



Frequency comparative study of coal-fired fly ash acoustic agglomeration

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Received 18 December 2010; revised 20 May 2011; accepted 30 May 2011

Abstract

Particulate pollution is main kind of atmospheric pollution. The fine particles are seriously harmful to human health and environment. Acoustic agglomeration is considered as a promising pretreatment technology for fine particle agglomeration. The mechanisms of acoustic agglomeration are very complex and the agglomeration efficiency is affected by many factors. The most important and controversial factor is frequency. Comparative studies between high-frequency and low-frequency sound source to agglomerate coal-fired fly ash were carried out to investigate the influence of frequency on agglomeration efficiency. Acoustic agglomeration theoretical analysis, experimental particle size distributions (PSDs) and orthogonal design were examined. The results showed that the 20 kHz high-frequency sound source was not suitable to agglomerate coal-fired fly ash. Only within the size ranging from 0.2 to 0.25 μm the particles agglomerated to adhere together, and the agglomerated particles were smaller than 2.5 μm . The application of low-frequency (1000–1800 Hz) sound source was proved as an advisable pretreatment with the highest agglomeration efficiency of 75.3%, and all the number concentrations within the measuring range decreased. Orthogonal design L16 (4)³ was introduced to determine the optimum frequency and optimize acoustic agglomeration condition. According to the results of orthogonal analysis, frequency was the dominant factor of coal-fired fly ash acoustic agglomeration and the optimum frequency was 1400 Hz.

Key words: coal-fired fly ash; acoustic agglomeration; frequency; agglomeration kernel; orthogonal design

DOI: 10.1016/S1001-0742(10)60652-3

Citation: Liu J Z, Wang J, Zhang G X, Zhou J H, Cen K F, 2011. Frequency comparative study of coal-fired fly ash acoustic agglomeration. *Journal of Environmental Sciences*, 23(11): 1845–1851

Introduction

Air pollution has become increasingly concerns as the economic development (Bi et al., 2007; Fenger, 2009). Particulate pollution as a main type of atmospheric pollution is seriously harmful to human health and environment (Du et al., 2010; Lee et al., 2007; Pope et al., 2002). It has raised a great concern in international community (Sandström et al., 2005; Seaton et al., 1995). Particulate pollution is mainly caused by the burning of fossil fuels (Liu et al., 2006; Zhang et al., 2008), vehicle exhaust emissions (Clarke et al., 1996; Guo et al., 2010; Zhang et al., 2007), industrial processes (Gu et al., 2010) and other human activities (Lewtas, 2007; Li et al., 2009a). Total suspended particulates (TSP) and particulate matter (PM) with the aerodynamic diameter less than 10 μm (PM₁₀) contain many toxic substances, such as polycyclic aromatic hydrocarbons and heavy metals (Li et al., 2009b). They are recognized as a serious public health concern, especially for the smaller particles with aerodynamic diameter less than 2.5 μm (PM_{2.5}). These particles can travel deeper into alveolus even into the capillaries and have been linked to premature deaths, chronic bronchitis and aggravated

asthma (Chow et al., 1994). The current conventional particle filtering devices, such as electrostatic precipitators, cyclone separators, bag houses and wet scrubbers, have higher total dust removal efficiency for the particles larger than 5 μm , but fall short of retaining very fine particulate matter. The pretreatment of flue gas before entering filtering devices is required to enlarge the particle average size to the point where the conventional filter operates. In this way, it can substantially improve the fine particle removal efficiency of the conventional device. Among other innovative approaches for small-particle filtration techniques, acoustic agglomeration has recently evolved as an efficient method for controlling small particle emissions prior to the actual filtration stage (Boulaud et al., 1984; Gallego-Juárez et al., 1999; Reethof, 1986). The application of acoustic waves to the aerosol forces particles to move toward each other and causes collisions between them. Once they collide, the particles are likely to adhere and form larger ones due to the meshing of their irregular surface and the strong surface molecular attraction force. The newly born particles continue to collide with other particles or agglomerates. This cascading process takes place simultaneously among all particles. The acoustic agglomeration results in a significant shift towards larger

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sizes in a short while of the particle size distributions (PSDs).

A lot of theoretical and experimental work has been done on acoustic agglomeration process (Boulaud et al., 1984; Hoffmann et al., 1993; Lee et al., 1982; Liu et al., 2009; Shaw and Tu, 1979; Song et al., 1994; Tiwary and Reethof, 1987; Wang et al., 2011), as the mechanisms peculiar to such process are very complex and their descriptions are not completely established. Orthokinetic interaction is based on the entrainment of different-size particles by the intensive sound wave in polydisperse aerosol (Cheng et al., 1983), and it is widely recognized as a primary acoustic mechanism. Hydrodynamic interactions results from the mutual influence of particles due to the nonlinear interaction of scattered waves (Lee et al., 1982), and it becomes particularly important to the monodisperse aerosol with the similar-size particles wherein orthokinetic interaction nearly vanishes due to the absence of relative oscillatory motion. In the acoustic agglomeration process, the agglomeration efficiency is affected by many factors, such as frequency, sound pressure level (SPL), aerosol initial concentration, resistant time. The agglomeration effect is a result of mutual constraint, mutual influence and coordinated development of these factors.

The most important and controversial factor is frequency. According to the orthokinetic mechanism there is an optimal frequency, f_{opt} . If the frequency is much lower than f_{opt} , the suspended particles are totally vibrated with sound at the same amplitude; if the frequency is much higher than f_{opt} , the particles remain stationary and no agglomeration takes place. Based on the mechanism of hydrodynamic interaction, the forces between the particles can be enlarged as the frequency increases. The influence of frequency has been investigated by many researchers. Cheng et al. (1983) performed an experimental investigations of acoustic agglomeration with the frequencies from 600 to 3000 Hz to deal with NH_4Cl aerosol and the results proved that the agglomeration effect was very sensitive to the frequency and the optimum frequency was 3 kHz. Tiwary (1985) utilized the acoustic agglomeration to deal with coal-fired boiler flue gas and found that the optimum frequency was 2 kHz. Hoffman et al. (1993) combined the 44 Hz low-frequency sound with the limestone with mean diameter of 88 μm as absorbent to agglomerate the coal-fired fly ash and observed a good agglomeration effect. Capéran et al. (1995) used 10 and 21 kHz sound sources to agglomerate the glycol fog aerosol and achieved a better efficiency with the frequency of 21 kHz than that of 10 kHz. Gallego-Juárez et al. (1999) developed an agglomeration chamber driven by four high-power and highly directional acoustic transducers of 10 and/or 20 kHz to deal with the fume generated by a fluidized bed coal combustor, and found that using the 20 kHz sound source could get a better agglomeration effect. Riera-Franco de et al. (2000) utilized the 10 and 20 kHz sound sources to agglomerate the submicron particles in the diesel exhaust and found that using the agglomeration effect of 20 kHz was better than that of 10 kHz. Overall the investigated frequency varied from 44 to 21000 Hz; however the conclusion has not yet

reached a consensus on the value of f_{opt} .

In this article, we utilized the acoustic agglomeration to deal with the coal-fired fly ash, and comparative studies between high-frequency and low-frequency sound source were carried out to investigate the influence of frequency on agglomeration efficiency. Acoustic agglomeration theoretical analyses, experimental particle size distributions (PSDs) and orthogonal design were examined.

1 Experimental setup

The acoustic agglomeration system (Fig. 1) consists of agglomeration chamber, sound source, fly ash and seed particle feeding, aerosol sampling and measurement. The agglomeration chamber is made of a vertical tube with an inside diameter of 99 mm and a length of 1500 mm. The sound source is installed on the top of the chamber. Two sets of sound sources with high-frequency and low-frequency are studied. The high-frequency sound source consists of high-power piezoelectric ceramic transducer (ZJS-2000, Hangzhou Success Ultrasonic Equipment Co., Ltd., China) and ultrasonic control. The ultrasonic emission area is 50 cm^2 and SPL can be up to 150 dB in the atmosphere. The low-frequency sound source is made up of a horn and compression driver (YF-513, Yong Fa Electronics Co., Ltd., China), with the maximum input power of 80 W and the frequency ranging from 180 to 5500 Hz. The compression driver is combined with an amplifier (QSC RMX2450, QSC Audio Products, LLC, USA), which is powered by a signal generator (Goodwill SFG-1013, Good Will Instrument Co., Ltd., Taiwan). A foam rubber is placed at the bottom of the agglomeration chamber to prevent the sound wave reflection to ensure a relatively homogeneous travelling wave field in the chamber. Three acoustic measurement points are set along the chamber wall at regular intervals.

To simulate the testing aerosol similar to the coal-fired flue gas, the coal-fired fly ash particles are collected from an electrostatic precipitator of a coal-fired boiler. These particles are continuously given by the micro feeder, and then they are mixed with clean air-stream in the Venturi mixer. Coarse particles in the initial aerosol are removed by a cyclone with a cut-diameter of 10 μm . Then the aerosol

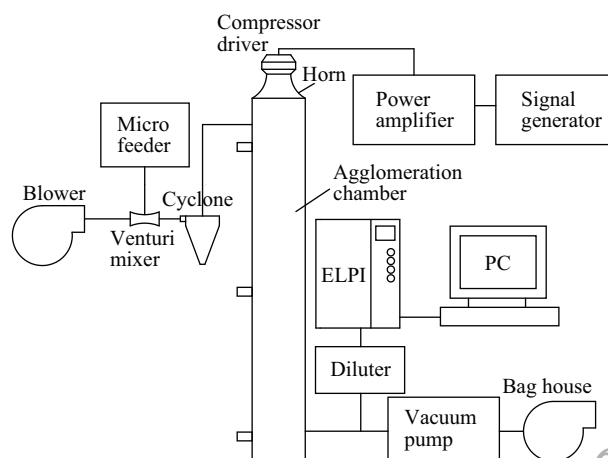


Fig. 1 Schematic diagram of acoustic agglomeration.

enters the agglomeration chamber. Residence time ranging from 3 to 7 sec are controlled by the flow ranging from 6 to 14 m³/hr. Electrical low pressure impactor (ELPI; Dekati type, Finland) is installed at the outlet of the chamber to measure the aerosol real-time PSDs. It measures the particle size distribution in the size range 0.03–10 µm with 12 channels. Before entering the ELPI, the aerosols are diluted by a two-stage diluter (DEKATI DI-1000; Dekati type, Finland) at the ratio of 1/64.

All the experiments are carried out at ambient temperature (25°C), and all the measurements are performed after the system is operated steadily for more than 10 min.

2 Results and discussion

2.1 Theoretical analyses

The interaction of a single spherical particle suspended freely in aerosol through which a plane acoustic wave is propagating leads two interesting results: one is called particle entrainment, which means the particle gains momentum from the wave and attempts to oscillate with the gas medium; the other is called wave scattering, which means the incident wave is partially scattered from the surface of the particle.

The local acoustic velocity u can be calculated by Eq. (1).

$$u = \text{Re}(U_0 e^{-j\omega t}) \quad (1)$$

where, Re means the real quantity of an unreliable figure, U_0 is the velocity amplitude of the gas medium, ω is the angular frequency, t is the time, and j is the imaginary number. Assuming U_0 is much smaller than the sound speed, and the particle diameter, d_p , is much smaller than the acoustic wavelength, the particle oscillating velocity, u_p , can be calculated by Eq. (2) (Song et al., 1994).

$$u_p = \text{Re}(H U_0 e^{-j\omega t}) \quad (2)$$

where, H called the complex entrainment function is defined as the ratio of the complex velocity amplitude of the particle to that of the gas medium. It is a measure of the particle entrainment. The magnitude of H is zero when the particle remains motionless and the magnitude of H is unity when the particle oscillates fully with the medium. The acoustic entrainment factor, μ_p , which is defined as the ratio of the particle amplitude to gas amplitude in the sound wave, can be calculated by Eq. (3) (Temkin, 1994).

$$\mu_p = \frac{u_{p0}}{u_0} = \frac{1}{\sqrt{1 + (\omega\tau_d)^2}} \quad (3)$$

where, τ_d is the particle dynamic relaxation time, given by Eq. (4) (Tiwary and Reethof, 1987).

$$\tau_d = \frac{\rho_p d_p^2}{18\mu} \quad (4)$$

where, ρ_p is the particle density, and μ is the dynamic viscosity of the gas medium. Combining Eq. (1) with

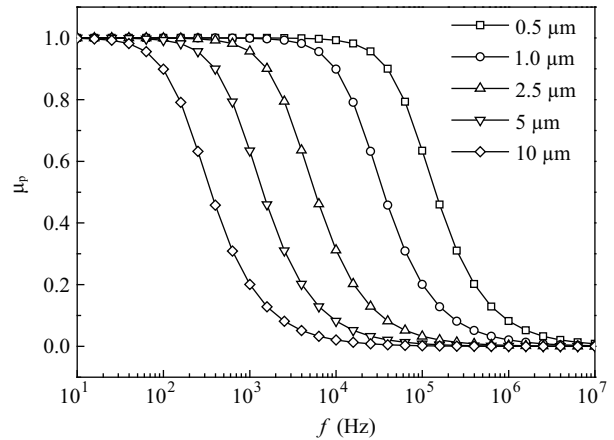


Fig. 2 Influence of frequency (f) on entrainment factor (μ_R) with different particle sizes ranging from 0.05 to 10 µm. u_p is particle oscillating velocity.

Eq. (2) can find that the entrainment factor is related to many factors. While in the acoustic agglomeration the density and the dynamic viscosity of the gas medium are considered as constants, and the particle density is also considered as constant, therefore, the entrainment factor is mainly related to the frequency and particle size.

In this experimental condition, the particle density is 2500 kg/m³, and the gas dynamic viscosity is 1.79×10^{-7} Pa·sec, thus the influences of frequency on entrainment factor with different particle sizes ranging from 0.05 to 10 µm are calculated (Fig. 2).

The entrainment factor becomes lower as sound frequency becomes higher. It means the particle vibration speed is smaller. There are two threshold values f_{\min} and f_{\max} for the frequency influence on entrainment. When the frequency is lower than f_{\min} , the entrainment factor is close to 1 and the particle oscillates fully with the medium. When the frequency is larger than f_{\max} , the entrainment factor is close to 0 and the particle is almost at rest. It also can be found that the frequency thresholds become larger for smaller particle. When the particle size is smaller than 0.05 µm, the entrainment factor is very close to 1. It means no matter how much the frequency is, the fine particle oscillates fully with the medium in the sound field. For the particle with the size ranging from 1 to 2.5 µm, the entrainment factor will decrease rapidly when the frequency is larger than 1000 Hz. If the particle size is larger than 5 µm, the downward trend of entrainment factor is more obvious.

In the aerosol dynamics calculation, agglomeration kernel K is used to describe the collision and agglomeration probability of two particles. It is defined as the number of agglomeration times of two particles, a and b , in the unit time, unit volume and unit particle number concentration, which can be expressed by Eq. (5).

$$K(a, b) = \frac{dN}{n_a n_b dt} \quad (5)$$

where, N is agglomeration times, and n_a and n_b is the number concentrations of particle a and particle b . To simulate the actual agglomeration three assumptions are introduced. First, assuming the particles in the aerosol can

be seen as spheres and also can be divided into two sizes, d_a and d_b ($d_a > d_b$). The larger particle plays a role as the core. The cylindrical area around the core is considered as the agglomeration volume. Second, assuming the particle number concentration in the agglomeration volume is the same as in the gas medium. It ensures that the particles can fill into the agglomeration volume in an instant. Third, assuming all the smaller particles can collide with the core particle and all the collisions lead to the agglomeration. Therefore, the agglomeration volume can be calculated by Eq. (6) (Sheng and Shen, 2006).

$$V_{a,b} = \frac{\pi}{4} (d_a + d_b)^2 \times \frac{u_0}{\omega} \mu_{a,b} \quad (6)$$

And the number of small particles within the agglomeration volume can be calculated by Eq. (7):

$$N_a = V_{a,b} n_a \quad (7)$$

According to the definition of agglomeration kernel K , combining of Eqs. (3), (6) and (7), $K(a, b)$ can be expressed by Eq. (8):

$$K(a, b) = \frac{N_a}{n_a n_b T} = \frac{\pi}{4} u_0 f (d_a + d_b)^2 \frac{|\tau_a - \tau_b|}{\sqrt{(1 + (\omega\tau_a)^2)(1 + (\omega\tau_b)^2)}} \quad (8)$$

Two sets of simulation results of the agglomeration kernels for micro with sub-micro particles are compared under an SPL of 150 dB (Fig. 3).

The peak frequency can be considered as f_{opt} . Figure 3a shows that f_{opt} of sub-micron and micron particles is higher than 10^4 Hz, and f_{opt} becomes higher as the particles become smaller. Compared with Fig. 3b, it can find that f_{opt} of the larger particles is much lower than that of submicron and micron particles. Actually not all the smaller particles near the core particle move straightly until hitting the core particle, the airflow detours the core particles and the trajectories of fine particles will deflect from the straight lines. Collision efficiency ε is introduced, and defined as the proportion of fine particles colliding with core particle (Zhang et al., 2009). The agglomeration kernel K can be amended as K' (Eq. (9)).

$$K'(a, b) = \varepsilon K(a, b) \quad (9)$$

Compared $K(a, b)$ with the modified agglomeration kernel $K'(a, b)$, it has some slight changes, but the trends are similar. To simplify the analysis of the frequency influence, the simplified model is still viable.

After the application of agglomeration, the initial particle number concentration is expected to decrease. The agglomeration efficiencies, η , is calculated by Eq. (10):

$$\eta_1 = \left(1 - \frac{N_1}{N_0}\right) \times 100\% \quad (10)$$

where, N is the particle number concentration, subscripts 0 and 1 represent before and after acoustic agglomeration, respectively.

2.2 Influence of high-frequency sound on agglomeration effect

The application of 20 kHz high-frequency sound source changes the particles size distribution immediately (Fig. 4).

The initial PSD shows as bimodal distribution and the peak particle diameters are 0.071 and 0.76 μm respectively. The application of high-frequency sound with the SPL of 150 dB, for the particles with the size smaller than 0.201 μm the number concentration decreases, and for the particle with the size from 0.201 to 1.945 μm the number concentration increases. The submicron particles presumably agglomerate to larger ones in the high-frequency sound field. The calculation results show that the number concentration of total aerosol decreases 10.38% from the initial of $3.37 \times 10^5 \text{ cm}^{-3}$ to the final of $3.02 \times 10^5 \text{ cm}^{-3}$. Figure 4b gives another example of the high-frequency acoustic agglomeration with the different initial PSDs. When the SPL of the high-frequency sound field is smaller, the agglomeration efficiency is a little lower, but the variation tendency of PSDs is similar.

Further study finds that the application of high-frequency sound decreases the total aerosol concentration, but the agglomeration effect is slight; besides only the particles with the size smaller than 0.25 μm adhere to agglomerate, and the sizes of the agglomerated particles are still smaller than 2.5 μm . It can be explained by entrainment factor as shown in Fig. 2. When the sound frequency is over 10^4 Hz, for the particle with size larger than 2.5 μm the entrainment factor is very close to 0. It proves that these larger particles are motionless in the

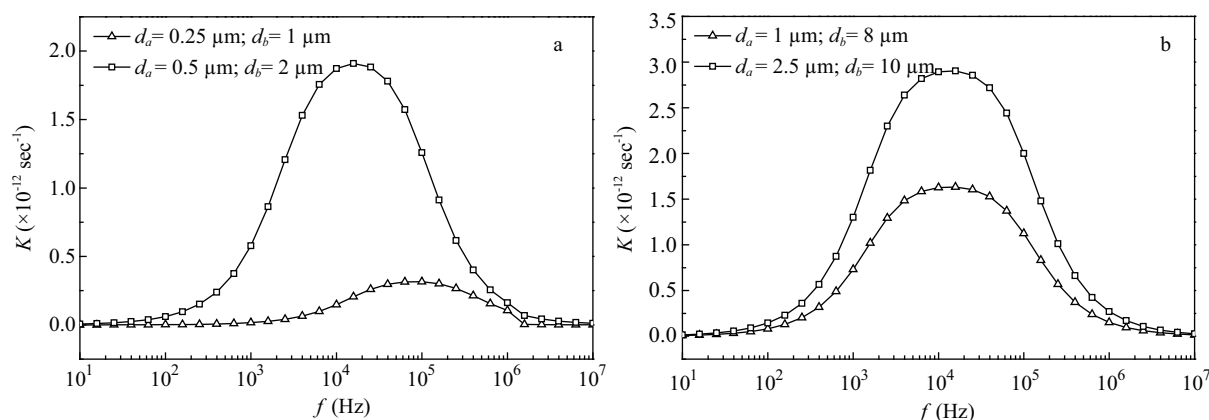


Fig. 3 Influence of frequency on agglomeration kernel for particles with different sizes.

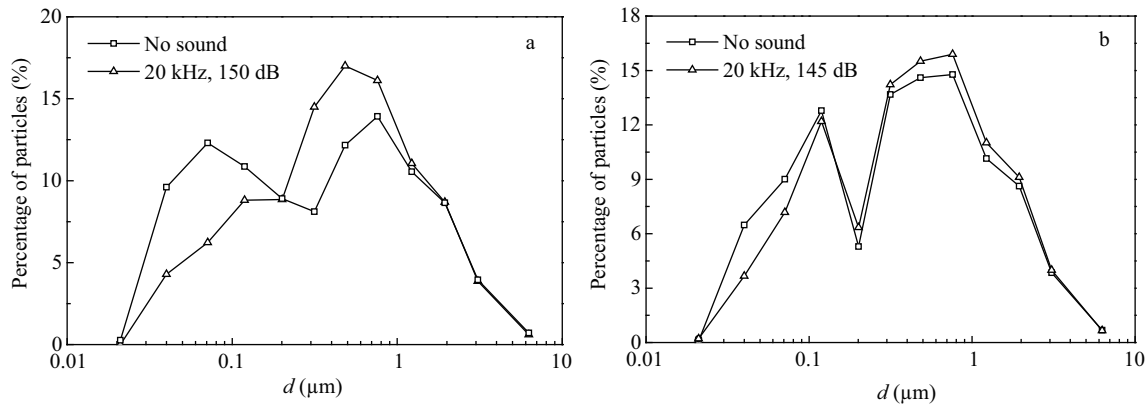


Fig. 4 Particle size distribution (PSDs) without and with high-frequency sound application.

20 kHz sound field and no relative motion can cause the collision or agglomeration. Nevertheless for micron or submicron particles with different sizes, the differences of entrainment factors are obvious. It means that in the 20 kHz sound field the submicron particles collide with each other to adhere together. It can also be proved by the agglomeration kernel. As shown in Fig. 3, the f_{opt} for submicron and micron particles is around 20 kHz, while the f_{opt} for larger particles is much lower than 20 kHz. In the industrial application, the fine particles are required to agglomerate larger than $\text{PM}_{2.5}$ for the conventional equipments to remove from the flue gas. The application of 20 kHz high-frequency sound source has some slight agglomeration effects on micro or submicron particles, but the agglomerated particles are still smaller than $\text{PM}_{2.5}$. Therefore high-frequency sound source is not an advisable pretreatment for coal-fired fly ash.

2.3 Influence of low-frequency sound on agglomeration effect

The application of low-frequency sound source also changes the particles size distribution immediately, but compared with the application of high-frequency sound source, the PSDs varies wildly (Fig. 5).

Compared with Fig. 5a and b, the initial aerosol PSDs are different, but after the applications of low-frequency sound the particle concentrations decrease for all the size range 0.03–10 μm with 12 channels. According to experimental results the total number concentrations decrease

75.3% and 62.7% respectively. It proves that the low-frequency sound is suitable to agglomerate the coal-fired fly ash particles.

Based on previous experimental and theoretical analyses, the influence of frequency is non-linear and the value of the f_{opt} is the most important and complex factor for the operating condition of acoustic agglomeration. If the frequency deviates from f_{opt} , the agglomeration efficiency will decrease (Shuster et al., 2002). Other factors, such as SPL and residence time, also affect the acoustic agglomeration efficiency. The influences of these factors mentioned above are not necessarily linear and may interact upon each other. The orthogonal design form $\text{L}_{16}(4)^3$ with total of sixteen tests is introduced to find out the optimum frequency and optimize acoustic agglomeration condition, including frequency, SPL and residence time. If a traditional full factorial design is employed to examine the effects of three factors, each at four levels, a total of 64 (4^3) tests have to be conducted. It is obvious that the orthogonal design can significantly reduce the experimental work load.

According to our previous study (Liu et al., 2009), the optimum frequencies for coal-fired fly ash were in the narrow range of 1400–1700 Hz. To achieve an efficient agglomeration, an SPL higher than 140 dB was required. As higher SPL consumed more energy, SPL of 140–150 dB was suitable. Residence time with the order of 1 sec was practicable for commercial industrial applications. In this study, these three factors are at the four levels as follows:

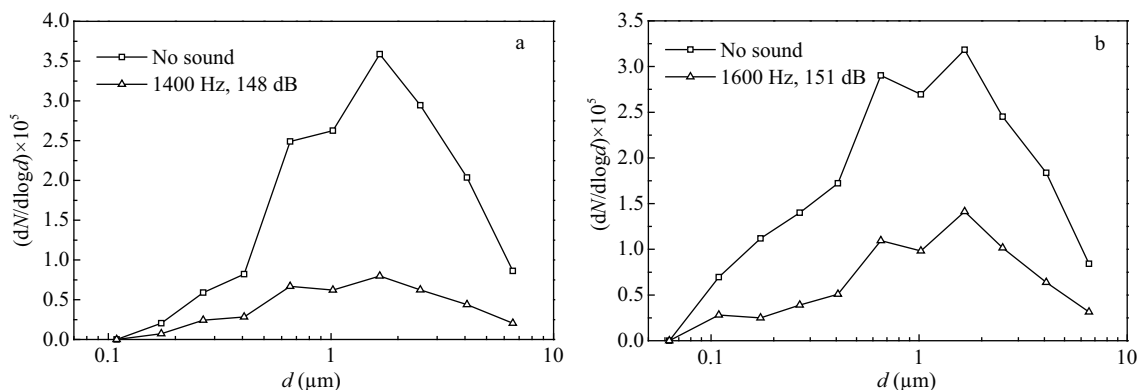


Fig. 5 PSDs without and with the application of low-frequency. (a) For the frequency of 1400 Hz, SPL of 148 dB, initial aerosol fly ash concentration of $3.16 \times 10^5 \text{ cm}^{-3}$ and residence time of 5 sec; (b) for the frequency of 1600 Hz SPL of 151 dB, initial aerosol fly concentration of $3.71 \times 10^5 \text{ cm}^{-3}$ and residence time of 4 sec.

frequency of 1000, 1400, 1600 and 1800 Hz, SPL of 135, 140, 145 and 150 dB, residence time of 4, 5, 6 and 7 sec. Table 1 shows the experimental and difference analysis results of the orthogonal design.

Based on the values of R , it proves that the influences of frequency, SPL and residence time on the agglomeration efficiency decreases in the order: $f > \text{SPL} > t$. The frequency is the dominant factor on acoustic agglomeration.

According to the level effects of influence, SPL and residence time (Fig. 6), the influence of frequency is non-linear within the range from 1000 to 1800 Hz, and the peak frequency is 1400 Hz. And the agglomeration efficiency is higher as the SPL became larger. High SPL favors the agglomeration effect because larger displacement of the particles and stronger acoustically induced turbulence can be expected. The influence of residence time is relatively small. It should be noted that too long residence time does not facilitate the improvement of agglomeration efficiency, as it offers the opportunities for the agglomerated particle to break up because of mutual collisions. Based on the above analyses, the optimal conditions of acoustic agglomeration alone are as follows: $f = 1400$ Hz, SPL = 150 dB, $t = 4$ sec.

eration alone are as follows: $f = 1400$ Hz, SPL = 150 dB, $t = 4$ sec.

Besides high-frequency sound approach has some drawbacks, such as the high energy consumption of the transducers, difficulties to penetrate into larger gas volumes, and costly sound source technology. Low-frequency sound field are much cheaper to design and easier to operate at high power than that of high-frequency counterparts. Also at lower frequency the acoustic penetration depth is much higher. Therefore the application of low-frequency is an advisable pretreatment for the conventional equipments to agglomerate the coal-fired fly ash.

3 Conclusions

In this article, comparative studies between high-frequency and low-frequency acoustic agglomeration are carried out to investigate the frequency influence on acoustic agglomeration. Theoretical analysis, experimental PSDs and orthogonal design are examined.

The results prove that although the application of 20 kHz high-frequency sound source decreases the total number concentration of the aerosol about 10%, the PSDs show that only the particles within the size ranging from 0.2 to 0.25 μm agglomerate to adhere together, and the agglomerated particles are still smaller than $\text{PM}_{2.5}$. It means high-frequency sound source is not suitable to removal coal-fired fly ash particles.

The application of low-frequency (1000–1800 Hz) gets a better agglomeration efficiency for the coal-fired fly ash acoustic agglomeration and all the number concentrations within the measuring range decrease. The highest obtainable agglomeration efficiency is up to 75.3%. The comparative study proves that low-frequency acoustic agglomeration is an advisable pretreatment for the conventional equipments to removal coal-fired fly ash particles.

Orthogonal design is introduced to investigate the influences of frequency and other factors on agglomeration efficiency. According to the orthogonal analysis results, the frequency is the dominant factor. The optimal conditions are as follows: $f = 1400$ Hz, SPL = 150 dB, $t = 4$ sec.

Acknowledgments

This work was supported by the National Basic Research Program (973) of China (No. 2010CB227001), the National Natural Science Foundation of China (No. 50576083), the Program New Century Excellent Talents University (No. NCET-04-0533) and the Zhejiang Provincial Natural Science Foundation of China (No. Y1100299).

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Table 1 Experimental and analysis results of the orthogonal design

Test No.	Factors			Agglomeration efficiency (%)
	(A) f (Hz)	(B) SPL (dB)	(C) t (sec)	
1	1000	135	4	28.15
2	1000	140	5	25.82
3	1000	145	6	24.74
4	1000	150	7	23.77
5	1400	135	5	37.71
6	1400	140	4	39.33
7	1400	145	7	44.85
8	1400	150	6	51.41
9	1600	135	6	29.44
10	1600	140	7	37.53
11	1600	145	4	41.68
12	1600	150	5	39.91
13	1800	135	7	28.18
14	1800	140	6	30.93
15	1800	145	5	35.25
16	1800	150	4	40.21
k_1	102.48 ^a	123.48	149.37	
k_2	173.31	133.62	138.69	
k_3	148.57	146.52	136.53	
k_4	134.57	155.31	134.34	
R	70.83 ^b	31.82	15.03	
Optimal level	1400	150	4	

^a $k_i^A = \sum \text{agglomeration efficiency at } A_i$, ^b $R_i^A = \max\{k_i^A\} - \min\{k_i^A\}$.

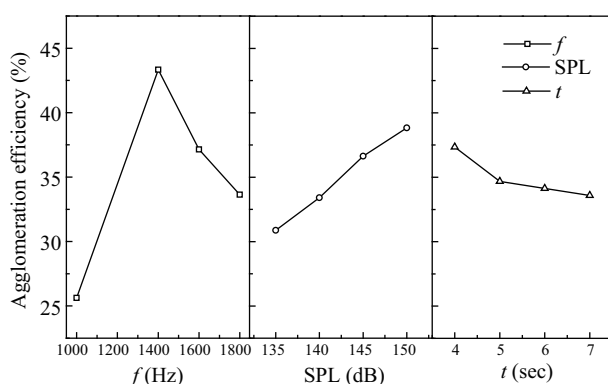


Fig. 6 Level effects of frequency (f), SPL and residence time (t).

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