

JINGJING LIU
 RUILIN MAN
 SHEXIA MA
 JUANSHEG LI
 QI WU
 JUANYING PENG
 and
 HUI XI

Study on ash composition and acidic gases emissions during co-combustion of domestic waste and coal

This study examines separately the composition and metal content of the ash generated during the combustion of domestic waste and combustion of coal mixed with different proportion of domestic waste and the impact of domestic waste mixing proportion on acidic gas of gases generated during fuel combustion as well as influencing factors analysis. The results show that when the garbage content is less than 60%, it can be used as fuel substitute materials of cement. SiO_2 , CaO , Al_2O_3 , Fe_2O_3 content of mixed fuel ash are more than 10%, the total content of four kinds of oxide is between 75% and 85%, alkali content ($0.658\text{K}_2\text{O}+\text{NaO}$) is between 2.71% and 6.22% , and five heavy metals (Pb, Ni, Cd, As, Hg) contents are all less than that generated during the combustion of coal alone. Mixed fuel with domestic waste can reduce NO_x and SO_2 emissions of combustion gases to a certain extent, and HCl gas emissions is increasing with increasing proportion of domestic waste. When garbage content reaches 30%, acidic gases emissions generated during combustion of mixed fuel is optimal. Combustion temperature and air flow rate influence NO_x and SO_2 gas emissions during mixed fuel combustion, but have little effect on HCl gas emissions.

Key words: domestic waste; coal; ash; heavy metals; acidic gas.

1. Introduction

With the rapid development of industrialization, domestic waste is gradually used as fuel substitute materials so that the garbage can set disposed harmlessly and recycled [1-3], which can not only reduce the

use of coal and save fossil energy, but also bring good economic benefits [4-7].

Cement industry is a major source of CO_2 emissions. With the gradual increase of China's annual cement production, carbon emissions of cement industry are also increasing, so the carbon abatement technology of cement industry has gradually attracted worldwide attention. Currently, the main ways to reduce carbon emissions of cement industry are thermal efficiency improvement, fuel substitution, clinker substitution, etc., in which the fuel substitution technology is the best way of emission reduction at present. Domestic waste can be used as a fuel substitute material of cement plant, which can not only achieve the purpose of carbon abatement of cement plant, but also can solve the problem of garbage pollution. Therefore, the research that the domestic waste can be used as a fuel substitute material of cement plant has drawn more and more researchers' attention. In recent years, coal ash is gradually used to make cement, and is co-grinded with cement clinker and cement is made. Researches of some researchers [8-11] noted, adding coal ash during the process of cement production can not only improve the micro-structure and performance of concrete to a large extent, but also improve the crack resistance and durability of concrete greatly but have good volume stability at the same time. Therefore, the research ash composition of the blended fuel and heavy metal content is of great importance.

Meanwhile, with the increasingly serious environmental problems, human environmental awareness is also growing, and the state has introduced standard for harmful ingredient emissions of relative industrial fuel combustion gas, therefore, the study on harmful gas emissions during co-combustion of domestic waste as a fuel substitute material and coal is particularly important. Alfonso AU[12], Dong Changqing[13] etc. have studied combustion gas generated during co-combustion of domestic waste and coal and found SO_2 and CO_2 gas emissions is reducing with the increase of garbage. Coal mixed with household waste burning gaseous fuels,

Messrs. Jingjing Liu, Ruilin Man, Qi Wu, School of Chemistry Engineering, and also Key Laboratory of Nonferrous Metal Resource Chemistry, Central South University, Changsha, Hunan; Juangsheng Li, Juanying Peng, Hui Xi, Changsha Environmental Protection College, Changsha, Hunan and Shexia Ma, South China Institute of Environmental Sciences, MEP, Guangzhou, China. Corresponding authors: Email: rlman@mail.csu.edu.cn and mashexia@scies.org

found with increasing dosage of garbage, SO₂, reducing CO₂ gas emissions; Chen Guoyan [14] etc. have studied the relationship between polluting gases emissions generated during garbage combustion and the temperature and found that SO₂, NO_x emissions is increasing with increasing combustion temperature; Wang Wengang [15] etc. have studied technology of polluting gas control during garbage combustion and put forward optimal control combination process; Currently, a large amount of studies focus on harmful gas emissions during the combustion of garbage alone, garbage mixture and coal alone, while acidic gases emissions during co-combustion of domestic waste and coal is rarely involved

This study examines the composition and metal content of the ash generated during the combustion of domestic waste and combustion of coal mixed with different amount of domestic waste as well as the impact of domestic waste mixing amount on acidic gas of gases generated during fuel combustion, studies the impact of combustion temperature, air flow rate on acidic gases (SO₂, NO_x, HCl) emissions during combustion of mixed fuel and aims to provide the experimental parameters during co-combustion of domestic waste as industrial fuel and coal.

2. Experiment

2.1 EXPERIMENTAL MATERIALS

Experimental material is domestic waste and coal. The domestic waste is from campus, and mainly consists of combustible domestic waste: 55% of kitchen waste (leftovers, peels and egg shells, etc.), 15% waste paper, 20% plastic and 10% cloth (mass fraction wt%, the same below). The coal selects qualified bitumite of a coal mine in Chenzhou, Hunan Province, and its heat value is 25856.9kJ·kg⁻¹.

Industrial analysis for domestic waste and coal are done according to the “Sampling and Physical Analysis of Domestic Waste” (HJ/T 313-2009) and “Industrial Analysis of Coal”(GB/T 212-2008) and elemental analyzer is used to analyze the element of sample. Their industrial and elemental analysis results are shown in Table 1.

2.2 EXPERIMENTAL CONTENT

2.2.1 Determination of ash content

Garbage and coal are prepared as mixed fuel at mixing proportions 0%, 20%, 40%, 60% and 80%; Take 40g mixed fuel of different ratio respectively with electronic analytic balance (AUY220, Shimadzu) and put them into the Muffle furnace for combustion (temperature 900°C) until they become ash, which would be put in plastic bags for dry storage; Weigh 0.1g different proportions of mixed fuel ash and measure the composition and heavy metal content of ash with inductively coupled plasma spectrometer (ICPS-7510, Shimadzu).

2.2.2 Determination of acidic gas

Set different combustion temperatures (700, 800, 900, 1000°C) and different air flow rates (0.1, 0.2, 0.3L/min), analyze emission characteristics of three acidic gases (NO_x, SO₂, HCl) of fuel gases generated during the combustion of mixed fuel (proportion of domestic waste is 30%): Put 1g fuels (coal, garbage, 50% garbage and 50% coal,) and 1g mixed fuel (proportion of domestic waste is 30%) into a tube furnace (SRJK-2.5-13, Teste) and burning for 2h, and then detect three gases (NO_x, SO₂, HCl) content.

2.3 ANALYTICAL METHOD

2.3.1 Determination of heavy metals in the ash

Determination of the eight heavy metals (Zn, Hg, Cr, Pb, Cu, Ni, Cd, As) content of gases generated during mixed fuel combustion: “Ambient air and stationary source emission - Determination of metals in ambient particulate matter Inductively coupled plasma/mass spectrometry (ICP-MS)” (HJ 657-2013)

2.3.2 Determination of acidic gas of mixed combustion flue gases

Determination of SO₂ content: “Determination of sulphur dioxide from exhausted gas of stationary source Iodine titration method” (HJ/T 56-2000);

Determination of NO_x content: “Stationary source emission-Determination of nitrogen oxide-Ultraviolet spectrophotometric method” (HJ/T 42-1999);

TABLE 1 TECHNICAL AND ELEMENT ANALYSIS OF DOMESTIC WASTE AND COAL

Raw material	Industrial analysis/%				Element analysis/%				
	Mad	Aad	Vad	FCad	w(C)	w(H)	w(O)	w(N)	w(S)
Kitchen waste	72.01	0.30	20.89	6.80	40.28	5.83	43.31	2.51	0.22
Waste paper	7.83	9.49	72.49	10.18	39.62	5.88	44.21	0.23	0.31
Plastic	0.41	0.34	91.82	7.43	60.89	8.18	3.43	0.05	0.13
Cloth	2.25	0.94	86.71	10.10	41.23	6.86	45.36	0.72	0.78
Coal	8.52	7.59	30.08	53.81	54.86	3.4	7.13	0.85	0.94

Remarks: Mad, Aad, Vad, FCad are dried basis contents.

TABLE 2 THE ASH COMPOSITION CONTENT OF THE MIXED FUEL AT DIFFERENT RATIO/(%)

Garage proportion	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	SO ₃
0%	41.35	15.75	12.14	15.65	1.67	2.94	0.78	2.83
20%	38.87	14.85	12.00	17.95	2.77	4.01	1.55	2.81
40%	36.52	13.16	10.70	19.14	3.35	4.64	1.49	2.52
60%	34.50	11.09	10.14	21.66	5.90	4.74	2.97	2.43
80%	31.69	9.58	9.49	22.28	7.37	5.70	4.89	2.15
100%	30.03	8.69	8.48	22.89	8.36	6.12	5.23	2.05

Determination of HCl content: “Stationary source emissions – Determination of hydrogen chloride – (Silver nitrate titration)” (HJ 548-2009).

3 Experimental results and analysis

3.1 MIXED FUEL ASH COMPOSITION ANALYSIS

The main contents of the cement clinker are SiO₂, CaO, Al₂O₃, Fe₂O₃, and In the “Symposium of 2013 Exchange of New Technologies Conference and Exhibition of Cement Grinding at Home and Abroad”, H. Baier etc. [16] propose that when these four ingredients of the mixed fuel are over 50% of the raw material, then it can be used as cement clinker substitute materials. “Rotary Kiln Dust Used in Cement” (JC/T 742-2009) points out that when kiln dust is used as cement clinker, its SO₃ content should be not more than 3% and alkali content (0.658K₂O+NaO) should be not greater than 8%.

The ash composition content of the mixed fuel is determined at different ratio (0%, 20%, 40%, 60%, 80%, 100% respectively). The results are shown in Table 2.

Table 2 shows that the content of the SiO₂, CaO, Al₂O₃, Fe₂O₃ in mixed fuel are higher and basically more than 10%, of which the highest is SiO₂ and is followed by CaO, and the total content of these four oxides is between 75% and 85%, which is in line with cement clinker composition standard. For all mixed fuel ash, SiO₂ content of ash is highest and reaches 41.35 percent when the coal combusts alone (garbage proportion 0%), and with the increasing proportion of domestic waste, SiO₂ content of combustion ash is decreasing and the lowest reaches 30.03% when the garbage proportion is 100%, which is mainly due to the reason that silicate is the main component of coal and garbage while its content in coal is higher than that in domestic waste; The content of CaO is increasing with the increasing proportion of domestic waste and is increasing gradually, which is mainly due to the high content of calcium and salt content in domestic waste.

At the same time, the content of SO₃ in all the mixed fuel ash is lower than 3%, which is in accordance with the requirements of the national standard.

The contents of alkali in the mixed fuel ash in Table 2 are calculated, and the results are shown in Table 3.

From Table 3, when the proportion of domestic waste is from 0% to 60%, the alkali content of combustion ash is

TABLE 3 CONTENT OF ALKALI IN THE MIXED FUEL ASH/%

Garage proportion	0%	20%	40%	60%	80%	100%
0.658K ₂ O+NaO	2.71	4.30	4.67	6.22	8.79	9.26

(0.658K₂O + NaO) between 2.71% and 6.22% but no more than 8%, but when the proportion of domestic waste is 80% and 100%, and its alkali content is 8.79%, 9.26% respectively, and both are more than 8%. Therefore, when no more than 60% domestic wastes are added to coal, ash composition of mixed fuel is in line with Portland cement production standards.

3.2 ANALYSIS OF HEAVY METALS IN MIXED FUEL ASH

According to cement clinker substitute material standard put forward by the national standard “Cement Plant Design Specifications” (GB50295-2008) and “Symposium of 2013 Exchange of New Technologies Conference and Exhibition of Cement Grinding at Home and Abroad”, the heavy metals content in mixed fuel ash should meet certain requirements in the process of cement clinker production, that is, kiln dust is used as clinker alternative material, and its Ni, Pb, As content cannot exceed 100 ppm, Cr content is not more than 150ppm, Cd content is no more than 50 ppm, and Hg content cannot exceed 2 ppm.

Table 4 shows that in addition to Mn, the other eight kinds of heavy metals content are up to ppm level. When the domestic waste content is 0% to 40%, the heavy metal contents are in line with the clinker substitute materials requirements and as increasing proportion of domestic waste content, five heavy metals contents of relatively large risk (Pb, Ni, Cd, As, Hg) are decreasing. When the proportion reaches 60%, Cr content reaches 170 ppm and exceeds the national standard; when proportion of domestic waste is 80% and 100%, the content of Cr exceeds, and the Cu content is higher. Thus, when domestic waste proportion is 60% or less in coal, ash content can reach the standards of cement clinker production, the property of which is better than ash cement clinker.

3.3 ACIDIC GAS EMISSION OF MIXED FUEL COMBUSTION GAS

3.3.1 Coal, garbage combustion alone

Control the combustion temperature to 900°C and air flow rate to 0.2L/min, then burn the coal, garbage, fuel with 50% garbage and 50% coal respectively to obtain the relationship between combustion flue gas emissions and proportion of

TABLE 4 CONTENTS OF HEAVY METALS IN THE MIXED FUEL ASH /(PPM)

Garage proportion	Zn	Mn	Cr	Pb	Cu	Ni	Cd	As	Hg
0%	160	2600	130	44	190	83	28	90	0.68
20%	220	2500	140	41	196	71	18	70	0.63
40%	330	1700	150	33	260	76	14	80	0.61
60%	160	2000	170	39	268	71	19	90	0.52
80%	190	1200	220	27	270	90	13	110	0.57
100%	220	1000	230	21	278	96	15	120	0.54

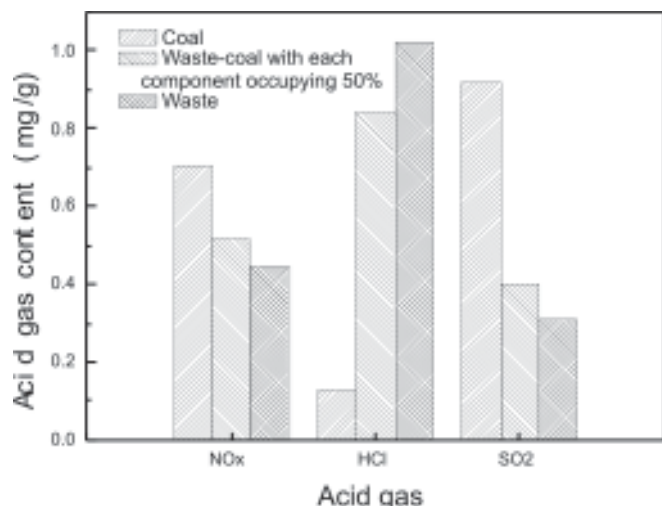


Fig.1 Acid gas emission conditions for combustion of domestic waste and coal respectively

domestic waste in fuel, as is shown in Fig.1.

Generally there are two kinds of ways generating NO_x during combustion: one is the thermal type NO_x generated by the reaction of N₂ and O₂ at the temperature at least 1300°C[17]; The other is the fuel type NO_x transformed from combustion of chemical nitrogen in the fuel. In this experiment, the combustion temperature is 900°C, therefore NO_x in the flue gas is fuel type, that means NO_x generation in the experiment cannot be affected by N₂ in air.

Figure 1 shows, NO_x content in flue gas is 0.449mg/g, SO₂ content is 0.311mg/g, and HCl content is 0.128mg/g during the combustion of domestic waste alone; NO_x content in flue gas is 0.704mg/g, SO₂ gas content is 0.918mg/g, and HCl gas content is 0.128mg/g during the combustion of coal alone, which are far higher than the NO_x and SO₂ content of domestic waste combustion flue gases; While NO_x, SO₂ content in fuel gases released during the combustion of 50% coal and 50% garbage are between the two, and the emissions are below half of the two's sum. This shows NO_x, SO₂ content released during the combustion of domestic waste is lower than that of coal, and the mixing with domestic waste helps reduce NO_x, SO₂ emissions of fuel combustion. Coal nitrogen content is higher than domestic waste, and reducing gas released during combustion of domestic waste reduces NO_x into N₂, therefore, the mixing with domestic waste can

reduce NO_x emissions from coal to a certain extent; secondly, coal contains high pyrite and elemental sulfur, whose sulfur content is much higher than domestic waste, and elemental sulfur and pyrite will produce SO₂ at high temperature combustion, therefore, SO₂ released during coal combustion is much higher than that released during domestic waste combustion.

HCl gas content released during coal combustion alone is 0.128mg/g, well below HCl gas content which is 1.021mg/g released during domestic waste combustion, which is mainly due to the chlorine content in coal is lower than the domestic waste, and the chlorine in coal generally present in the form of inorganic salts, and it is difficult to generate HCl gases during combustion, while most chlorine in garbage present in the form of organic and can be easily converted to HCl gas during combustion.

We can also see from the Figure 2 that HCl content the combustion emissions released during the combustion of 50% coal and 50% garbage is between the two and higher than half of their sum, indicating that HCl gas emission becomes more as the increasing mixing proportion of domestic waste. HCl gas detection is an important part of combustion flue gas detection in the cement plant, the HCl gas in the flue gas reacts with CaO, Ca(OH)₂ and CaCO₃ and forms calcium chloride affecting the hardness of cement, HCl gas emission

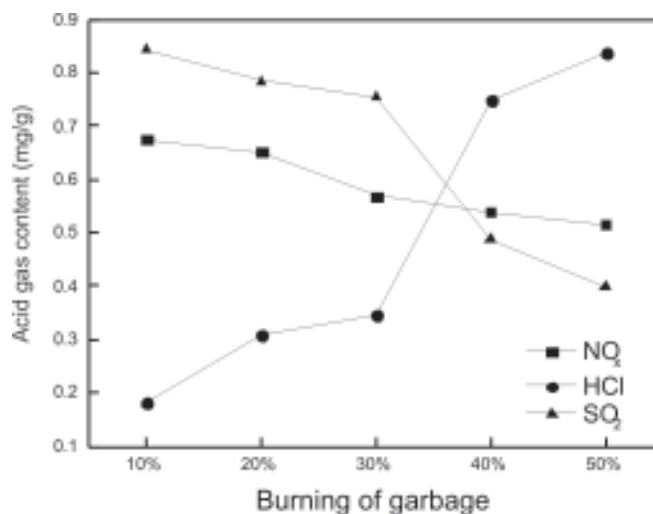


Fig.2 Acid gas emission conditions for waste-fuel blend combustion with different proportions

TABLE 5 ACIDIC GAS EMISSION REDUCTION IN FUEL GASES DURING MIXED FUEL COMBUSTION (%)

proportion of domestic waste in mixed fuel (%)	10	20	30	40	50
SO ₂ emission reduction	8.06	14.52	17.74	46.77	56.41
NO _x emission reduction	3.98	7.39	19.18	23.44	26.56
HCl emission increase	42.19	142.18	170.31	483.59	555.47

has been strictly controlled in the new flue gas emission standards in cement plant issued by the State, so we can see from the experimental results that the mixing proportion of domestic waste cannot be higher.

2.3.2 Different proportions of mixed fuel combustion

Take 1g mixed fuel (garbage proportion is 10%, 20%, 30%, 40%, 50%) respectively for combustion, determine SO₂, NO_x and HCl gas content in the flue gas and get the relationship diagram between proportion of domestic waste and acidic gas emissions, as is shown in Fig.2.

Fig.2 shows that with the gradually increase of mixing proportion of domestic waste in the mixed fuel from 10% to 50%, SO₂, NO_x contents in combustion flue gas decrease, that is, SO₂ decreases from 0.844mg/g to 0.400mg/g while NO_x decreases from 0.676mg/g to 0.517mg/g. when mixing proportion of domestic waste is greater than 30%, SO₂ emissions in fuel gases declines sharply and SO₂ gas emission reduction rises rapidly, which indicates that mixing domestic waste can effectively reduce SO₂, NO_x gas emissions in flue gases and reduction effect gets better as the increasing proportion of domestic waste. The main reason is sulfur content in coal is higher than that in the domestic waste, and the domestic waste itself contains a certain amount of humic acid and lignin with strong SO₂ adsorption capacity which can largely reduce SO₂ precipitation speed, meanwhile the domestic waste contains a number of alkali metals (Ca, K, Na, etc.) and alkaline earth metal (mainly Ca) which will react with SO₂ and forms sulfate or sulfite [18], therefore, the higher mixing proportion of domestic waste, the lower the SO₂ emissions; Secondly, the nitrogen content in coal is higher than that of domestic waste, and the conversion rate of NO_x is higher than that of domestic waste during combustion; It will be volatilize at low temperatures and snatch O₂ with coal ash for combustion so that local coal combustion zone becomes oxygen-depleted zone and restrain the generation of NO_x during coal combustion; At the same time, reducing gas released during combustion of domestic waste will be reduce NO_x to N₂. Thus, mixing coal with domestic waste is good for the reduction of NO_x, and reduction effect gets better with the increase of mixing proportion of domestic waste.

Meanwhile, HCl content in the combustion flue gas is gradually increasing from 0.182mg/g to 0.839mg/g. And when mixing proportion of domestic waste is greater than 30%, HCl emissions increases dramatically, mainly due to high content of chlorine contained in plastic, rubber and other ingredients

of domestic waste, which results chlorine of domestic waste is much higher than coal and a lot of HCl gas is generated by chlorine element during combustion; Secondly, during coal combustion, more iron, aluminum oxide will be generated, when co-combustion with domestic waste, these metal oxides react with HCl gas released during combustion and generate chloride salt, which can reduce the HCl gas emissions to some extent. Therefore, with the increase proportion of domestic waste, HCl gas is increasing gradually.

Analysis of acidic gas emission reduction in fuel gases during mixed fuel and coal alone combustion and the results are shown in Table 5.

Table 5 shows that, compared with coal combustion alone and with the gradually increase of mixing proportion of domestic waste in the mixed fuel from 10% to 50%, SO₂ emission reductions increase from 8.06% to 56.41%, especially when the mixing proportion of domestic waste is greater than 30%, SO₂ emission reductions in fuel gases increase from 17.74% to 46.77% rapidly, SO₂ emission reduction increases sharply; NO_x emission reductions increase from 3.98% to 26.56%. When the mixing proportion of domestic waste is greater than 30%, emission reductions increase from 7.39% to 19.18%, NO_x emission reduces obviously; HCl emission increase increases from 42.19% to 555.47%; when the mixing proportion of domestic waste is greater than 30%, HCl emission increase dramatically and its increase increases from 170.31% to 483.59%.

From the experimental results, we can see that when the mixing proportion of domestic waste is gradually increasing, NO_x and SO₂ gas reduction effect is gradually increasing which is in favor of the reduction of industrial NO_x and SO₂ gas emissions, but HCl gas emissions is gradually increasing, especially when the mixing proportion of domestic waste is greater than 30%, HCl gas emission increase has a sharp increase. HCl gas is very harmful to the human body, and its emissions cannot be too high according to the limit values of HCl emission in combustion flue gases issued by the state.

3.4 EFFECT OF COMBUSTION CONDITIONS ON THE ACIDIC GAS EMISSIONS OF MIXED FUEL

3.4.1 Effect of combustion temperature

Set different combustion temperatures (700, 800, 900, 1000°C), burn the mixed fuel with 30% proportion of domestic waste and determine of the acidic gas content of the flue gas. The results are shown in Fig.3.

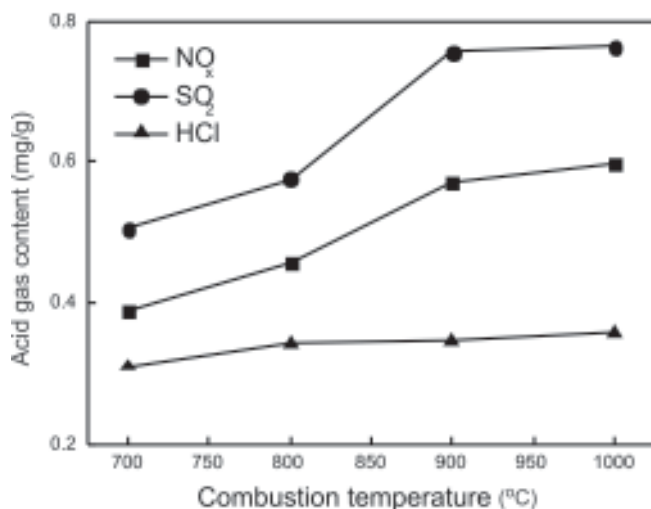


Fig.3 Acid gas emissions under different combustion temperatures

Figure 3 shows that, as the combustion temperature rises from 700°C to 1000°C, NO_x, SO₂ gas content of fuel gases released during the combustion of mixed fuel shows an increasing trend; When the temperature rises from 800 to 900°C, NO_x, SO₂ emissions are of significant rise. This is mainly because with the increase of the combustion temperature, nitrogen-containing organic and sulfur-containing organic in the mixed fuel combustion gases burn very fast and NH₃, H₂S and other reducing gases in volatile components of combustion gas is accelerated with increasing temperature and reduces the emission of reducing volatile gas; Secondly, with the increase of combustion temperature, nitrate and sulfate in mixed fuel will gradually decompose NO_x and SO₂, and the sulfur, nitrogen mineral in the fuel will be gradually burning as the increase of the combustion temperature; but when the temperature exceeds 900°C, NO_x and SO₂ gas emissions does not increase substantially, mainly because when the temperature exceeds 900°C, the combustion temperature is already high and it has no effect on whether the volatile component burns completely or not and nitrate and decomposition of nitrate and sulfate or combustion of mineral containing sulfur and nitrogen.

HCl gas emissions are essentially the same, the main reason is that the HCl generates mainly from the combustion of organic in fuel, for example, the combustion of chlorinated organic substances in plastic and cloth generates HCl gas, when the temperature exceeds 700°C, these chlorinated organic are able to burn completely. So as the rise of temperature, HCl gas emissions remain basically unchanged.

3.4.2 Effect of air flow rate

We analyze the acid gas emissions of fuel gases released during combustion of mixed fuel composed by 30% domestic waste and coal under different air flow rate (0.1, 0.2, 0.3L/min) and obtain NO_x, SO₂ and HCl gas emissions under their different combustion temperatures. Detail information is shown in Fig.4.

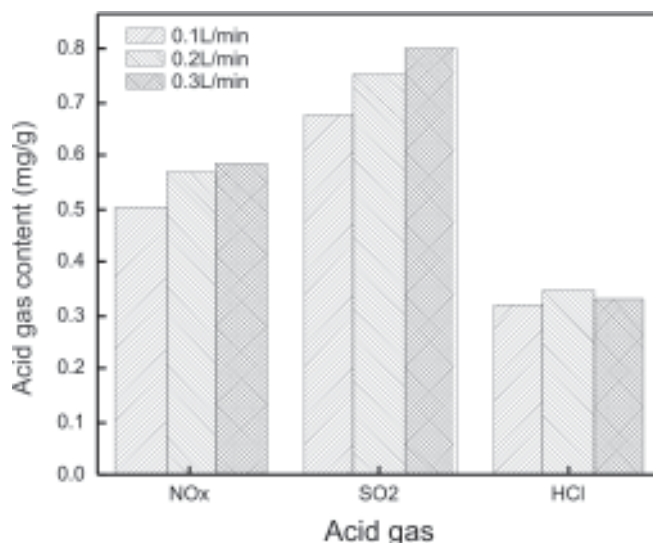


Fig.4 Polluting gas emission conditions under different combustion temperatures

Fig.4 shows that with the increase of air flow rate, NO_x and SO₂ gas emissions in combustion fuel gases of mixed fuel show an increasing trend. In air flow rates increasing from 0.1L/min to 0.3L/min, NO_x emissions increases from 0.502mg/g to 0.587mg/g and SO₂ emissions increases from 0.678mg/g to 0.803mg/g. The main reason is when the air flow rate increases, N, S gas contained in NH₃, H₂S etc. of volatile components will be burnt gradually and generate NO_x and SO₂. Therefore, with the increase of air flow rate, NO_x, SO₂ gas emissions will be increased at the same time.

HCl gas emissions in combustion fuel gases of mixed fuel are almost the same. After flow rate increasing from 0.1L/min to 0.3L/min, HCl gas emissions keeps at 0.33mg/g or so. The reason is that the generation of HCl does not need the participation of O₂ and is mainly from the decomposition of chlorinated organic substances of fuel. Therefore, when the air flow rate is changing, an HCl gas emission remains unchanged.

4. Conclusion

(1) When the garbage content is less than 60%, it can be used as fuel substitute materials of cement. SiO₂, CaO, Al₂O₃, Fe₂O₃ content of mixed fuel ash are more than 10%, the total content of four kinds of oxide is between 75% and 85%, alkali content (0.658K₂O+NaO) is between 2.71% and 6.22%, and five heavy metals (Pb, Ni, Cd, As, Hg) contents are all less than that generated during the combustion of coal alone.

(2) Mixing domestic waste at mixed fuel can reduce NO_x and SO₂ emissions of combustion gases to a certain extent, and HCl gas emissions is increasing with increasing proportion of domestic waste. When garbage proportion reaches 30%, acidic gases emissions generated during combustion of mixed fuel is optimal.

(3) NO_x and SO_2 gas emissions of fuel gases have close relationship with combustion temperature and air flow rate during the combustion of mixed fuel, which have little effect on HCl gas emissions.

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