Effects of Municipal Solid Waste Torrefaction and Classification Pretreatment on Hydrogen Chloride Release

Guangtao Hu1, 2, *, Xing Ning2, Borui Zhu1, Shuang Wang1

1 School of Energy Engineering, Yulin University, Yulin, China
2 School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an, China

Email address:
ylhuguangtao@126.com (Guangtao Hu)
*Corresponding author

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Abstract: The incineration treatment of municipal solid waste is widely used, but gas-phase hydrogen chloride (HCl) generated during the incineration seriously threatens the heating surface of boiler tail, and even causes serious harm to environment. Through incineration experiments, this paper found that many factors (temperature, oxygen concentration, residence time and moisture content) affect the HCl emission characteristics in the process of municipal solid waste incineration treatment. In order to effectively cut back the HCl emission in incineration treatment of municipal solid waste, this paper proposes torrefaction and classification pretreatment of municipal solid waste. Torrefaction pretreatment has a significant inhibitory effect on HCl emission during the incineration of mixed municipal solid waste. At the same time, compared with the incineration treatment of mixed municipal solid waste, sawdust and plastic municipal solid waste of the same quality after classification pretreatment can effectively curb HCl emissions within the entire temperature range. In particular, HCl emissions are reduced by 35% during sawdust incineration treatment at 400°C. Furthermore, by studying the reaction mechanics of HCl generation during combustion at different temperatures, and the reaction rate of HCl generation is: mixed municipal solid waste > plastic > sawdust. Therefore, this paper believes that torrefaction and classification pretreatment of mixed municipal solid waste is beneficial to inhibit the precipitation of chlorine during the incineration process and promotes the resource utilization of municipal solid waste, which is of significant significance for energy conservation and emission reduction.

Keywords: Municipal Solid Waste (MSW), Torrefaction, Classification, HCl

1. Introduction

With the rapid development of our country economy, the output and growth rate of municipal solid waste ranks first in the world and grows at a rate of 8%-10% per year [1, 2]. In order to avoid pollution to environment, it has become urgent to solve the rational treatment of municipal solid waste in the process of industrialization. Nowadays, municipal solid waste landfills, composting and incineration are the main treatments. Compared with the long time period of landfill and compost, however, incineration has advantages of reducing energy consumption, fast treatment speed and high temperature sterilization, and has become an important way to deal with municipal solid waste [3-6]. According to statistics, China's municipal solid waste production was 215,509 kilotons in 2017, a year-on-year increase of 5.7% [5, 6]. Therefore, study of renewable cycle of municipal solid waste on the atmospheric environment management, energy conservation and emission reduction has far-reaching and long-term significance [7].

Because of the change of time and space, complex composition of municipal solid waste is complicated, diverse and uneven. Municipal solid waste mainly includes high chlorine waste plastic and medium-content chlorine timber, which mostly exist in the form of organic chlorine, but also includes a small number of inorganic chloride [8, 9]. Yang Z-C has found that due to the complexity of solid waste composition the distribution of chlorine varies, but the source of chlorine is consistent [10, 11]. Thus, he has considered that it provides a theoretical basis for simulating a solid waste chlorine mixture. A large number of scholars have also shown
that municipal solid waste in the middle-low temperature of 200–600°C commonly in the form of HCl release, and in high temperature mainly in the form of alkali metal chloride precipitation [12-14]. In the case of full combustion, chlorine in poly-vinyl chloride (PVC) substances can be converted into HCl. When the presence of moisture, the increase of moisture in municipal solid waste can provide more H free radicals under high temperature combustion conditions, so that C free radicals have more opportunities to effectively collide with H free radicals [15]. The release has little effect at low temperature, but it aggravates the release of HCl during combustion at medium-high temperature. At the same time, mixed burning of municipal solid waste will show a higher release of HCl pollutants gas, not only will corrode the metal pipe wall, shorten the service life of the equipment, at the same time, in the presence of carbon sources will produce dioxin precursors, which seriously harm to life and ecological environment [21, 22].

Thence, this paper analyzed that the difference in HCl release characteristics of municipal solid waste wet mixture and dry mixture (plastic and sawdust) in an orderly manner, thereby revealing influence of municipal solid waste torrefaction on the HCl release characteristics. Effects of multi-factors (combustion temperature, combustion oxygen concentration, combustion reaction time) were studied on the HCl release characteristics of municipal solid waste torrefaction mixture, and reaction kinetics of HCl release was further proposed during combustion of municipal solid waste torrefaction mixture. After that, municipal solid waste is classified and then burned (plastic, sawdust) to analyze the HCl release characteristics. Finally, comparative research of influence of municipal solid waste torrefaction and classification on waste incineration pollutant emission based on the perspective of HCl release, which is aimed at providing a theoretical basis for municipal solid waste torrefaction and classification to reduce pollutant emission.

2. Experimental System and Condition

Take 2–3 mm municipal solid waste wet mixture (plastic and sawdust) and the classified wet waste (plastic, sawdust) sample particles for torrefaction in a drying oven at 105°C for 12 h, and obtained a mixture of torrefaction and classified municipal solid waste. After connecting the system devices and checking the air tightness, the combustion experiment of each working condition is carried out in the two-dimensional quartz reaction tube. Simultaneously, the NaOH solution is used to collect the precipitation of HCl at the other end, and the measurement method is silver nitrate capacity method.

The silver nitrate capacity method can be frequently utilized to determine the content of chloride ions in the absorption liquid: first, the sample solution after sampling is transferred into a white porcelain dish and then the phenolphthalein indicator is added, and then the potassium chromate indicator is added when its solution red is just faded, finally, titrate 0.01 mol/L silver nitrate standard solution under constant stirring until a non-disappearing light brick red color is produced. Another capacity absorption solution will be used for blank titration. The HCl concentration is calculated as follows:

\[ HCl(mg) = \left( V - V_0 \right) \times C_{AgNO_3} \times 36.45 \times 1000 \]

\( V \)—the volume of silver nitrate standard solution consumed by the titration sample solution (ml);
\( V_0 \)—the volume of the silver nitrate standard solution consumed by the blank solution (ml);
\( C_{(AgNO_3)} \)—concentration of silver nitrate standard solution (mol / L);
36.45—The mass of HCl equivalent to 1 L of 1 mol/L silver nitrate standard solution (g).

3. Results and Discussion

3.1. HCl Release Characteristic of Municipal Solid Waste

3.1.1. Influence of Moisture Content on HCl Release Under Different Temperatures

After municipal solid waste mixture is burned in an air atmosphere with a reaction time of 20 min, Cl-HCl conversion rate continues to increase within the temperature range of 200-700°C. It is especially obvious that Cl-HCl conversion rate of municipal solid waste is sharp at 300-400°C, but the rise tends to be relatively flat at 400-600°C, which is shown in Figure 1. Most of the chlorine is released in the form of HCl during combustion of municipal solid waste. Meanwhile, in two temperature ranges, the former is release of organic chlorine, and the latter is precipitation of inorganic chlorine [14, 23, 24]. It can be seen from the figure that combustion temperature affects the conversion of Cl-HCl, and the conversion rate of Cl-HCl increases significantly with the increase of combustion temperature during combustion process. This is mainly due to the significant increase in the number of activated molecules in temperature, which increases the effective number of collisions between H free
radical and Cl free radical within the residence time, and the chemical reaction is relatively sufficient, so the conversion rate of Cl-HCl is significantly increased [5, 16, 25]. In the process of municipal solid waste combustion, HCl has begun to be precipitated at 200°C, and the conversion rate of Cl-HCl is stable at 600°C, indicating that majority of chlorine in municipal solid waste is precipitated at 600°C.

What’s more, the presence of moisture in combustion process of municipal solid waste remarkably accelerates release of chlorine in the waste, which indicates that part of chlorine release reaction occurs with the participation of moisture vapor. When moisture exists, effect of chlorine precipitation from municipal solid waste combustion is that combustion temperature is meaningfully reduced with the same chlorine precipitation rate. When the presence of moisture, sawdust waste that is not free of volatile chlorine released HCl gas during the incineration process after the inorganic chloride salt is obtained by transfer [15]. because of the existence of kitchen waste with high moisture content from municipal solid waste, the amount of HCl released during combustion of municipal solid waste increases. Therefore, reducing the moisture content of kitchen waste through torrefaction is a technical measure to decrease precipitation of chlorine during the combustion of municipal solid waste.

3.1.2. Influence of Burning Time on HCl Release Under Same Temperatures

Figure 2. Cl-HCl conversion rate of municipal solid waste mixture with different reaction time when burned at 300°C.

Municipal solid waste mixture is burned for different time under 300°C in an air atmosphere, and its Cl-HCl conversion rate is related to the duration of stay in the furnace. As can be seen from Figure 2, as Cl-HCl conversion rate increases rapidly with the increase of the length of stay, and the longer the stay, the fuller the reaction. Furthermore, the process of HCl release of municipal solid waste mixture in combustion process is obviously divided into two stages. From beginning of experiment until 15 min, most of chlorine precipitated by municipal solid waste mixture after heating process is active organic chlorine. At the same time, the precipitation rate at this stage is very fast, and most of municipal solid waste mixture of chlorine has been analyzed by 15 min. But HCl precipitated after 15 min may be precipitation of chlorine in the form of chloride ions, which is relatively slow. This is mainly due to the highest chlorine content of plastic waste, meanwhile, the de-chlorination and combustion process of PVC will occur in the temperature range of 220-320°C, resulting in a large number of active organic chlorine production [26]. But, inorganic chlorine is relatively stable, its chlorine analysis is slow. Therefore, HCl release rate during combustion process of municipal solid waste mixture 15 minutes ago is relatively large, and HCl release is more. In practical engineering applications, municipal solid wastes mixture, especially the residence time of waste plastic in high temperature areas, should be reduced as much as possible, which can significantly slow down generation of HCl, thereby cutting down high temperature corrosion of HCl on the boiler over-heater.

3.1.3. Influence of Oxygen Content on HCl Release Under Same Temperatures

Figure 3. Cl-HCl conversion rate of municipal solid waste mixture with different reaction time when burned at 300°C.

There are oxidizing and reducing alternating atmospheres during combustion process of municipal solid waste mixtures, and different atmospheres have a serious impact on the precipitation of chlorine. In order to study the effect of alternating atmosphere on precipitation of chlorine during combustion process of municipal solid waste mixtures, Cl-HCl conversion rate of different oxygen concentrations was analyzed at a combustion temperature of 300°C and a reaction time of 20 min, the results shown in Figure 3. In the pyrolysis reaction where the oxygen concentration is zero, the Cl-HCl conversion rate of the municipal solid waste mixture is about 2%, indicating that a certain amount of chlorine precipitation exists at 300°C, which is typically due to part of organic chlorine in municipal solid waste mixture has chlorine precipitation during pyrolysis process. With the increase of oxygen, chlorine emission rate increased to about 30%, and
chloride generation changed greatly. As the transformation of atmosphere from weak reductive atmosphere to strong oxidation atmosphere, chlorine precipitation rate increased significantly. Oxygen provides more active particles and sites for the combustion process of municipal solid waste mixtures, so the increase of oxygen concentration effectively increased combustion intensity of municipal solid waste mixture, and brought more HCl release. Therefore, oxygen plays a vital role in the thermal conversion of municipal solid waste mixture chlorine. At the same time, it can be seen that the larger the amount of oxygen, the more conducive to precipitation of HCl, and in order to effectively inhibit the precipitation of HCl, the amount of oxygen cannot be too high.

3.1.4. Reaction Kinetics of HCl Formation During the Combustion of Municipal Solid Waste Mixture

Suppose that municipal solid waste mixture is burned in a tube furnace, and when air passes through a two-dimensional quartz reaction tube at a constant flow rate, the chlorine in municipal solid waste mixture is basically precipitated in the form of HCl during chlorine generation reaction. The relationship between burning time and the conversion rate of Cl-HCl in municipal solid waste mixture at different temperatures is shown in Figure 4. The reaction kinetic rate at different temperatures can also be calculated using the Arrhenius equation:

\[ k = A e^{-\frac{E}{RT}} \]

Molar gas constant: \( R = 8.134 \text{ KJ/mol} \)

Reaction kinetic equation of HCl generated during combustion process of municipal solid waste mixture is shown in Figure 5. \( (k = 1.5092e^{-\frac{10470.878}{RT}}) \)

3.2. HCl Release Characteristics of Sawdust and Plastic

3.2.1. Influence of Moisture Content on the Release of HCl

Earlier in this paper, release of HCl in combustion process of municipal solid waste mixture at different temperatures and moisture contents was studied, and it was pointed out that the presence of moisture vapor aggravates the chlorine extraction in combustion process of municipal solid waste mixture. Nowadays, release of HCl in combustion process of the classified municipal solid waste (sawdust, plastic) at 500°C in different moisture vapor content is studied separately, results is shown in Figure 6. With the increase of moisture vapor content, the change of sawdust is particularly prominent compared with the change of Cl-HCl conversion rate of plastic. When moisture vapor content increased from 0 to 30%, the Cl-HCl conversion rate of sawdust increased by 30.2%. Furthermore, with the increase of moisture vapor content, the Cl-HCl conversion rate of sawdust tends to level off. However, for the Cl-HCl conversion rate of plastics, increasing in moisture vapor content does not change much, and its fluctuations range is even smaller than that of sawdust. Release of most of the organic chlorine in PVC in the high-chlorine plastic basically ends at 500°C, and its inorganic chlorine is relatively small, so the increase in moisture vapor

Figure 4. The relationship between burning time and the conversion rate of Cl-HCl in municipal solid waste mixture at different temperatures.

Figure 5. Reaction kinetic equation of HCl generated during combustion process of municipal solid waste mixture.

Figure 6. Cl-HCl conversion rate of municipal solid waste (sawdust, plastic) with different moisture contents when burned at 500°C.
content has little effect on it. However, it is different for medium-chlorine sawdust. With the increase of moisture vapor content, under the hydrolysis of chlorinated minerals, sawdust that non-volatile chlorine-free sawdust by transfer to obtain inorganic chlorine salt, and the solid chlorine equivalent into gaseous chlorine in the incineration process to analyze HCl.

3.2.2. Influence of Combustion Temperature on the Release of HCl

The Cl-HCl conversion rate of the classified municipal solid waste at different combustion temperatures is shown in Figure 7. After sawdust combustion in the air atmosphere of 20 min, the Cl-HCl conversion rate is continuously improved in the temperature range of 200 to 600°C, especially the Cl-HCl conversion rate has reached 50% at about 430, and the Cl-HCl conversion rate in the temperature range of 600 to 800°C is flat. The large proportion of the chlorine source of medium-chlorinated sawdust that precipitates in the form of inorganic chlorine at medium-low temperatures of 200-600°C. However, for plastic, the Cl-HCl conversion rate rises sharply in the temperature range of 300-400°C, and the Cl-HCl conversion rate is as high as 74% at 400°C, and the Cl-HCl conversion rate has flattened in the temperature range of 500 to 800°C. This is mainly due to the fact that the precipitation of most organic chlorine in high-chlorine plastics, and the proportion of inorganic chlorine is relatively low. At the same time, through the slope, it can be found that the order of Cl-HCl conversion rate in the temperature range of 200-600°C is: plastic>sawdust. The temperature range of HCl released by inorganic chlorine in sawdust during combustion process is usually 200-600°C, while the temperature range of HCl released by organic chlorine in plastics during combustion is relatively narrow but the conversion is faster. Therefore, the combustion temperature has a certain effect on the HCl gas phase migration transformation of plastic and sawdust with different chlorine content. As the temperature increases, so does the conversion rate of all chlorine sources Cl-HCl, but the organic chlorine Cl-HCl conversion rate is higher at the same temperature. Thus, the classification of municipal solid waste is beneficial to define the balance point of thermal energy conversion and reduction of HCl release, thereby further promoting the effective utilization of municipal solid waste resources.

3.2.3. Influence of Burning Time on the Release of HCl

Figure 8 shows classification of municipal solid waste at a different reaction time Cl-HCl conversion rate at 300°C in an air atmosphere. The conversion rate of chlorine in sawdust is higher than that of plastic, and the conversion rate of Cl-HCl in 5 min is about 12%. The chlorinated sawdust can precipitate HCl in a short time when temperature rises rapidly. Meanwhile, for plastic, the HCl release law is roughly the same as that of sawdust when incinerated. However, it is worth noting that the Cl-HCl conversion rate of high-chlorine plastics is relatively low in 300°C air atmosphere after the classification of municipal solid waste. The precipitation of gaseous chloride is related to combustion temperature and residence time. It can be seen from the figure that at the same combustion temperature, with the increase of residence time, both the release amount and conversion rate of combustible solid waste HCl show an increasing trend. Furthermore, regardless of whether it is high-chlorine plastic or medium-chlorine sawdust, the Cl-HCl conversion rate shows a two-stage change as the combustion time increases, and the Cl-HCl conversion rate increases sharply within 5-15 min. But the Cl-HCl conversion rate is relatively slow within 5-25 min. The release of HCl from the organic chlorine during the thermal conversion process, which is mainly in the early stage of the reaction, and its release is relatively concentrated, while the precipitation of chlorine becomes slow with the increase of time, and the precipitation of inorganic chlorine is mainly at this time. Organic and inorganic chlorine combustible solid wastes release HCl at a different rate and concentration time period, which illustrates the difference in the internal mechanism of HCl release, and has certain guiding significance for the classification treatment of...
municipal solid wastes.

3.2.4. Influence of Oxygen Concentration on the Release of HCl

![Figure 9. Cl-HCl conversion rate of municipal solid waste (sawdust, plastic) with different oxygen concentrations.](image)

Sawdust and plastic have oxidizing and reducing atmosphere changes during the combustion process, in view of the municipal solid wastes high volatility and combustion characteristics, sawdust and plastic in combustion process of change of oxidation and reduction atmosphere is more obvious, therefore, it is very important to study the effect of atmosphere on chlorine analysis. Figure 9 shows the Cl-HCl conversion rate of different oxygen concentrations of each material at a combustion temperature of 300°C at 20 min after the classification of municipal solid waste. The larger oxygen concentration is beneficial to precipitation of chlorine in sawdust and plastic during combustion. When oxygen concentration is zero, that is pyrolysis process, the conversion rates of Cl-HCl for sawdust and plastic are 2.2% and 1.9%, respectively. At the same time, the Cl-HCl conversion rate of sawdust is significantly higher than that of plastics in the entire oxygen concentration range. When oxygen concentration is low, furnace presents a reducing atmosphere and chlorine precipitation rate is low. But, when the amount of oxygen increases, the conversion rate of Cl-HCl increases, which shows that combustion of material is more beneficial to promote the conversion of Cl-HCl, and increasing in the amount of oxidation is beneficial to the conversion of chlorine. However, within the range of 30-40% oxygen content, the increase in the Cl-HCl conversion rate of each material tends to be flat, mainly because the decisive factor affecting the Cl-HCl conversion in inorganic chlorine is temperature. Therefore, addition of oxygen in the thermal conversion process of combustible solid waste accelerates the decomposition of materials. However, it can also be seen from the figure that changes in oxygen concentration have different promotion effects on HCl during the thermal conversion of inorganic and organic chlorine combustible solid wastes.

3.2.5. Reaction Kinetics of HCl Formation During the Combustion of Sawdust and Plastic

Suppose that sawdust and plastic are burned in a tube furnace, and when air passes through a two-dimensional quartz reaction tube at a constant flow rate, the chlorine in the sawdust and plastic basically precipitates in the form of HCl during the process of generating chlorine. Figure 10 show the relationship between the burning time and the Cl-HCl conversion rate in sawdust and plastic at different temperatures. The Arrhenius equation can also be used to calculate the reaction kinetic rate of HCl generated in the combustion process:

Arrhenius equation:

$$k = A e^{\frac{-E}{RT}}$$

Molar gas constant: $R=8.134$ KJ/mol

The reaction kinetic equation of HCl generated during the combustion of sawdust and plastic are shown in Figure 11.

(sawdust: $k = 0.2306e^{\frac{548.554}{RT}}$, plastic: $k = 0.9009e^{\frac{874.107}{RT}}$)

![Figure 10. The relationship between the burning time and the Cl-HCl conversion rate in sawdust and plastic at different temperatures.](image)
3.3. Effect of Torrefaction and Classification on the Release of HCl from Municipal Solid Waste

The inhibitory effect of torrefaction and classification on the release of chlorine during combustion of municipal solid waste is shown in Figure 12, Figure 13 and Figure 14.

Figure 12 shows the amount of chlorine precipitation before and after the torrefaction of mixed, sawdust and plastic municipal solid waste with 50% moisture content at 500°C. Compared with the original municipal solid waste, release of HCl after torrefaction municipal solid waste in the combustion process is reduced, and the total release is also greatly decreased, indicating that torrefaction as a pretreatment means can indeed curb the release of municipal solid waste in the furnace combustion process HCl, thereby preventing the risk of chlorine chloride from municipal solid waste to the boiler heat surface corrosion. Some fuels disappear in the gas phase in the form of volatile hydrocarbons during the torrefaction pretreatment, which can release part of the chlorine in the form of CH$_3$Cl [27]. Simultaneously, CH$_3$Cl competes with HCl for chlorine freelance produced during combustion in urban solid waste, which leads to the chlorine content of the municipal solid waste after torrefaction pretreatment is lower than chlorine content of the original municipal solid waste, and a significantly lower HCl resulting from HCl from combustion of primary municipal solid waste [21, 28]. Furthermore, it can be seen from Figure 12 that the reduction of chlorine precipitation during combustion process after the torrefaction pretreatment of plastic solid waste is less reduced,
but the reduction of chlorine precipitation is the largest after the torrefaction pretreatment of sawdust solid waste, while mixed municipal solid waste is centered. This is mainly because part of the chlorine in sawdust solid waste torrefaction pretreatment is fixed by alkali metals, alkaline earth metals and their oxides in the ash after the sawdust is burned, therefore, its inhibitory effect is the most obvious [29]. The torrefaction of municipal solid waste has an important inhibitory effect on precipitation of chlorine and effectively improves the incineration treatment.

Figure 13 shows the difference between the chlorine precipitation content of sawdust and plastic municipal solid waste after classification and the chlorine precipitation content of unclassified mixed municipal solid waste during the combustion process at different temperatures. Figure 14 shows the comparison of the chlorine precipitation reaction kinetics of sawdust and plastic municipal solid waste after classification under the same quality with that of unsorted mixed municipal solid waste.

A large number of previous studies have shown that chlorine sources of municipal solid waste mainly derived from plastic with high chlorine content and sawdust with medium chlorine content [30, 31]. However, HCl release of municipal solid waste after classification was revealed in this paper. As can be seen from Figure 13, HCl release of sawdust after classification at 400°C has the largest difference with HCl release of mixed municipal solid waste under the same mass, and classification has the most prominent inhibitory effect on its HCl release. At the same time, compared with classified plastic, the amount of HCl released during the combustion of sawdust after classification in the full temperature range is also significantly reduced. However, in general, HCl emission of municipal solid wastes of the same quality after classification decreased. In fact, any change in the composition of municipal solid waste will significantly affect the combustion and further change the amount of HCl released. The classified municipal solid waste has a single chlorine source structure, stable incineration, and the coordination of organic chlorine and inorganic chlorine is hindered, thereby effectively reducing the release of HCl [32, 33]. Further known from Figure 14, reaction kinetics relationship generated by HCl, when the combustion temperature is higher than 300°C, the HCl generation of mixed municipal solid waste is significantly increased, and then at the same temperature the HCl growth of the size of the relationship is: mixed >plastic> sawdust. Therefore, the classified combustion treatment of municipal solid waste is beneficial to curb the release of HCl, thereby further reducing the risk of corrosion on the heating surface of the boiler, and it is of great significance to suppress pollutant emissions.

4. Conclusions

Mixed municipal solid waste could release more HCl during combustion treatment. When municipal solid waste contains moisture, it will further increase the precipitation of HCl, and torrefaction can effectively reduce the precipitation of chlorine, especially for inorganic chlorine at high temperatures. At the same time, it was found that the amount of HCl released from sawdust and plastic was significantly decreased during the combustion process when municipal solid waste was classifying, and the classification inhibited the release of HCl. Therefore, the orderly adjustment of combustion conditions, the introduction of torrefaction and classified pretreatment can effectively lower the amount of HCl released during the incineration of municipal solid waste, thereby effectively reducing the risk of corrosion on the heating surface of the boiler, which further promotes energy conservation and emission reduction.

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Conflicts of Interest

All the authors do not have any possible conflicts of interest.

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