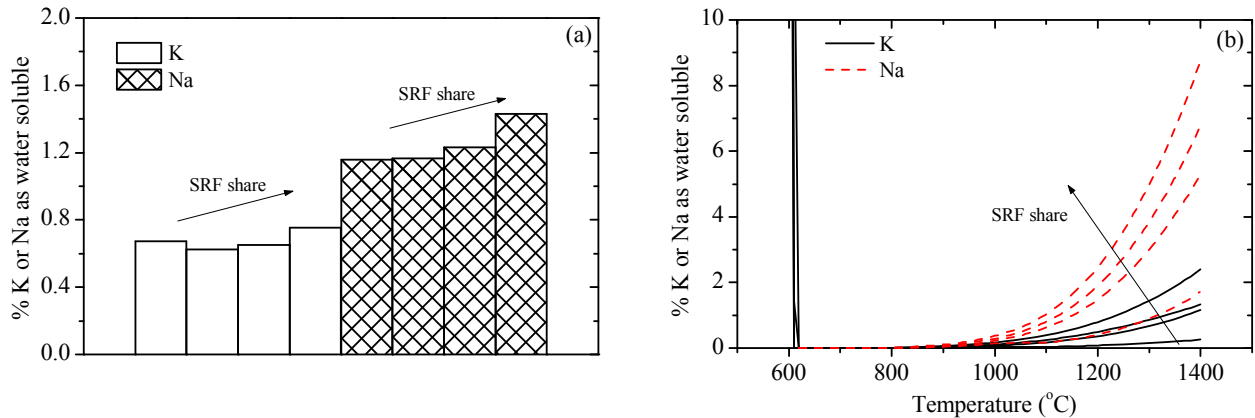


Fig. 2 (a) Percentage of K or Na present as water soluble form in the fly ashes from the EFR experiments, with SRF share of 0%, 7.9%, 14.8% and 25%; (b) Simulated percentage of K or Na present as water soluble form at different temperature, with SRF share of 0%, 7.9%, 14.8% and 25%.

Fig. 2a shows the percentage of K or Na present as water soluble form in the fly ashes from EFR experiments with different share of SRF. It is seen that percentage of K present as water soluble form is in a range of 0.62–0.75% at different co-combustion experiments, whereas the percentage of Na present as water soluble form is generally higher than that of K, which is in a range of 1.16–1.43%. For K, it seems that the different SRF share does not have a noticeable effect on the percentage of K present as water soluble form. For Na, it seems that the percentage of Na present as water soluble form is slightly increased in the experiments with higher share of SRF, particularly for the experiment with 25% SRF.

The simulated percentage of K or Na present as water soluble form at different SRF share conditions is given in Fig. 2b. The significant increase of the water soluble K observed at temperatures below 620 °C is due to a conversion from K-aluminosilicates to K-sulphate. However, such a conversion is not likely to happen in the experiments due to the short residence time in this temperature range. By neglecting the results below 620 °C, Fig. 2b shows that at equilibrium temperature of 800 °C all of the K and Na species would be present as aluminosilicates which cannot be dissolved by water. At equilibrium temperature of 1000 °C, the percentage of K present as water soluble form is in a range of 0.04–0.17% (increases with SRF share), and the percentage of Na present as water soluble form is in a range of 0.14–0.37%. Similar parameter at 1400 °C is 0.25–2.41% for K and 1.73–8.76% for Na. The general tendency reflected by Fig. 2b is that the percentage of Na present as water soluble form is normally higher than that of K. In addition, the percentage of K and Na present as water soluble form increases with increasing share of SRF.

Through comparing the experimental and simulation results, it is obvious that global equilibrium is not reached in the EFR experiments, since water soluble K and Na species are detected in the fly ash collected at about 600 °C. This indicates that part of the KCl (g) and NaCl (g) which is thermodynamically stable at high temperature is not totally converted to alkali-aluminosilicates when the flue gas is cooled to about 600 °C. There are several possible reasons for the observed discrepancies between the experimental and simulation results. The conversion of KCl (g) and NaCl (g) to aluminosilicates may be kinetically limited. The total residence time of the fuel particles in the EFR is around 1s, and the residence time in the temperature range of 1200–600 °C is even shorter. In such short

residence time, it may be difficult for the reactions between alkali chlorides and aluminosilicates to reach equilibrium. Besides the kinetic limitations, it has been found that the reaction of gaseous alkali and aluminosilicates (such as Kaolinite) would mainly occur on the surface of aluminosilicates particles, as the formation of alkali-aluminosilicates may result in a catastrophic melt and deactivate the aluminosilicates particles [13]. Thus the aluminosilicates that are available for reacting with KCl (g) and NaCl (g) would be less than that considered in the equilibrium calculation. Furthermore, it is well-known that the association of inorganic species in coal is complicated, with a significant fraction of inorganic species present as included or excluded minerals such as quartz, kaolinite and illite [22]. In a practical combustion system, the interactions among these mineral particles (particularly for the excluded minerals) are limited, as it would be difficult for these particles to meet and reach global equilibrium condition.

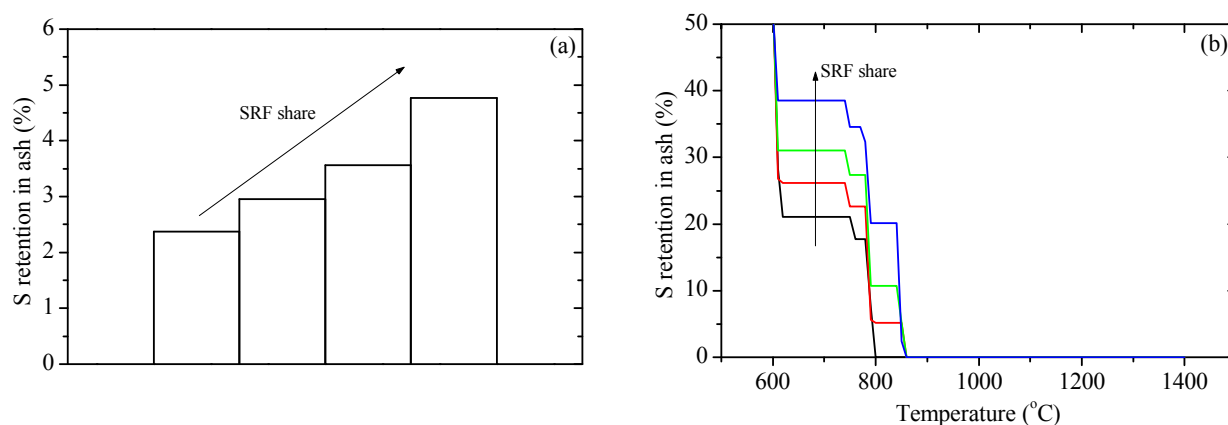


Fig. 3 (a) Percentage of fuel sulphur retained in the ashes from the EFR experiments, with SRF share of 0%, 7.9%, 14.8% and 25%; (b) Simulated percentage of fuel sulphur retained in ashes, with SRF share of 0%, 7.9%, 14.8% and 25%

Some qualitative agreements are achieved between the experimental and simulation results. In the experiments, the percentage of Na present as water soluble form is found to be higher than that of K, which is consistent with the tendency in the equilibrium calculation. This indicates that the formation of NaCl (g) is more thermodynamically favorable than the formation of KCl (g) at the simulated co-combustion conditions. In addition, the experimental results show that the percentage of Na present as water soluble form increases with increasing share of SRF. Similar trend is also seen in the simulation results. However, the predicted percentage of Na present as water soluble form in co-combustion of coal and 25% SRF is always several times (>2) higher than that in dedicated coal combustion. This deviates from the experimental results which find that increasing the SRF share only slightly enhances the formation of water soluble Na. One of the possible explanations is that the effect of Ca on the partitioning of alkali species may be over-predicted in the equilibrium calculation. The distribution of Si species shows that the formation of $\text{CaAl}_2\text{Si}_2\text{O}_8$ (s2) is greatly increased with increasing SRF share, due to the relatively high Ca content in SRF. This would inhibit the partitioning of Na to Na-aluminosilicates and promote the formation of NaCl (g) . However, in a practical combustion system, the majority of the Ca in the SRF would be in solid phase at high temperature, and the interactions between the aluminosilicates in coal and the Ca in SRF would be quite limited. By neglecting the effect of Ca in simulation (i.e. assuming there is no Ca in SRF), the percentage of Na present as water soluble form is reduced to 0.14% at 1000 °C and 4.41% at 1400 °C during co-combustion coal and 25% SRF. The results clearly show that if the Ca in SRF is neglected in equilibrium calculation the partitioning of Na to NaCl (g) can be inhibited,

and the obtained simulation results would be closer to the experimental results. The Ca in SRF has similar impact on the partitioning of K, i.e. the formation of $\text{CaAl}_2\text{Si}_2\text{O}_8$ (s2) would promote the partitioning of K to KCl (g). This may partly explain the more pronounced effect of SRF share on K partition in the equilibrium simulation, as compared with the experimental results.

Fig. 3a illustrates the percentage of fuel sulphur retained in ash during the EFR experiments with different SRF share. It can be seen that the sulphur retention is increased with increasing share of SRF. For dedicated coal combustion, the sulphur retention in ash is about 2.4 %, and the value for co-combustion of coal and 25% SRF is 4.8%. The sulphur retention in ash predicted from the equilibrium calculation is depicted in Fig. 3b. Similar as aforementioned, the simulation results below 620 °C should be ignored as a conversion from K-aluminosilicates to K-sulphate is unlikely to occur in practical. According to the simulation results above 620 °C, it can be seen that the predicted sulphur retention increases with increasing share of SRF, mainly due to the relatively high Ca content in the SRF. Such an increasing tendency is in agreement with the experimental results. However, from quantitative point of view, the sulphur retention predicted by equilibrium modeling is about 10 times higher than that in the experiments. The significant differences between experimental and simulation results are likely related with kinetic limitations. According to the equilibrium calculation results shown in Fig. 1, the retention of sulphur in ash is mainly through the formation of CaSO_4 (s), which occurs at temperatures below 850 °C. In the EFR, the residence time at the flue gas temperature below 850 °C is estimated to be quite short (<0.1s), which would be a main limiting factor for the sulphur retention to approach equilibrium.

3. 2 Influence of PVC addition

The distribution of K, Na, S and Cl species during the addition of 2% and 4% PVC are depicted in Fig. 4. As compared with the results in Fig. 1, it can be seen that the addition of PVC has greatly affected the distribution of K and Na in co-combustion of coal and SRF, whereas the influences on the S and Cl distribution are not pronounced. With the addition of PVC to the mixture of coal and SRF, the formation KCl (g) is increased considerably in the temperature range of 850–1400 °C. With 2% and 4% PVC addition, the equilibrium percentage of KCl (g) at 1400 °C is 11.6% and 22.3% respectively, which is significantly higher than the percentage without PVC addition which is 1.3%. For Na distribution, the effect of PVC addition is even more pronounced. At 1400 °C, the percentage of Na present as NaCl (g) is 62.0% and 99.4% respectively for 2% and 4% PVC addition, whereas the percentage of NaCl (g) in co-combustion of coal and 14.8% SRF is only about 6.7%.

The simulation results imply that an increase of the chlorine content in the fuel mixture would induce a transition of alkali-aluminosilicates (KAlSi_2O_6 and $\text{NaAlSi}_3\text{O}_8$) to alkali chlorides (KCl and NaCl), particularly at temperatures above 1000 °C. The occurrence of such transformations may result in a significant increase of the KCl (g) and NaCl (g) concentration in the flue gas, and may cause severe ash deposition and corrosion in the convective part of the boilers. Therefore it would be important to control the chlorine content in SRF during co-combustion of coal and SRF.

The influence of PVC addition on the partitioning of alkali species in a practical combustion system may deviate from that predicted by equilibrium calculation. Fig. 5a shows the percentage of K or Na present as water soluble form in the fly ash from the EFR experiments without PVC addition and with 2% PVC addition. It can be seen that with the addition of 2% PVC, the percentage of K present as water soluble form is increased from 0.65% to 2.76%, and the percentage of water soluble Na is increased from 1.23% to 3.67%. However, the experimental results for 4% PVC addition are not available due to the amount of ash collected in the experiments is not sufficient for analysis.

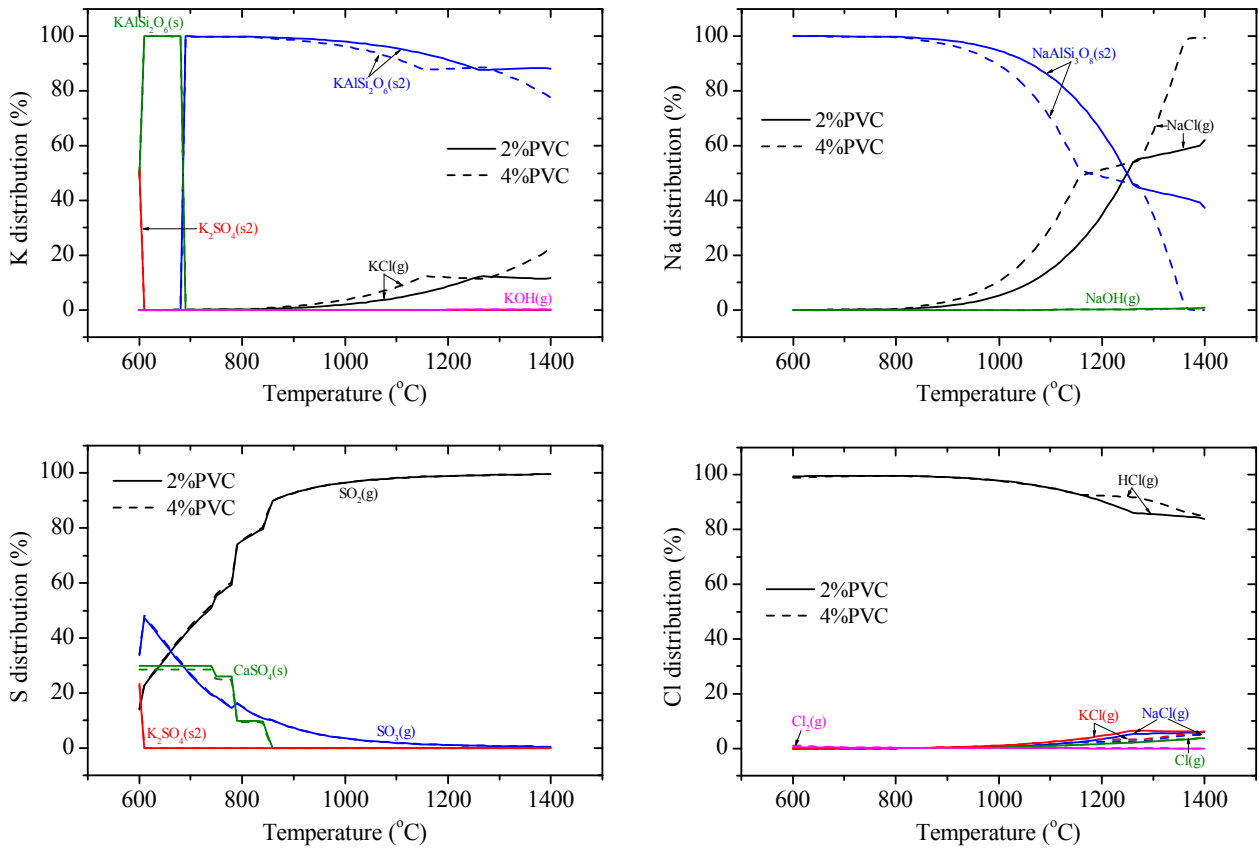


Fig. 4 Equilibrium distributions of K, Na, S and Cl species during the addition of 2% PVC and 4% PVC to the mixture of coal and SRF, with the mass share of coal fixed at 85.2%.

The estimated percentage of K or Na present in water soluble form is given in Fig. 5b. It is seen that the formation of water soluble K and Na at temperatures higher than 850 °C is greatly promoted by the addition of 2% PVC. As an example, the predicted percentage of K present as water soluble form at 1000 °C is 0.11% for the case without PVC addition, and is 1.92% for the case with 2% PVC addition. For Na, the simulated percentage of Na present as water soluble form at 1000 °C is 0.28% for the case without PVC addition, and 5.24% for the case with 2% PVC addition. At temperatures below 800 °C, the presence of water soluble alkali is generally negligible. However, a significant increase of the water soluble K at temperatures below 620 °C is observed. Such an increase is because of the formation of K-sulphate from K-aluminosilicates, but it is not likely to happen in the experiments due to the limitation of residence time.

By comparing the experimental results with the simulation results, it is clearly that global equilibrium is not reached in the EFR experiment with PVC addition. This suggests that not all of the KCl (g) or NaCl (g) appearing at high temperature are converted to aluminosilicates during the flue gas cooling in the EFR, with the possible reasons given in Section 3.1.5. However, there are some qualitative agreements between the experimental and simulation results. Both results show the tendency that the percentage of K or Na present as water soluble form is increased by the addition of 2% PVC, and the percentage of Na present as water soluble form is generally higher than that of K. This implies that the predicted transition

of alkali-aluminosilicates to alkali chlorides during PVC addition may occur in the EFR experiment, and is most likely a primary reason for the increased water soluble alkali content in fly ash. However, in the equilibrium calculation, the percentage of K and Na present as water soluble form is predicted to be increased by a factor of at least 8 when 2% PVC is added to the mixture of coal and SRF, whereas the percentage of K or Na present as water soluble form is only increased by a factor about 3 in the EFR experiment. This discrepancy suggests the effect of PVC on the partitioning of K and Na may be over-predicted by the equilibrium calculation. In the EFR experiment, the transition of alkali-aluminosilicates to alkali chlorides during PVC addition may be kinetically limited, thus the actually alkali that present as alkali chlorides would be less than predictions. In addition, not all of the PVC particles would be immediately decomposed to HCl (g) upon injection, which would also confine the combustion system to reach equilibrium at high temperatures.

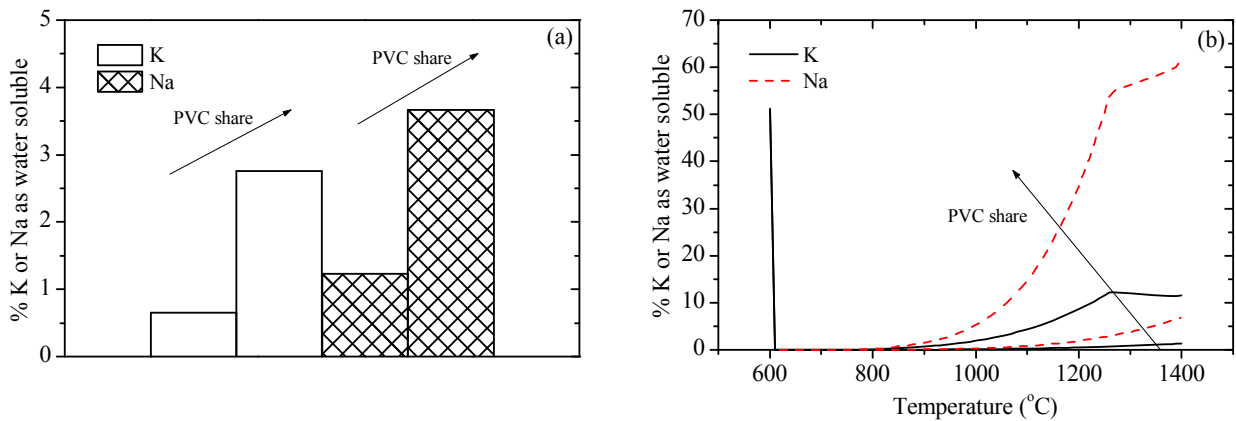


Fig. 5 (a) Percentage of K or Na present as water soluble form in the fly ashes from the EFR experiments with 0% PVC addition (coal+14.8% SRF) and with the addition of 2% PVC (coal+12.8% SRF+2% PVC) (b) Simulated percentage of K or Na present as water soluble form at different temperature, with 0% PVC addition and with the addition of 2% PVC

3. 3 Influence of NaCl addition

Fig. 6 shows the distribution of K, Na, S and Cl species when 1% and 2% NaCl are added to the mixture of coal and SRF. As compared with the results in Fig. 1, it can be seen that the influences of NaCl addition are significant on the distribution of these species in co-combustion. For K species, the addition of NaCl significantly increases the formation of KCl (g) at temperatures higher than 800 °C. As an example, the percentage of K present as KCl (g) at 1000 °C is 5.7% and 39.6% respectively with the addition of 1% and 2% NaCl, while the percentage in co-combustion of coal and 14.8% SRF is only about 0.1%. Similar as the tendency of KCl (g), the formation of NaCl (g) at temperatures above 800 °C is also increased considerably when 1% and 2% NaCl is added to the mixture of coal and SRF. The percentage of Na present as NaCl (g) at 1000 °C is 9.4% and 25.1% for the addition of 1% and 2% NaCl respectively, whereas the percentage of NaCl (g) in co-combustion of coal and 14.8% SRF is approximately 0.3%. Besides the influence on NaCl (g) distribution, the addition of 1% NaCl also leads to the formation of NaAlSiO₄ (s) in the temperature range of 780–1060 °C as well as the formation of Na₂SO₄ (s) in the temperature range of 690–770 °C. For the addition of 2% NaCl, it can be seen that NaAlSiO₄ (s,s2) is present in the temperature range of 760–1350 °C. In addition, the formation of Na₂SO₄ (liq) is observed in the temperature range of 890–960 °C, and Na₂SO₄ (s2) is present at temperatures below 890 °C. From the distribution of chlorine species, it can be seen that the addition of

NaCl has greatly shifted the distribution of chlorine from HCl (g) to alkali chlorides. As an example, at 1000 °C HCl (g) represents for about 98% of the total chlorine in co-combustion of coal and 14.8% SRF. However, the percentage of HCl (g) is decreased to 87.4% with the addition of 1% NaCl, and it is further decreased to 54.4% with the addition of 2% NaCl.

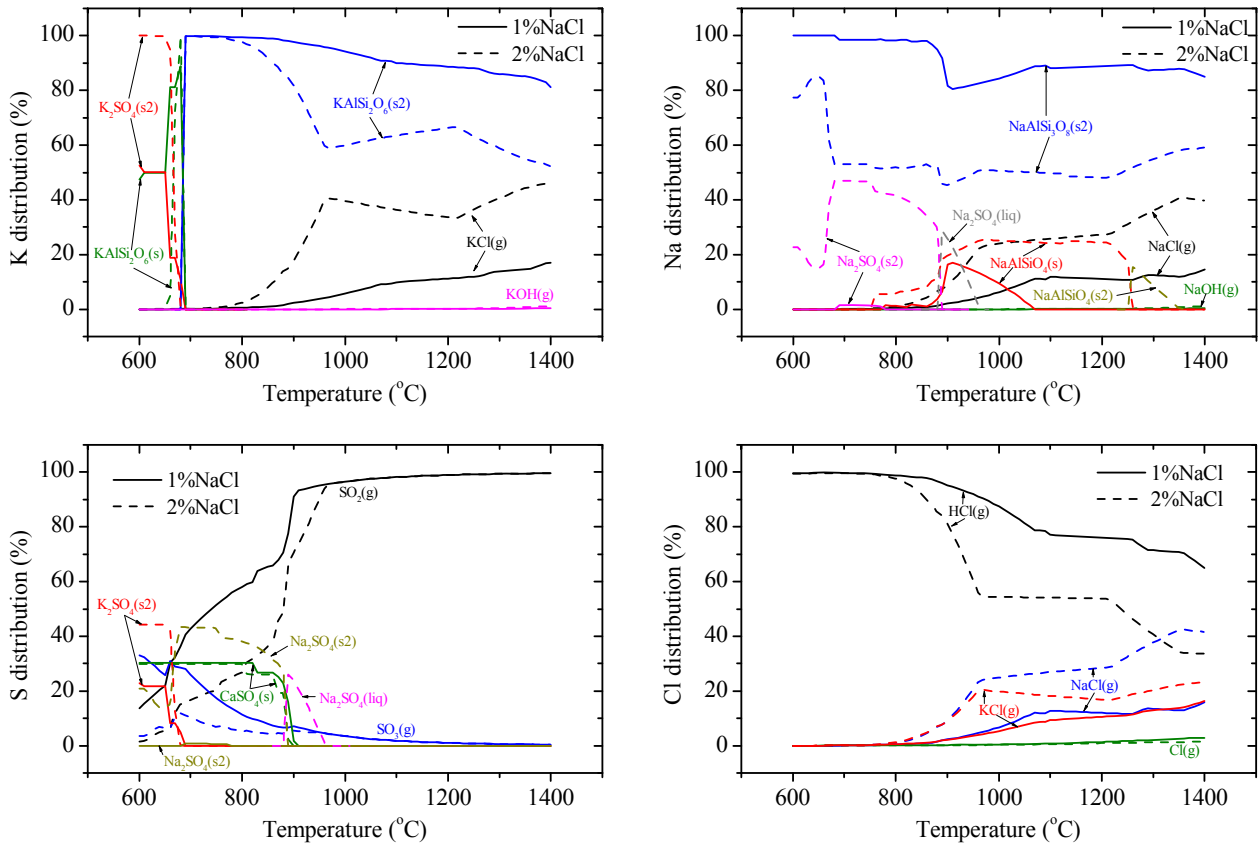


Fig. 6 Equilibrium distributions of K, Na, S and Cl species during the addition of 1% NaCl and 2% NaCl to the mixture of coal and SRF, with the mass share of coal fixed at 85.2%.

The simulation results shown in Fig. 6 demonstrate that the addition of NaCl to the mixture of coal and SRF would considerably increase the concentration of KCl (g) and NaCl (g) at temperatures above 800 °C. This is mainly due to that the addition of NaCl would shift the partitioning of alkali species from alkali aluminosilicates to alkali chlorides at high temperatures. Moreover, the addition of NaCl also increases the content of Na in the fuel mixture, thus the concentration of NaCl (g) can be increased even if the partitioning of Na species is not changed. The high concentration of KCl (g) and NaCl (g) at temperature range of 800–1000 °C may lead to significant ash deposition and corrosion in the convective part of the boilers.

Fig. 7a shows the percentage of K and Na present as water soluble form in the fly ashes from the EFR experiments with different share of NaCl (0%, 1% and 2%). It can be seen that the percentage of K and Na present as water soluble form increases with increasing share of SRF, particularly for the case with 2% NaCl addition. When 2% NaCl is added to the mixture of coal and SRF, the percentage of K and Na present as water soluble form in fly ash is 16.5% and 20.7% respectively, which is significantly higher

than that in 1% NaCl addition (2.8% for K and 4.5% for Na) and that in 0% NaCl addition (0.65% for K and 1.23 for Na).

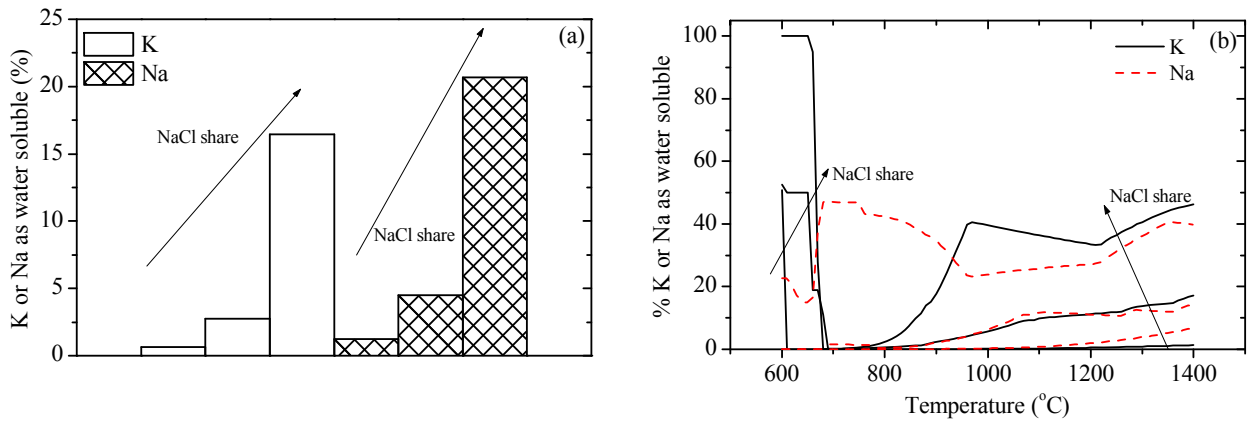


Fig. 7 (a) Percentage of K or Na present as water soluble form in the fly ashes from the EFR experiments without NaCl addition (coal+14.8% SRF), and with the addition of 1% NaCl (coal+13.8% SRF+1% NaCl) and 2% NaCl (coal+12.8% SRF+2% NaCl) (b) Simulated percentage of K or Na present as water soluble form at different temperature, without PVC addition, and with the addition of 1% and 2% NaCl.

The simulated percentage of alkali present as water soluble form is illustrated in Fig. 7b. It is seen that the presence of water soluble K is almost negligible in the temperature range of 700–800 °C, as the majority of K would be present as KAlSi_2O_6 (s2). The significant increase of water soluble K below 700 °C is because of a conversion from KAlSi_2O_6 (s2) to K_2SO_4 (s2), which is not likely to happen in the EFR experiments due to the short residence time in the temperature range (<0.1 s). The presence of water soluble K above 800 °C is mainly due to the formation of KCl (g), and it can be seen that the percentage of K present as water soluble form increases significantly with increasing share of NaCl. For Na, the presence of water soluble Na is only observed above 800 °C for the cases with 0% and 1% NaCl addition. However, under the condition with 2% NaCl addition, the presence of water soluble Na is found at the whole temperature range. At temperatures above 980 °C, NaCl (g) is the dominant water soluble Na species, whereas the presence of Na_2SO_4 (liq, s2) is found at temperatures below 980 °C and becomes the dominant Na species below 800 °C. In general, the percentage of Na present as water soluble form is found to be increased with increasing share of NaCl.

By comparing the experimental and simulation results, it is clear that the equilibrium distribution of K and Na species at 600 °C do not represent the distribution of these species in the fly ashes collected in the EFR. The presence of water soluble K and Na in fly ash is most likely because of the NaCl (g) and KCl (g) present at high temperatures has not been totally converted to aluminosilicates during flue gas cooling. From Fig. 6, it can be seen that the conversion of alkali chlorides to alkali aluminosilicates during NaCl addition mainly happens in the temperature range of 800–1100 °C. In the EFR experiments, the residence time within this temperature range is short (<0.5 s), suggesting that the conversion of alkali chlorides to alkali aluminosilicates may be kinetically limited. This hypothesis is supported by the obtained percentage of water soluble K and Na in fly ash, which is also in between the equilibrium values at 1100 °C and 800 °C.

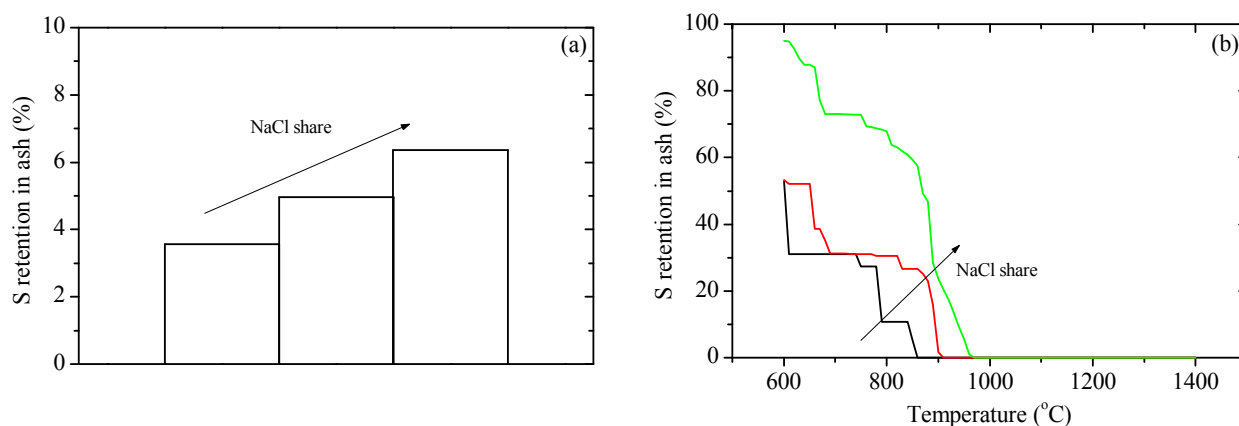


Fig. 8 (a) Percentage of sulphur retention in ash in the EFR experiments without NaCl addition (coal+14.8% SRF), with the addition of 1% NaCl (coal+13.8% SRF+1% NaCl) and 2% NaCl (b) Simulated percentage of sulphur retention in ash at different temperature, without PVC addition, and with the addition of 1% and 2% NaCl.

There are some qualitative agreements between the simulation results and experimental results. Both results show that the addition of NaCl would increase the percentage of K and Na present as water soluble form, and the effect is especially pronounced with 2% NaCl addition. For K, it is likely that the addition of NaCl would induce a transition of potassium aluminosilicates to KCl (g) in the EFR experiments. From Fig. 6, it can be seen that such transition is thermodynamically favorable at a temperature range of 1000–1400 °C. In general, the reaction rates would be faster at higher temperatures and it would be easier to achieve equilibrium. Therefore, it is likely that a significant increase of KCl (g) has happened in the high temperature zones (>1000 °C) of the EFR, when NaCl is added to the mixture of coal and SRF. Although the formed KCl (g) is expected to be converted to K-aluminosilicates at temperatures lower than 1000 °C, the conversion is probably quite limited in the EFR experiments, as indicated by the significantly high water soluble K content (~16.5%) in the fly ash collected during 2% NaCl addition. Similar reasons may be attributed to the increase of water soluble Na in the fly ash collected during NaCl addition. For Na, another possible reason is that part of the added NaCl may not be totally vaporized in the EFR which would also increase the percentage of Na present as water soluble form in fly ash. However, for the case with 2% NaCl addition, the added NaCl represents for above 75% of the Na in the fuel, whereas the Na present as water soluble form in fly ash is only about 21%. This implies that a significant fraction of the added NaCl has been converted to aluminosilicates during the experiments. Such conversion is thermodynamic favorable in the temperature range of 1000–1400 °C, since the equilibrium percentage of Na present as NaCl (g) is about 20–40% which is much lower than that in the fuel.

Fig. 8a shows the percentage of sulphur retention in ash in the EFR experiments without NaCl addition and with the addition of 1% and 2% NaCl. It can be seen that the sulphur retention increases with increasing share of NaCl. According to the simulated sulphur retention shown in Fig. 8b, it is likely that the increased sulphur retention during 1% NaCl addition is related with an increase of the initial temperature for CaSO_4 (s) formation. It can be seen that the initial temperature for CaSO_4 (s) formation is advanced by approximately 50 °C with the addition of 1% NaCl. Therefore, the sulphur retention in ash would be increased as the reaction rate between gaseous sulphur and calcium species would be faster at higher temperature, and the reaction time would be longer. For the addition of 2% NaCl, an additional reason for the increased sulphur retention is that part of the NaCl (g) can be converted to Na_2SO_4 (liq, s2)

at temperatures lower than 960 °C. Although the tendency agrees well, it should be noted that the experimental sulphur retention is found to be significantly lower than the predictions, suggesting that this process may be kinetically limited.

5. Conclusion

Co-combustion of coal and SRF has been studied by global equilibrium calculations. It is found that the equilibrium distribution of the K and Na species in co-combustion of coal and 25% SRF is quite similar as that in dedicated coal combustion, but deviates significantly from that in 100% SRF combustion. The results suggest that the co-combustion of coal and SRF could significantly reduce the formation of NaCl (g) and KCl (g) in moderate temperature (800–1000 °C), as compared with the situation in 100% SRF combustion. It indicates that co-combustion of coal and SRF may be a feasible method to increase the electrical efficiency of utilizing waste, as the aluminosilicates content in the coal would mitigate the adverse effect caused by the relative high chlorine and alkali content in the SRF. In addition, the retention of sulphur in the ashes may be increased by co-combustion of coal and SRF, mainly due to the relatively high calcium content in the SRF. The comparisons between experimental results from an entrained flow reactor and the global equilibrium calculation support that co-combustion of coal and 25% SRF would not alter the distribution of K and Na species in combustion significantly, and the sulphur retention in ash would be increased by co-combustion. However, quantitative discrepancies are found between the experimental and simulation results, and the possible reasons are discussed.

The influence of SRF property variation on co-combustion has been studied by adding different percentage of PVC or NaCl to the mixture of coal and SRF. The main effect of PVC addition is that the formation of KCl (g) and NaCl (g) is increased at temperatures higher than 850 °C, and the impact appears to be more significant on the formation of NaCl (g) than that of KCl (g). The simulation results reveal that the addition of PVC would induce a transition of alkali aluminosilicates to alkali chlorides (g), particularly at temperatures above 1000 °C. Such tendency is supported by the experimental results with 2% PVC addition, showing that the percentage alkali present as water soluble form is increased with 2% PVC addition. From the equilibrium calculation, it is also seen that the addition of NaCl would increase the formation of both KCl (g) and NaCl (g) at temperatures higher than 800 °C. It may result in a significant increase of the concentration of KCl (g) and NaCl (g) in moderate temperature range (800–1000 °C) and cause severe ash deposition and corrosion problems in the boiler. Comparison with experimental results suggests that the tendency of the alkali behavior in the experiments with NaCl addition is in agreement with the simulation results. The addition of NaCl to the mixture of coal and SRF would increase the formation of KCl (g) and NaCl (g) at high temperatures (>1000 °C), and a significant fraction of these KCl (g) and NaCl (g) has not been converted to aluminosilicates during the flue gas cooling in the experiments. The results suggest that it is important to control the chlorine and alkali content in the SRF, in order to facilitate co-combustion of coal and SRF in a coal-fired power plant.

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