ANALYTICAL METHODS FOR DETERMINATION OF AEROSOLS BY MEANS OF MEMBRANE ULTRAFILTERS. XIX.*

EFFICIENCY MEASUREMENT OF NUCLEAR PORE FILTERS BY MEANS OF LATEX AEROSOLS

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The filtration properties of Nuclear Pore Filters were studied by using them for latex aerosol filtration. The filtration efficiency, the selectivity and the filtration kinetics of these filters were measured by means of model latex aerosols in the range of particle sizes 0·3 through 1·1 μm and by means of a particle counter Royco. The former filtration equation of a pore filter was modified according to the experimental results. The selectivity of these filters was found excellent and the clogging for larger particle sizes very slow.

In some previous communications[1-5] the structure and filtration properties of nuclear pore filters have already been described. It was also found, that the agreement between filtration theory and experimental results of measurement was practically quantitative[1] in the range of particle sizes 0·005 through 0·5 μm (diameters). The radioactive labelled condensation aerosols[6,7] were used for this measurement.

The measurement of filtration efficiencies by means of aerosols with larger particle sizes of latex aerosols, the studying of filtration selectivity and the comparison of these results with our previous filtration theory are subject of this communication.

EXPERIMENTAL

Model filters. American analytical NPF[1] (production by General Electric, Pleasanton, U.S.A.) of pore sizes from 0·5 to 8·0 μm in diameter were used as a model pore filters. Their structural properties (pore sizes, porosities, thickness *etc.*) were measured applying the former described methods[1,4,5].

Model aerosols. The filtration efficiencies were measured by means of monodisperse LA. The suspensions of monodisperse latex (Dow Chemical Company, Serva Heidelberg, Germany) with particle sizes (diameters) of 0·357 μm, 0·714 μm and 1·099 μm were used for aerosol genera-

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tion. The aerosols were generated by atomizing a dilute suspension of these latex particles in a glass atomizer. The latex suspension was diluted from its original concentration (10%) according to the theory of dilution[8,9]. Practically the solutions of monodisperse latex in bidistilled water of concentration about 0·01 through 0·2% were found the best. The electric charges of these aerosols were not measured. But satisfactory data[10] had already been published demonstrating that these charges were relatively high (10 to 20 e).

Equipment. The schematic of the equipment for filter efficiency measurement is in the Fig. 1. A plexiglass sphere with a diameter of about 80 cm was a good aerosol chamber working under conditions of a dynamical equilibrium. Monodisperse aerosols of polystyrene latex particles were produced by atomizing a dilute suspension of latex particles in a glass atomizer (*G*). The electrical charge of those particles was decreased by irradiation with a polonium source (Po, 2 mCi). The dry pressure gas for atomizer (*PG*) was filtered (F_2) and its pressure was measured (M_1, 0·9 atm). After atomization the aerosol was mixed with dry filtered air (F_1) in the sphere. The LA coming into the sphere are moved then through the measured filter (*PF*) with a flow rate Q_2 and through the pump (*P*) with a flow rate Q_1. The total flow rate through the sphere is Q (l/min). The flow rates are measured by flowmeters (Q_1, Q_2, Q). The pressure in the flowmeter Q_2 and the pressure drop in the filter *PF* is measured by a manometer M_2. To determine the particle concentration before and behind the filter *PF*, an optical particle counter (Royco PC 200) was connected with the filter holder. The numbers of counted particles were registered by means of a recorder (*R*).

Procedure. The glass atomizer was filled with a diluted suspension of latex. The plexiglass sphere was "cleaned out" from any particles. The air filtered by means of an absolute filter (F_1) was sucked by the pump (*P*) through the sphere. The particle concentration in the sphere was measured by means of the particle counter Royco. The atmosphere in the chamber was considered clean when the particle concentration was smaller that about 20 particles in a liter of air. Then the aerosol was generated and introduced into the sphere. The aerosol concentration before the filter (*PF*) was measured continuously. If the aerosol concentration is stabile (it takes about 20–30 minutes) and not too high (the aerosol concentration should be smaller than 50 cm^{-3}), the measurement of filter efficiency could start. The aerosol concentrations before and behind the filter were measured continuously and registered as a function of time. It was possible to

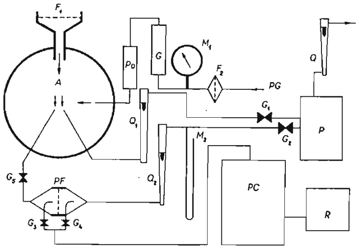

FIG. 1

Schematic Diagram of Experimental Apparatus for Efficiency Determination
Description in the text.

change the pore sizes of filters, the particle sizes of latex aerosols and the filter face gas velocities. The filter efficiency could therefore be measured as a function of particle radius (r), pore radius (R) and filter face gas velocity (q).

RESULTS AND DISCUSSION

Verification of Former Pore Filter Equations

In a previous communication[1] the equation for a total collection efficiency of a pore filter was verified by means of different condensation aerosols, in the particle ranges 0·005 to 0·5 μm (diameters) and under conditions

$$N_R = r/R \ll 1 \, .$$

The agreement of experiments with the theory was relatively good and the empirical factor for the direct interception collection, δ, was found to be $\approx 0{\cdot}15$.

Using latex aerosols also the total collection efficiencies under conditions of $N_R \leq 1$ were able to be measured. The measured data were compared with the theoretical total collection efficiency using our former[1] equations:

$$E_1 = \varepsilon_i + \varepsilon_D - \varepsilon_i \varepsilon_D \, , \qquad (1)$$

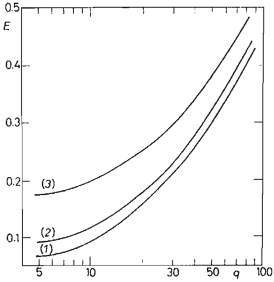

FIG. 2

Dependence of Efficiency E on Flow Rate q(cm/s)

R 4·0 μm, r 0·357 μm: computed according to equations indicated in the figure.

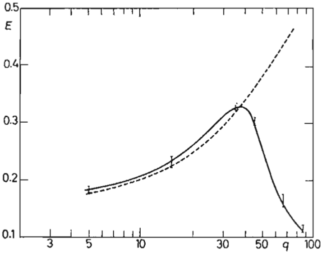

FIG. 3

Measured Dependence of Efficiency E on Flow Rate q (cm/s)

R 4·0 μm, r 0·357 μm, pointed curve means the theory.

for the impaction (ε_i) and diffusion (ε_D) mechanisms, and

$$E_2 = \varepsilon_i + \varepsilon_D + \delta\varepsilon_R - \varepsilon_i\varepsilon_D - \delta\varepsilon_i\varepsilon_R , \qquad (2)$$

for the impaction, diffusion and direct interception (ε_R) mechanisms. (The equations describing ε_D, ε_i and ε_R were already published[1]; ε_i, ε_D, ε_R are the partial efficiencies for apparent mechanisms).

As the agreement between theory (equations (*1*), (*2*)) and experimental results under those conditions was not good enough (the experimental efficiencies were higher than the theoretically predicted ones), we have tried to find the reason. As the electric charges of aerosol particles were eliminated, the collection by means of the direct interception mechanism seemed to be more important for the total collection efficiency than the factor $\delta = 0{\cdot}15$ permitted.

TABLE I

Efficiency Table of a Nuclear Pore Filter (Theory and Experiment)

Aerosol: r 0·549 μm; s 1·0 g cm^{-3}; filter: R 1·000 μm; P 0·063; L 11·5 μm; temperature 273K; viscosity 181·10^{-6} g cm^{-1} s^{-1}; ε_R 0·797.

q cm s^{-1}	ε_i	ε_D	E_1	E_2	E_3	E_{exp}
0·1	0·006	0·136	0·141	0·260	0·784	—
5·0	0·266	0·038	0·294	0·381	0·769	0·771 ± 0·011
10·0	0·442	0·037	0·462	0·529	0·823	—
15·0	0·556	0·036	0·572	0·625	0·859	0·862 ± 0·009
20·0	0·632	0·036	0·645	0·689	0·883	—
25·0	0·685	0·036	0·697	0·734	0·901	—
30·0	0·724	0·036	0·734	0·767	0·913	—
35·0	0·753	0·036	0·762	0·791	0·922	0·960 ± 0·014
40·0	0·775	0·036	0·783	0·810	0·929	—
45·0	0·793	0·036	0·801	0·825	0·935	—
50·0	0·808	0·036	0·815	0·838	0·939	—
55·0	0·820	0·035	0·826	0·848	0·942	0·902 ± 0·012
60·0	0·830	0·035	0·836	0·856	0·946	—
65·0	0·838	0·035	0·844	0·863	0·949	—
70·0	0·846	0·035	0·851	0·869	0·951	—
75·0	0·852	0·035	0·857	0·875	0·953	—
80·0	0·857	0·035	0·862	0·879	0·954	—
85·0	0·862	0·035	0·867	0·884	0·956	—
90·0	0·867	0·035	0·871	0·887	0·957	—
95·0	0·870	0·035	0·875	0·891	0·959	—
100·0	0·874	0·035	0·878	0·893	0·959	—

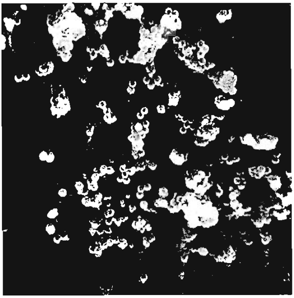

FIG. 7

Latex Particles Separated on the NPF Surface, Mostly by Impaction Mechanism $R(2{\cdot}55\ \mu m$, $r\ 0{\cdot}044\ \mu m$, $0{\cdot}214\ \mu m$ and $0{\cdot}398\ \mu m)$.

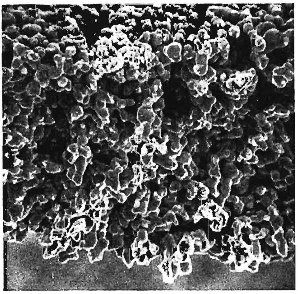

FIG. 10

The Structure of a Silver Membrane Filter (1 : 4500)

After the evaluation of many experiments, we have decided for an empirical factor $\vartheta^{(1-N_R)}$ and have found that $\vartheta \approx 0{\cdot}63$. This coefficient seems to be useful for a larger scale of N_R and therefore also more common. Then the equation for the total efficiency was

$$E_3 = \varepsilon_i + (1 - \varepsilon_i)\left[\varepsilon_D + \varepsilon_R \vartheta^{(1-N_R)}\right] . \qquad (3)$$

It is necessary to add that the equations for the total efficiency (*2*) and (*3*) are semi-empirical equations, not exactly derived as it was the case of the equation (*1*), respecting the two mechanisms (impaction and diffusion), only. Because of this reality and because of the influence of many others not well known factors, the agreement found could be considered as successful. The equation (*3*) permits to predict a total collection efficiency of a model pore filter. It is often a little smaller than the efficiency of a real pore filter.

TABLE II

Efficiency Table of a Nuclear Pore Filter (Theory and Experiment)

Aerosol: r 0·179 μm; s 1·0 g cm^{-3}; filter: R 2·500 μm; P 0·078; $L = 14{\cdot}0$ μm; temperature 273 K; viscosity $181 \cdot 10^{-6}$ g cm^{-1} s^{-1}; ε_R 0·138.

q cm s^{-1}	ε_i	ε_D	E_1	E_2	E_3	E_{exp}
0·1	0·000	0·113	0·114	0·134	0·204	—
5·0	0·014	0·037	0·051	0·071	0·140	—
10·0	0·028	0·036	0·063	0·083	0·150	0·171 ± 0·016
15·0	0·041	0·036	0·076	0·095	0·162	—
20·0	0·055	0·036	0·088	0·108	0·173	—
25·0	0·068	0·036	0·101	0·120	0·185	0·210 ± 0·013
30·0	0·081	0·036	0·113	0·132	0·196	—
35·0	0·093	0·036	0·125	0·144	0·207	—
40·0	0·106	0·035	0·137	0·156	0·217	0·210 ± 0·011
45·0	0·118	0·035	0·149	0·167	0·228	—
50·0	0·130	0·035	0·161	0·179	0·239	—
55·0	0·142	0·035	0·172	0·190	0·249	—
60·0	0·153	0·035	0·183	0·201	0·259	—
65·0	0·165	0·035	0·194	0·211	0·269	0·113 ± 0·019
70·0	0·176	0·035	0·205	0·222	0·279	—
75·0	0·187	0·035	0·215	0·232	0·288	—
80·0	0·197	0·035	0·226	0·242	0·298	—
85·0	0·208	0·035	0·236	0·252	0·307	0·091 ± 0·014
90·0	0·218	0·035	0·246	0·262	0·316	—
95·0	0·229	0·035	0·256	0·272	0·325	—
100·0	0·239	0·035	0·265	0·281	0·333	—

TABLE III

Efficiency Table of a Nuclear Pore Filter (Theory and Experiment)

Aerosol: r 0·357 µm; s 1·0 g cm^{-3}; filter: R 4·000 µm; P 0·051; L 12·2 µm; temperature 273 K; viscosity 181·10^{-6} g cm^{-1} s^{-1}; ε_R 0·171.

q cm s^{-1}	ε_i	ε_D	E_1	E_2	E_3	E_{exp}
0·1	0·000	0·045	0·046	0·072	0·158	—
5·0	0·032	0·035	0·066	0·091	0·175	0·179 ± 0·009
10·0	0·063	0·035	0·096	0·120	0·201	—
15·0	0·093	0·035	0·125	0·148	0·227	0·231 ± 0·012
20·0	0·121	0·035	0·152	0·175	0·251	—
25·0	0·149	0·035	0·179	0·201	0·275	—
30·0	0·175	0·035	0·205	0·226	0·298	—
35·0	0·201	0·035	0·229	0·250	0·319	0·321 ± 0·008
40·0	0·225	0·035	0·253	0·272	0·340	—
45·0	0·249	0·035	0·275	0·294	0·359	0·301 ± 0·016
50·0	0·271	0·035	0·297	0·316	0·379	—
55·0	0·293	0·035	0·318	0·336	0·397	—
60·0	0·313	0·035	0·338	0·355	0·415	—
65·0	0·333	0·035	0·357	0·374	0·432	0·164 ± 0·014
70·0	0·352	0·035	0·375	0·392	0·448	—
75·0	0·370	0·035	0·393	0·409	0·464	—
80·0	0·388	0·035	0·409	0·425	0·478	0·112 ± 0·013
85·0	0·405	0·035	0·426	0·441	0·493	—
90·0	0·421	0·035	0·441	0·456	0·506	—
95·0	0·436	0·035	0·456	0·470	0·519	—
100·0	0·450	0·035	0·470	0·484	0·532	—

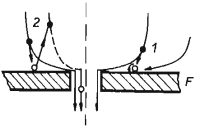

FIG. 4

Schematic Diagram for Probable Explanation of Particle Removing from the Filter Surface

The curves in the Fig. 2 and the Tables I–III show that the semiempirical equation (3) seems to be the most suitable one. Measuring the efficiency curves $E = f(q)$ an important deviation from the theory was found, if q or r were great enough. It has already been reported[11], that the collection efficiency of a fibrous filter was decreasing with increasing q and r, if these both parameters were great ($r > 0·8$ µm,

$q > 100\ \mathrm{cm\ s^{-1}}$). Qualitatively the same effect has been found at the NPF (Fig. 3). The explanation does not seem to be easy. The surface of a NPF is relatively smooth and the latex particles could be removed from this surface, because of hydrodynamical forces or after rebounding of particles (Fig. 4).

The Influence of the Electric Charge of Particles

The possibility that electrostatic effects of NPF might be responsible for the increasing of filter collection efficiency was discussed in the previous communication[1]. The electrostatic effects of aerosol particles, *e.g.* their electrostatic charges might also be responsible for increasing or decreasing of the filter collection efficiency. As mentioned the electric charge of polystyrene latex aerosols produced by atomization-drying process was high enough[10]. Therefore the measurement of filter collection efficiencies was done under and without irradiation with polonium source (Fig. 1). It means the electric charge of latex particles was decreasing by radioactive irradiation. The influence of this electrostatic effect of particles was found the greater the larger were the particle sizes and the smaller were the face filter velocities. A good example of the influence of this effect is shown in Fig. 5.

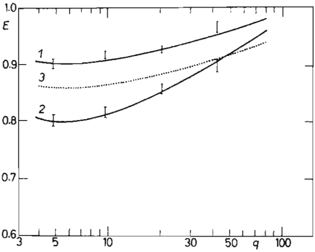

FIG. 5

The Influence of Charged Particles on the Efficiency *E*

R 2·5 μm, *r* 0·357 μm; *1* without irradiation, *2* irradiation with polonium, *3* theoretical curve, flow rate *q* in cm/s.

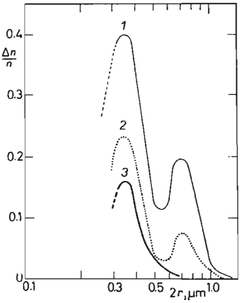

FIG. 6

The Selectivity Curves for a Mixture of Two Types of Latex Particles: *r* 0·179 μm and *r* 0·357 μm

Measuring before (*1*) and after (*2* and *3*) a nuclear pore filter ($R = 1{\cdot}0$ μm). Flow rates: *1* and *2* 3·1, *3* 61·2 cm/s.

As the theory of this effect on a pore filter is not known yet, no quantitative evaluation was possible. But for all measurements of efficiencies this effect was diminished by irradiation of latex aerosol by means of a polonium source.

For practical purposes of aerosol sampling by means of NPF this effect does not seem to be very important, because the efficiency of these filters (*e.g.* NPF with $R = 0{\cdot}5$ μm or $R = 0{\cdot}8$ μm) for non charged particles is mostly higher than 98%.

The Selectivity

It has also been already shown, on the basis of computing[3], that the NPF should have a relatively good selectivity for different particle sizes. If we have a polydisperse aerosol, which is filtering through a NPF with a larger pore size, the penetrating aerosol can have a relatively narrow particle size distribution curve. The optimum conditions can be found by computing.

Using the particle counter Royco and the latex model aerosols, it was possible to find such conditions that the selectivity of a NPF were high. A good example is shown in the Fig. 6.

The Clogging

Time dependence on pressure drop and filter collection efficiency is a function of a clogging process. This process differs in dependence on filtration mechanisms. Some preliminary considerations have already been published[1,2]. If the diffusion in the capillary is the most important separation mechanism, it means, if particle sizes are very small ($r \leq 0{\cdot}1$ μm), the pore walls are coated with particles, effectively decreasing their diameters. When the diameter ultimately decreases to zero at some point, filtration occurs through a porous plug of increasing filter thickness.

The time dependence on pore radius by filtering solid and liquid aerosols was found under some simplified conditions[1]. The pressure drop and also the total collection efficiency are due to increase during this clogging process. This effect is dependent on pore size and particle size, on particle concentration and face filter velocity.

If the diffusion is a negligeable filtration mechanism, *e.g.* the aerosol particles are larger ($r > 0{\cdot}3$ μm), the clogging of pores is going very slowly. In this case the impaction is the most important mechanism, mainly if q is great enough (≈ 50 cm/s). Then the particles are separated mostly on the filter surface (Fig. 7*), and the pores remain open. The time dependence on pressure drop and collection efficiency is negligeable during relatively long filtration time. For example, a NPF with larger pore sizes ($R = 1{\cdot}0$ μm and $R = 2{\cdot}5$ μm) can be used for the preparation of approximately monodisperse aerosols or for a dilution of a monodisperse latex aerosol. This

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* See insert facing p. 2860.

later possibility can be important for practical aerosol measurement by means of particle counter Royco. It has been mentioned that this apparatus can count the number of particles smaller than about 50 cm^{-3} min^{-1}, only. There are many complicated equipments for the dilution, which are often not so accurate. It is possible to use *e.g.* two or more NPF in a series and the particle concentration can be diluted as a function of filter collection efficiency (Fig. 8). We have worked with such a "diluter" and the results were sufficient enough. How long such a "diluter" can be used, shows a simple consideration:

Let us suppose we shall use a NPF ($R = 1{\cdot}0\,\mu\text{m}$, the number of pores $N_p = 3 \,.\, 10^7\,\text{cm}^{-2}$) and we shall filter a latex aerosol ($r = 0{\cdot}357\,\mu\text{m}$, concentration $\approx 10^4\,\text{cm}^{-3}$) with a flow rate 300 cm^3 min^{-1}. Fig. 7 demonstrates that each filter pore can collect as many as about 10 particles without changing its radius. If the filter surface is about 12 cm^2, we can use such a "diluter" more than 15 hours.

Membrane Filters and Silver Membrane Filters

We were also trying to apply this experimental technique for measurement of the filter collection efficiency at classical membrane filters and silver membrane filters. The results have shown that very little agreement is between the counted efficiencies and experimental data (Fig. 9). This disagreement is probably caused by the great

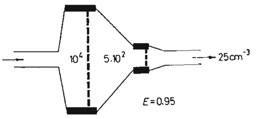

FIG. 8

Schematic Diagram of an NPF "Diluter"

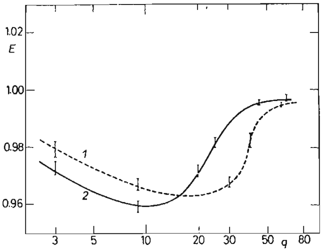

FIG. 9

Measured Dependence of Efficiency E on Flow Rate q (cm/s).

1 Membrane filter NCWP (R 7 µm); *2* silver membrane filter (R 2·5 µm); latex particles (r 0·357 µm). Theoretical efficiencies for q m 20 cm s^{-1}: *1* ~ 0·27, *2* ~ 0·43.

difference between the physical filter model and the real filter structure (Fig. 10*). The "pore diameter" of a membrane filter is more a fiction parameter than a reality. Nevertheless, the efficiency minimum could be found at both these filters.

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* See insert facing p. 2860.