Research Article

Design of a mono-disperse aerosol generator for efficiency testing of HEPA filter

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Abstract

A set of mono-disperse aerosol generator was designed to meet the requirement of efficiency testing for high efficiency particle air filter. The aerosol generation tests and performance tests were conducted by using evaporation–condensation method, with NaCl solutions at different concentrations as the condensation nucleus and respectively using the DEHS, DOP, PAO–4 as reagents. The results show that three reagents can generate mono-disperse aerosol particles by strictly controlling various parameters which affects the aerosol performance, where the particle size range is 0.33–0.36 μm for DEHS, 0.35–0.37 μm for DOP and 0.34–0.36 μm for PAO–4 and the concentrations of the aerosols lager than 10^6 cm^{-3}. The particle size characteristics and concentrations generated through such method basically conform to the requirements of efficiency testing for high efficiency particle air filter.

Keywords Mono-disperse aerosol · Efficiency testing · High efficiency particle air filter · Evaporation–condensation method

1 Introduction

The radioactive aerosols formed by suspension of radioactive materials in the air are one of the most important pollutant sources in the workplace environment, such as nuclear technology applications, nuclear facilities, and nuclear and radiation terrorist incidents [7, 15, 18, 19]. The radioactive aerosols can invade into the human body through the human breathing, skin absorption and other means, which is one of the main ways to cause the internal irradiation hazards in the human body, [1, 9, 12, 13]. The high efficiency particle air (HEPA) filter is usually used in nuclear facility air purification system to remove the radioactive aerosol. This type of filter is characterized by the filtration efficiency of not less than 99.97% for particles with particle size of 0.3 μm. According to the requirements of US Military Standards MIL-STD-282 [3], each HEPA filter is tested for its efficiency by using the mono-disperse DOP aerosol with particle size of 0.30 ± 0.03 μm before leaving the factory.

Aerosol generators are widely used in aerosol instrument calibration, aerosol movement law research, aerosol respiratory deposition law research, filter aerosol mechanism research, and aerosol filter testing. The test aerosol produced is mono-disperse or poly-disperse, spherical or non-spherical, solid or liquid particles [2]. The ideal aerosol generator is required to continuously produce stable spherical solid aerosol particles and conveniently control the particle size and concentration of the aerosol particles. The commonly used methods for producing spherical mono-dispersity aerosols include atomization, vibration, and condensation [4, 11, 16]. At present, the evaporation–condensation method is a commonly used generation method for mono-disperse aerosol [14, 21]. The condensation-type mono-disperse aerosol generator can quickly generate the sub-micron mono-disperse aerosol particles (geometric standard deviation $\sigma_g \leq 1.25$)

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with adjustable concentration and suitable for HEPA filter efficiency testing. This method was first proposed by Sinclair and Lamer, employing high-voltage electric sparks to heat NaC1 to produce condensation nucleus. It presented extremely high electrical insulation requirements and poor stability. Prodi improved the above device, used a sprayer to generate condensation nucleus, and adjusted the particle size of condensation nucleus by controlling the pressure of compressed air and the dilution ratio of the solution. In the past 20 years, the research on this type of aerosol generator has primarily used electromigration technology to produce nanometer aerosol particles [6, 17]. A series of research results and patented technology have made the mono-dispersion aerosol generator a product. Besides, it has exhibited relatively mature technology, stable performance, and broad application after gradual improvement and innovation. However, it is expensive. In China, there are no reports on the development of this equipment. This subject aims to master the generation method of condensation type mono-disperse aerosol through exploration and research, laying the foundation for the development of mono-disperse aerosol generator for HEPA filter detection. Considering that the DOP (Di-n-octyl-o-phthalate and 1-decene) reagents are carcinogenic and strongly irritative, there is a trend to use the non-toxic PAO (polydecene) liquids to substitute DOP in the world. Therefore, we have also studied the method to generate mono-disperse aerosols by using PAO.

This paper mainly introduces the design and establishment of a mono-disperse aerosol generator and emphatically introduces the test process of generating DEHS (Di-2-ethyl hexyl sebacate), DOP and PAO–4 (1-decene, tetramer mixed with 1-decene, commonly known as Emery 3004) mono-disperse aerosols. The best process parameters and particle size characteristic parameters of generated aerosols under the existing conditions are selected. The geometric mean diameter of the generated aerosol particles is about 0.35 μm, $\sigma_g \leq 1.25$ and its concentration is 10^6 cm^{-3} or over.

2 Theory

2.1 Basic principle

The condensation-type mono-disperse aerosol generator generates the monodisperse aerosol by the principle of evaporation–condensation, that is, heating a liquid state aerosol substance (such as DEHS) to evaporates it into the supersaturated vapor, and then suddenly cooling to make the supersaturated vapor condense to form the aerosol particles. Based on the presence or absence of condensation nucleus in the condensation process, the condensation test can be divided into homogeneous condensation and heterogeneous condensation. The homogeneous condensation does not provide the additional condensation nucleus in the condensation process, therefore, the steam condensation environment is greatly different and the monodispersity is also poor; the heterogeneous condensation has the condensation nucleus to evenly condensate the steam and can generate the aerosols with good monodispersity, therefore, it is widely used.

The basic principle of condensation-type mono-disperse aerosol generator is heterogeneous condensation and the source of condensation nucleus is generally the solid NaCl crystal particle. Figure 1 is the generation principle diagram. In Fig. 1, the clean nitrogen is used as the carrier gas and passes through the sprayer with inorganic salt solutions (generally NaCl solutions) at the certain pressure and flow rate, after the atomization, the small liquid drops are dried and crystallized into solid NaCl aerosol particle as condensation nucleus. The condensation nucleus particles are mixed with the aerosol substance evaporated from the saturator by heating and then enter into the reheater. The reheater heats the liquid drops in the mixture to completely evaporate into supersaturated vapor. Finally, the mixture (condensation nucleus particles, aerosol vapor, and nitrogen) enters into the condenser pipe and is suddenly cooled, and the vapor is condensed on the condensation nucleus to form the mono-disperse aerosol [5, 10, 20].

2.2 Technological process

According to the principle in Fig. 1, a process flow of mono-disperse aerosol generator as follows is designed

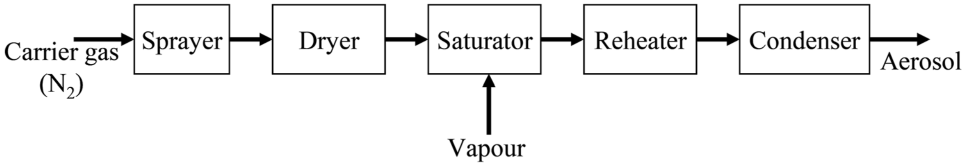


Fig. 1 Schematic diagram of evaporation–condensation method

(see Fig. 2). High-purity nitrogen enters into the sprayer under certain pressure after passing through the pressure regulating valve, and then sprays NaCl solution in the sprayer into fine NaCl drops which enter into the silica gel drying column and then form tiny NaCl solid crystal particles as the condensation nucleus after desiccation and crystallization. Later on, the condensation nucleus is divided into two parts: one part enters into the saturator containing liquid organic matters, and the other part directly enters into the reheater. The Liquid organic matters in the saturator are heated to evaporate and enter into the reheater under certain pressure and temperature, and those entering into the reheater are the mixture of condensation nucleus, nitrogen and vapor of organic matters. The mixture is further heated in the reheater to ensure no condensation prior to entry into condenser. Finally, in the condenser, the organic matters condensate on the condensation nucleus and form the mono-disperse aerosol.

2.3 Main technical specifications

Aerosols with particle size conforming to logarithmic normal distribution can be signified by geometric mean diameter d_g and geometric standard deviation σ_g:

$$d_g = \prod d_i^{\left(N_i/N_t\right)} \tag{1}$$

$$\sigma_g = \exp\{\sum [N_i(\ln d_g - \ln d_i)^2/N_t]\}^{1/2} \tag{2}$$

where d_i is the mean particle size of aerosol particle at the ith part; N_i is the number of aerosol particles at the ith part; N_t is total number of particles.

Aerosol dispersion is signified by relative standard deviation α, which is the ratio of the standard deviation of particle size to the mean radius of particles. Judgment standard for mono-disperse aerosol can be represented by $\alpha < 0.2$ ($\ln\sigma_g < 0.2$) or $\sigma_g < 1.25$. Hence, aerosol with $\sigma_g < 1.25$ is one condition for mono-dispersion, which is also main technical index of the designed device.

2.4 Particle size measurement

Particle size is the most attribute of aerosol behavior and characteristics. Different particle size testing technologies are the measurements on different particle characteristics, together with the measurement results to be different equivalent diameters. Common particle size measurement methods comprise: optical method, sedimentation method and scattering method, etc. Particle size measurement method used in this work is the scattering method.

Instrument used for measurement of aerosol particle size is Malvern Mastersizer2000 particle size measurer which measures the particle size by virtue of the scattering (diffraction) of particles to light, with particle size measurement in 0.02 μm ~ 2000 μm. Measurement principle of Mastersizer 2000 particle size measurer is as shown in Fig. 3. It consists of transmission, receiver and measuring window. Transmission part is constituted by the laser and beam processor, of which the laser can offer parallel lights of two different wave lengths. Measuring window is to obtain the particle size information of the sample. Receiver consists of Fourier lens and photodetector arrays. To enlarge the receiving angle of scattered light, a large-angle detection system is designed, which expands the lower detection limit of the instrument.

3 Experimental

3.1 Aerosol experiment with DEHS

According to Ref. [8], the mass concentration of NaCl solution C_1, total flow Q_t, saturator flow Q_b, bypass flow Q_p and saturator temperature T_1 are the main parameters influencing the geometric mean diameter d_g of aerosol particle.

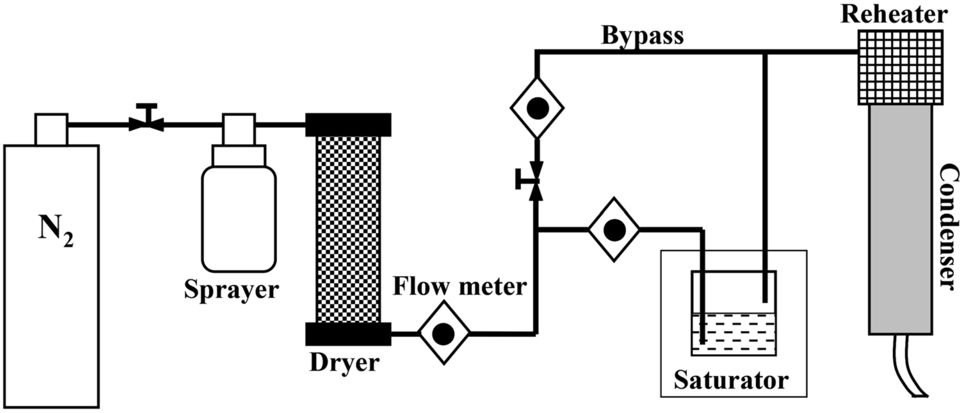


Fig. 2 The process flow of mono-disperse aerosol generator

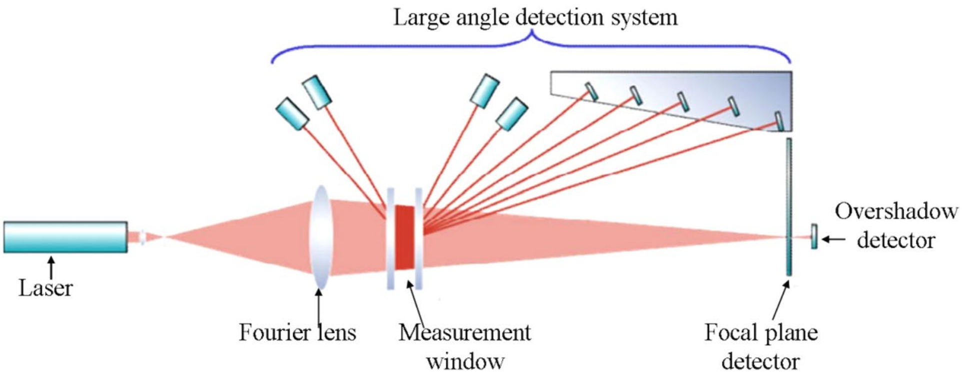


Fig. 3 Schematic diagram of the measuring principle of Mastersizer2000

Select the operating parameters recommended by Ref. [8]: concentration of NaCl solution $C_1 = 20$ mg L^{-1}, total flow $Q_t = 210$ L h^{-1}, saturator flow $Q_b = 150$ L h^{-1}, bypass flow $Q_p = 60$ L h^{-1} and saturator temperature $T_1 = 120 \sim 200$ °C, adjust the reheater temperature T_2 (100 ~ 400 °C) and then generate the aerosol with DEHS. Measurement on the samples under various conditions shall be made with Mastersizer2000 particle size measurer, and d_g and σ_g of samples are calculated.

3.2 Aerosol experiment with DOP

With different concentrations of NaCl solution selected, under C_1 respectively at (1) 10 mg L^{-1}; (2) 20 mg L^{-1}; (3) 40 mg L^{-1}; (4) 250 mg L^{-1}, total flow $Q_t = 210$ L h^{-1}, saturator flow $Q_b = 150$ L h^{-1}, bypass flow $Q_p = 60$ L h^{-1}, and $T_1 = 100 \sim 160$ °C, T_2 (from 100 ~ 400 °C) is adjusted, and aerosol is generated by DOP. Measurement is made to the samples under various conditions with Mastersizer2000 particle size measurer, and d_g and σ_g of various samples are calculated.

3.3 Aerosol experiment with PAO–4

With different concentrations of NaCl solutions selected, under C_1 respectively at (1) 10 mg L^{-1}; (2) 20 mg L^{-1}; (3) 250 mg L^{-1}, total flow $Q_t = 210$ L h^{-1}, saturator flow $Q_b = 150$ L h^{-1}, bypass flow $Q_p = 60$ L h^{-1}, and $T_1 = 100 \sim 190$ °C, T_2 (from 100 ~ 400 °C) is adjusted, and aerosol is generated by PAO–4. Measurement is made to the samples under various conditions with Mastersizer2000 particle size measurer, and d_g and σ_g of various samples are calculated.

4 Results and discussion

4.1 Experiment results of DEHS

According to the results (see Table 1) of measurement samples, $T_1 = 150$ °C is the optimal condition generating the mono-disperse DEHS aerosol, and T_2 is of a wide requirement range within 150 ~ 325 °C, with particle size range at 0.33 ~ 0.36 μm, $\sigma_g \le 1.25$, and the concentrations of aerosol generated under various temperature conditions above 10^6 cm^{-3}. Figure 3 is the particle size distribution diagram of one representative sample, and from the diagram, it can be seen that concentration of particle is up to 3.0×10^6 cm^{-3}.

4.2 Experiment results of DOP

According to the results of measurement samples, it can be obtained that, upon $T_1 = 120$ °C, DOP aerosols generated by NaCl solutions of four different concentrations are of best mono-dispersion, and all σ_g are basically less than 1.20 (except for very few samples), so that $T_1 = 120$ °C is the optimal condition.

According the data calculated from measured samples, d_g and σ_g of DOP aerosols generated by NaCl solutions of four different concentrations change with increase of T_2 (see Figs. 4 and 5). From Figs. 4 and 5, it can be seen that, when DOP aerosol with NaCl solutions of four different concentrations as condensation nucleus

Table 1 Effect of reheater temperature on DEHS aerosol

T_2 (℃)	d_g (μm)	σ_g
100	0.37	1.35
125	0.42	1.72
150	0.36	1.23
175	0.36	1.22
200	0.36	1.20
225	0.35	1.18
250	0.35	1.19
275	0.34	1.23
300	0.34	1.22
325	0.33	1.24
350	0.33	1.26
375	0.33	1.27
400	0.33	1.25

is at $T_1 = 120$ °C: (1) d_g of aerosol generated under different concentrations are within 0.34 ~ 0.38 μm; (2) When $C_1 = 10, 20, 40$ mg L^{-1}, d_g increases with the increase of T_2 at first and then decreases; Upon $C_1 = 250$ mg L^{-1}, from $T_2 = 150$ °C, d_g decreases with the increase of T_2, and in case of $T_2 = 400$ °C, d_g is minimum, at 0.34 μm; (3) All σ_g of aerosols generated under various concentrations are almost less than 1.25 (except for two points); (4) Upon $T_2 \geq 200$ °C, all σ_g of aerosols generated under various concentrations are less than 1.20; (5) When T_2 changes in 100 ~ 400 °C, all σ_g of aerosol generated upon $C_1 = 250$ mg L^{-1} are less than 1.20, which is the optimal condition for the generation of the mono-disperse aerosol.

Fig. 4 Effect of reheater temperature on DOP aerosols' geometric mean diameter. When $Q_t = 210$ L h^{-1}, $Q_b = 150$ L h^{-1}, $T_1 = 120$ °C

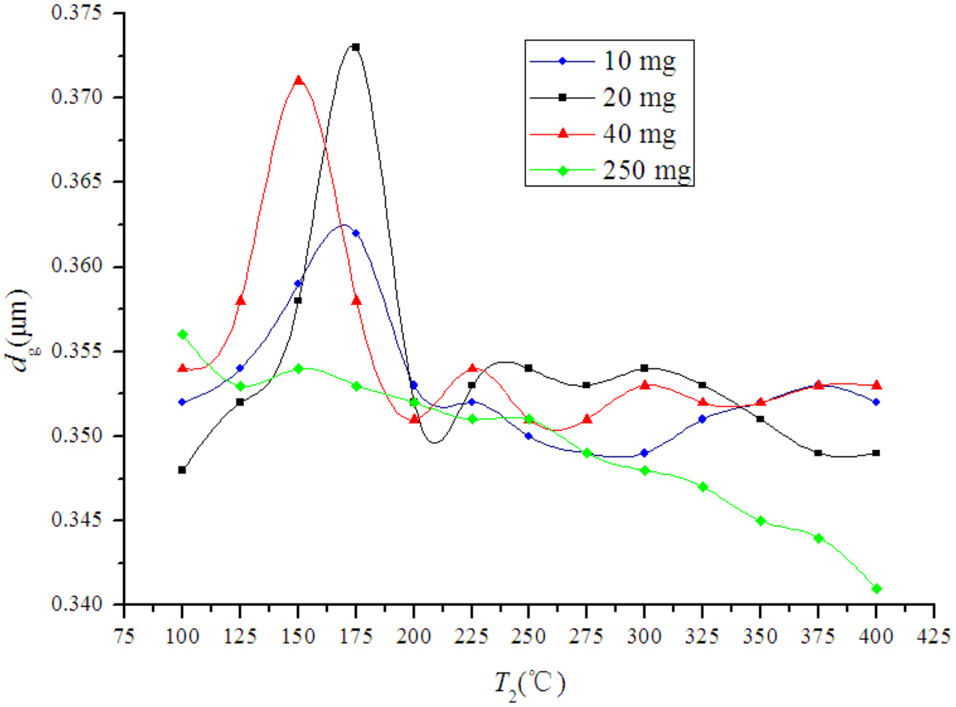
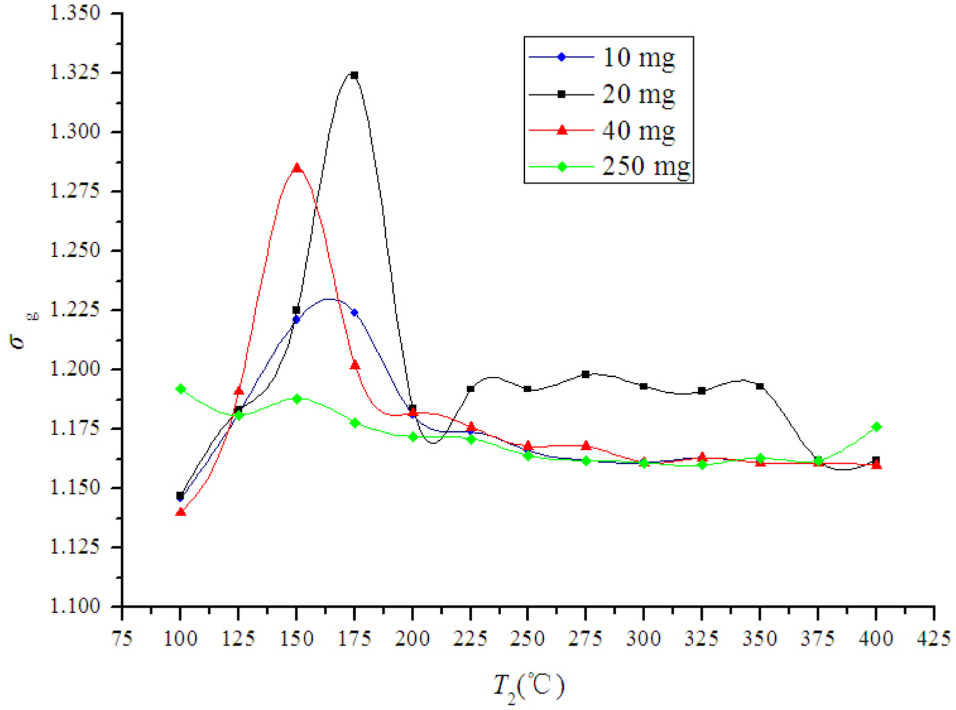


Fig. 5 Effect of reheater temperature on DOP aerosols' geometric standard deviation. When $Q_t = 210$ L h^{-1}, $Q_b = 150$ L h^{-1}, $T_1 = 120$ °C



In summary, in the test of DOP aerosol generated with NaCl solutions of four different concentrations as condensation nucleus, in case of controlled flow $Q_t = 210$ L h^{-1}, $Q_b = 150$ L h^{-1} and $Q_p = 60$ L h^{-1}, $C_1 = 250$ mg L^{-1} and $T_1 = 120$ °C are the optimal conditions for the generation of mono-disperse DOP aerosol, and T_2 is of a wide requirement range within 100 ~ 400 °C, with particle size range at 0.34 ~ 0.36 μm, $\sigma_g \le 1.20$, and corresponding particle concentration more than 10^6 cm^{-3}.

4.3 Experiment results of PAO–4

According to the measurement results, upon $T_1 = 150$ °C, PAO–4 aerosols generated by NaCl solutions of three different concentrations are of best mono-dispersion, and all σ_g are less than 1.20 except for individual samples, so that $T_1 = 150$ °C is a optimal condition.

According the data calculated from measured samples, d_g and σ_g of PAO–4 aerosols generated by NaCl solutions of three different concentrations change with increase of T_2 (see Figs. 6 and 7). From Figs. 6 and 7, it can be seen that, when PAO–4 aerosol generated with NaCl solutions of three different concentrations as condensation nucleus is at $T_1 = 150$ °C: (1) Upon $C_1 = 250$ mg L^{-1} and $T_2 = 100$ °C, d_g is the maximum at 0.425 μm, and meanwhile, σ_g is the maximum at 1.65. Later on, d_g decreases with increase of T_2. Upon $C_1 = 10$, 20 mg L^{-1}, from $T_2 = 100$ °C, d_g increases at first and then decreases, so as the change trend of σ_g; (2) Upon $T_2 \ge 150$ °C, d_g and σ_g curve of aerosols generated under various concentrations basically coincide, indicating small influence of NaCl solution concentration on d_g and σ_g upon T_2 within 150 ~ 400 °C; (3) Upon T_2 within 150 ~ 400 °C, all σ_g of aerosols generated under various concentrations are less than 1.20; Especially upon $T_2 = 200$ °C, d_g is about 0.35 μm, with minimum σ_g about 1.16, but best mono-dispersion.

In summary, in the test of PAO–4 aerosol generated with NaCl solutions of three different concentrations as condensation nucleus, in case of controlled flow $Q_t = 210$ L h^{-1}, $Q_b = 150$ L h^{-1} and $Q_p = 60$ L h^{-1}, $T_1 = 150$ °C is the optimal condition for the generation of mono-disperse PAO–4 aerosol, and T_2 is of a wide requirement range within 150 ~ 400 °C, with particle size range at 0.34 ~ 0.36 μm, $\sigma_g < 1.20$, and corresponding particle concentration more than 10^6 cm^{-3}.

5 Conclusions

A set of aerosol generation experimental device is design according to the aerosol generation principle of evaporation–condensation method. On the experimental device, with parameters able to affect aerosol performance under strict control, aerosols with good mono-dispersion ($\sigma_g \le 1.25$) and particle concentration more than 10^6 cm^{-3} can be generated with DEHS, DOP and PAO–4 reagents. The optimization conditions are slightly different, upon use of DOP, the saturator temperature is

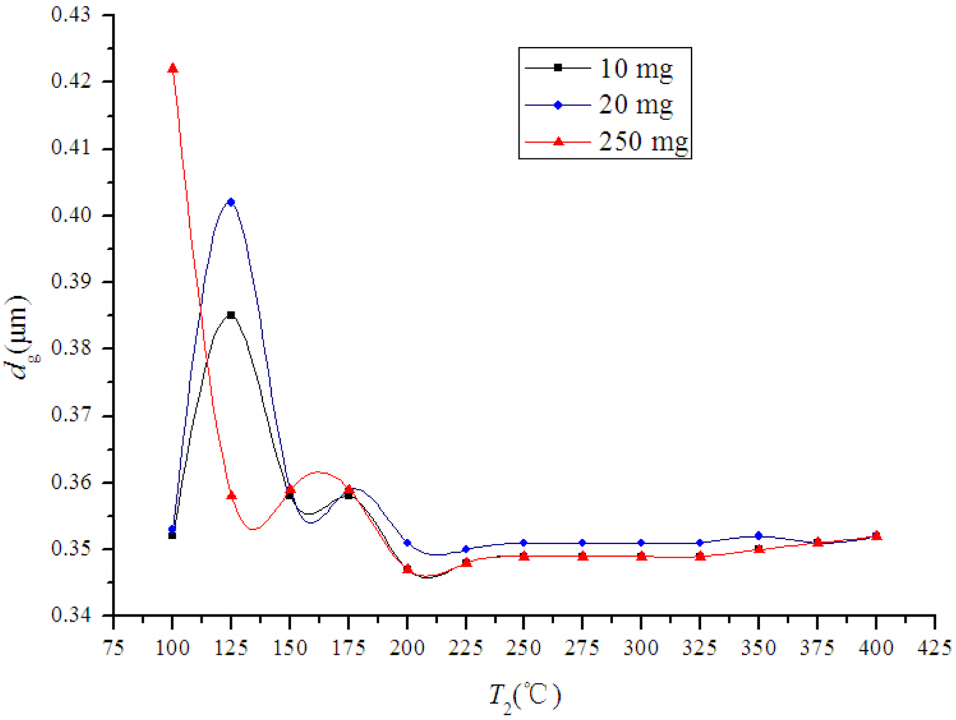


Fig. 6 Effect of reheater temperature on PAO–4 aerosols' geometric mean diameter. When $Q_t = 210$ L h^{-1}, $Q_b = 150$ L h^{-1}, $T_1 = 150$ °C

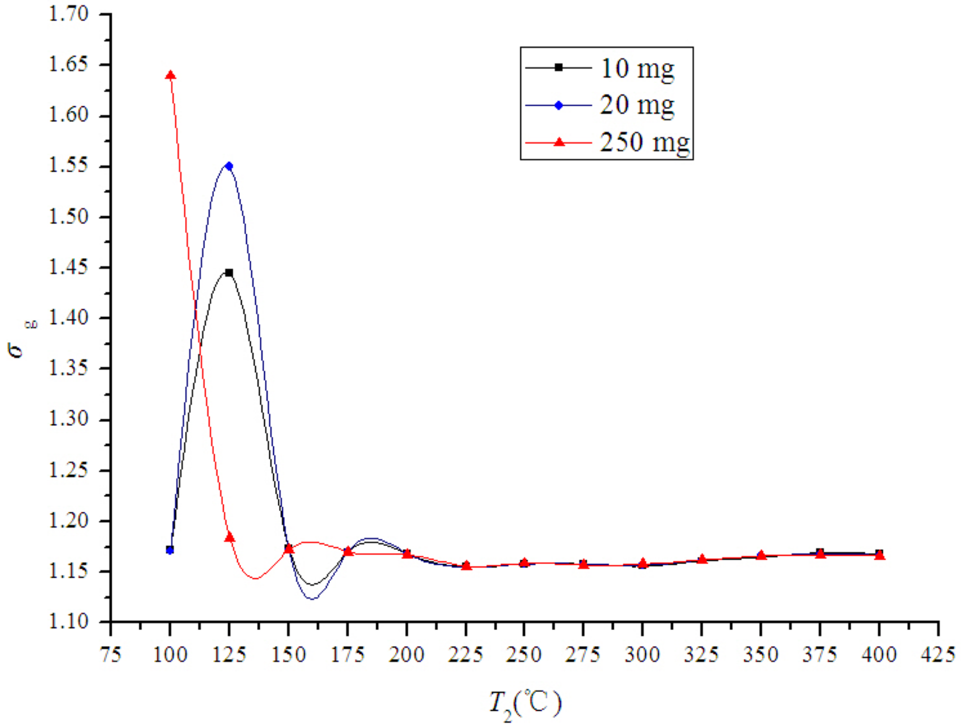


Fig. 7 Effect of reheater temperature on PAO–4 aerosols' geometric standard deviation. When $Q_t = 210$ L h^{-1}, $Q_b = 150$ L h^{-1}, $T_1 = 150$ °C

at 120 °C, while it is at 150 °C in using DEHS and PAO–4; concentrations of NaCl are also different, but which is easy to achieve. These three reagents all can be taken as reagents for efficiency testing of nuclear HEPA filter, of which DOP has been extensively applied in the production practices, but PAO-4 is of a better comprehensive performance. Particle size range of mono-disperse aerosols generated by the three reagents respectively is 0.33 ~ 0.36 μm, 0.35 ~ 0.37 μm, 0.34 ~ 0.36 μm and large than (0.30 ± 0.03) μm, but has a little influence on testing result; Mono-disperse aerosol particles (0.30 ± 0.03) μm generated from further experiment is our next assumption and target.

The evaporation–condensation method can produce aerosols with acceptable mono-dispersity, providing technical support for the efficiency testing of the HEPA filter. Besides, the secondary waste generated during the method operation is less, generating social and economic benefits. The built aerosol generator presents low cost, reliable performance, and flexible use. Moreover, the generated particles can meet the simulation of radioactive aerosol characteristics and the research on filtration purification technology. However, the aerosol generator needs to use high-pressure nitrogen as a carrier gas, limiting it to the laboratory environment. The use of the device can be effectively expanded if the sprayer device can be improved and the ultrasonic atomization method is used to achieve the atomization of the NaCl solution.

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