ORIENTAL JOURNAL OF CHEMISTRY
An International Open Free Access, Peer Reviewed Research Journal
www.orientjchem.org
Est. 1984

ISSN: 0970-020 X
CODEN: OJCHEG
2015, Vol. 31, (Spl Edn):
Month : Oct.
Pg. 43-51

Analysis of Applicability of Method of Aerosol Particles Permeability in Porous Materials for Incoming Control of Filtering Elements with Nanostructured Membranes used for Liquids Purification

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(Received: July 15, 2015; Accepted: October 03, 2015)

http://dx.doi.org/10.13005/ojc/31.Special-Issue1.05

ABSTRACT

An incomplete extraction of uranyl ammonium tricarbonate (UATC) from mother solutions is one of the problems concerning liquid radwaste accumulation. Implementation of processes and devices allowing for complete extraction of residual UATC from mother solutions would make it possible to significantly decrease expenses by the plant for radwaste utilization and environmental impact, and also to return UATC to the fuel production process. Update of mother solution handling technology is attributed in particular to the development and implementation of liquids purification systems using membranes. Successful introduction of membrane purification systems to the plants is primarily related to the quality of the membranes and the possibility of their total (100%) quality control. This paper represents the analysis of the possibility of using method based on aerosol particles permeability in porous materials as total incoming control of filtering elements with nanostructured membranes designed for removal of mechanical impurities from liquids[1, 2]. Analysis of this method was made in the following steps:

´ Measurement of the total number of particles and initial spectrum of aerosol particles;

´ Measurement of the total number of particles and spectra of aerosol particles transmission through porous base coats produced in various agglomeration modes;

´ Measurement of the total number of particles and spectra of aerosol particles transmission through the filtering elements with Ti – nano-membrane;

´ Calculation of coefficient of aerosol particles capture by Ti – membrane;

´ Evaluation of filtering elements quality.

Studies were carried out within the framework of Agreement14.625.21.0001 of 21.08.2014 between the Ministry of Education and Science of the Russian Federation and JSC "SSC RF "IPPE" (unique identifier of the project is RFMEF162514X0001).

Keywords:, uranyl ammonium tricarbonate (UATC), mother solutions, purification, nanostructured membranes, aerosol particles, filtering element, the porous substrate.

INTRODUCTION

To date, the share of nuclear power in the global energy production is steadily growing, and this growth rate strongly depends on the rate of increase of uranium mining and fuel production. Now the attention is being increasingly focused on the improvement of energy performance and safety of industrial production of uranium compounds and further uranium fuel fabrication.

In addition, there is a problem concerning accumulation and storage of liquid radwaste, which is primarily caused by incomplete extraction of uranyl ammonium tricarbonate (UATC) from mother solutions. Implementation of processes and devices allowing for complete extraction of residual UATC from mother solutions would make it possible to significantly decrease expenses by the plant for radwaste utilization and environmental impact, and also to return UATC to the fuel production process[3, 4].

Mother solution handling technique can be improved using membrane technology of liquids purification developed by the JSC “SSC RF – IPPE”. This technology entails the use of filtering elements with nanostructured metallic or ceramic membranes (made of titanium, zirconium, chromium etc. or their oxides, carbides and nitrides)[1, 2]. Hydrodynamics of liquid flow purified by the membrane filtering depends on the structure and composition of porous composite pair: “nanostructured membrane – base coat”. The lack of the total control of the composite pair may cause decrease of product quality and, as a result, the abrupt decrease of available period of stable filtration of mother solutions, decrease of filtering system lifetime or rapid rupture of porous materials. Complete quality evaluation of composite pair and nanostructured membrane by the standard methods (microscopic investigation, hydrodynamic performance check, structural analysis etc.) is time-consuming and cost-intensive procedure causing considerable increase of filtering material cost. In this view, it is viable to consider the possibility of using evaluation of spectra of aerosol particles penetrating through filtering element as a method of control of membrane filtering elements with required working characteristics. Results of evaluation of aerosol particles comparative permeability can also be used for the incoming control of membrane filtering elements intended for removal of mechanical impurities from liquids[5-7].

Analysis of the possibility of using method based on aerosol particles permeability in porous materials for total incoming control of filtering elements with nanostructured membranes designed for removal of mechanical impurities from liquids was made in the following steps:

- Measurement of the total number of particles and initial spectrum of aerosol particles;
- Measurement of the total number of particles and spectra of aerosol particles transmission through porous base coats fabricated in various agglomeration modes;
- Measurement of the total number of particles and spectra of aerosol particles transmission through the filtering elements with Ti – nanomembrane;
- Calculation of coefficient of aerosol particles capture by Ti – membrane;
- Evaluation of quality of filtering elements with Ti – nanomembrane[8].

METHOD

As a rule, at the stages of development and startup of production of filtering elements for liquids purification, selective control of their quality is performed using hydraulic characteristic $Q = f(\Delta P)$, where $\Delta P = P_1 – P_2$, P_1 – pressure at the membrane filter inlet, P_2 – pressure at the membrane filter outlet, Q – liquid flow rate in the filter[3]. In order to eliminate the effect of impurities layer formed in the course of filtering on hydraulic characteristic $Q = f(\Delta P)$ to be determined, liquid containing no impurities (distillate) is commonly used. However the use of this method for the total quality control is complicated by the design features of test equipment and duration of drying of tested filtering elements.

Advantages of method of incoming control of filtering elements using parameter of aerosol particles penetrability are fast response, accuracy, capability of gaining significant amount of statistical data, reliability of results etc.

Work on evaluation of aerosol particles penetrability through the filtering elements with nanostructured membrane was carried out taking into account the following technological characteristics:

- Modes of porous base coats fabrication[9,10].
- Modes of formation of plasma-chemical membranes on porous polymer base coat.

The use of low pressure ultrahigh-molecule polyethylene (LP UHMWPE) powder was required for base coat in order to meet the following requirements:

- Min grain-size dispersion of the composition to assure max uniformity of through pores;
- Min adhesion to the impurities in purified liquid;
- High softening temperature of agglomerated porous material.

Taking into account these requirements GUR 4120 powder (Germany) was used, its technical characteristics being as follows:

- Molecular mass: within the range of 1×10^6 – 8×10^6 g/mole;
- Average particle size: 50 – 180 µm;
- Pour density: 380 – 480 g/L;
- ash content in terms of titanium: ~ 3ppm.

Formation of plasma-chemical Ti-membrane on LP UHMWPE porous polymer base coat was carried out in UNM vacuum facility (Fig. 1).

The volume of filtering elements for the analysis of aerosol particles penetrability spectra in this study was only determined by the parameters of porous base coats fabrication process. The mode of formation of plasma-chemical Ti-membrane on the porous base coat is settled. Parameters of control of the process of Ti-membrane application on porous polymer base coats are shown in mnemonic diagram (Fig. 2).

Since all Ti-membranes were applied by the same technology, so the parameters of porous base coat fabrication process were the key determinants of aerosol particles permeability spectrum. The main parameters of porous base coat fabrication process are as follows: time of vibropacking of polymer powder in mold, and temperature and duration of agglomeration process[11, 12]. The total number of filtering elements with nanostructured membranes and porous base coats fabricated in various modes presented for the analysis of aerosol particles permeability spectra was 34.

The total number of particles and their spectra were measured using equipment shown in Fig. 3.

In Figure 4 presented is aerosol particles distribution spectrum typical for atmospheric air. Concentration of aerosol particles within the range of 27 nm – 947 nm (C_0) registered by N-WCPC counter (version 3788 Nano) is $3.4\cdot10^7$ particles/cm^3.

The basic criterion for evaluation of filtering elements quality using aerosol particles permeability method is integral permeability coefficient determined as the percentage ratio of the number of aerosol particles penetrated through the filtering element to the total number of aerosol particles in the air $k = (C_i/C_0)\cdot100$, expressed in percent. Another criteria of filtering elements incoming control is integral coefficient of aerosol particles capture by nanostructured membrane, determined as percentage ratio of the number of aerosol particles penetrated through the filtering element with applied nanostructured membrane to the number of aerosol particles penetrated through the corresponding porous base coat, $k' = (1 - C_{men}/C_{coat})\times100\%$ expressed in percent. Comparison of these coefficients to the function of impurities capture by the filtering element in the course of liquids purification would make it possible to adequately determine whether some failure occurred in the filtering elements fabrication process and, if so, at what stage this failure occurred[13-15].

RESULTS AND DISCUSSION

Evaluation of porous base coat quality

Table 1 gives the numbers of aerosol particles penetrated through the filtering element and corresponding porous base coat. Concentration of aerosol particles in the initial spectrum is $C_0 = 3.4\cdot10^7$ particles/cm^3.

Table 1: Integral permeability coefficients for porous base (K_{coats}) and filtering elements (K_{mem})

No.	C_{coat}, particles /cm³	C_{mem}, particles /cm³	κ_{coat}, $\cdot 10^{-3}$, %	κ_{mem}, $\cdot 10^{-3}$, %	No.	C_{coat}, particles /cm³	C_{mem}, particles /cm³	κ_{coat}, $\cdot 10^{-3}$, %	κ_{mem}, $\cdot 10^{-3}$, %
1	1280	265	3,76	0,78	2	2294	845	6,75	2,49
3	1350	303	3,97	0,89	4	1261	658	3,71	1,94
5	1181	360	3,47	1,06	6	1751	986	5,15	2,90
7	957	333	2,81	0,98	8	683	368	2,01	1,08
9	875	620	2,57	1,82	10	2136	445	6,28	1,31
11	1349	288	3,97	0,85	12	3127	178	9,20	0,52
13	1166	367	3,43	1,08	14	936	312	2,75	0,92
15	1050	301	3,09	0,89	16	988	389	2,91	1,14
17	905	613	2,66	1,80	18	652	106	1,92	0,31
19	3809	779	11,20	2,29	20	868	184	2,55	0,54
21	745	136	2,19	0,40	22	1008	270	2,96	0,79
23	385	210	1,13	0,62	24	1904	763	5,60	2,24
25	1034	301	3,04	0,89	26	800	125	2,35	0,37
27	832	112	2,45	0,33	28	3797	631	11,17	1,86
29	1264	213	3,72	0,63	30	1365	455	4,01	1,34
31	1048	280	3,08	0,82	32	1359	594	4,00	1,75
33	1408	1068	4,14	3,14	34	1563	1044	4,60	3.07

$C_{init.}$=3.4·10⁷ particles/cm³

Table 2: Integral coefficients of aerosol particles capture by nanostructured membrane (κ')

No.	C_{coat}, particles/cm³	C_{mem}, particles/cm³	κ', %	No.	C_{coat}, particles/cm³	C_{mem}, particles/cm³	κ', %
1	1280	265	79,3	2	2294	845	63,1
3	1350	303	77,6	4	1261	658	47,8
5	1181	360	69,5	6	1751	986	43,7
7	957	333	65,2	8	683	368	46,1
9	875	620	29,1	10	2136	445	79,2
11	1349	288	78,7	12	3127	178	94,3
13	1166	367	68,5	14	936	312	66,7
15	1050	301	71,3	16	988	389	60,6
17	905	613	32,3	18	652	106	83,7
19	3809	779	79,5	20	868	184	78,9
21	745	136	81,8	22	1008	270	73,3
23	385	210	45,5	24	1904	763	59,9
25	1034	301	70,6	26	800	125	84,4
27	832	112	86,5	28	3797	631	83,4
29	1264	213	83,2	30	1365	455	66,7
31	1048	280	73,3	32	1359	594	56,3
33	1408	1068	24,2	34	1563	1044	33,2

$C_{init.}$=3.4·10⁷ particles/cm³

Table 3: Coefficients of air purification (κ_{air}) and iron removal from water by filtering elements (κ_{Fe})

No. of specimen	κ_{air}, %	κ_{Fe}, %
33	99,99686	2,2
32	99,99825	49
26	99,99963	54,1

It can be seen from Table 1 that max specific number of aerosol particles penetrated through the porous base coat corresponds No. 19 specimen and is equal to C_{coat} = 3809 particles/cm^3, while min specific number corresponding to No. 23 specimen is equal to 385 particles/cm^3. Integral permeability coefficients of porous base coats of No. 19 and No. 23 specimens are equal, respectively, to $\kappa_{coat} = (3809/3,4\cdot10^7)\cdot100 = 11,20\cdot10^{-3}$ % and $\kappa_{coat} = (385/3,4\cdot10^7)\cdot100 = 1,13\cdot10^{-3}$ %. The difference between max and min values of integral permeability coefficients of porous base coats are of the same order and, hence, one can conclude that fabrication mode of No. 23 base coat is most close to optimum. The least integral permeability coefficient of filtering element is $\kappa_{mem} = (106/3,4\cdot10^7)\cdot100 = 0,31\cdot10^{-3}$ % (No. 18 specimen).

In order to find the most preferable modes of porous base coat fabrication, taking into account the above considerations, the average min value of integral permeability coefficients for porous base coats and filtering elements can be calculated, its dispersion being within 40 %[16].

Average min value of integral permeability coefficient for porous base coat is $1,85\cdot10^{-3}$ %. Taking into account the above dispersion (40 %) Nos. 23, 18, 8, 21, 26, 27, 20 and 9 specimens correspond to this value. For filtering element with Ti - membrane, the average min value of integral permeability coefficient is $0,47\cdot10^{-3}$ %, to which Nos. 18, 27, 26, 21, 12, 20, 23 and 29 specimens correspond within stated 40 % range: 6 out of 8 specimens, i.e. 75 % were fabricated in the similar modes. For Nos. 23, 27, 18, 20, 21 and 26 specimens, dispersion of values of polymer powder vibropacking time in the mold, and temperature and duration of agglomeration process is within 5%.

Evaluation of nanostructured membrane quality

In Fig. 5-6 presented are the most typical spectra of distribution of aerosol particles penetrated through porous base coat and filtering element.

Analysis of spectra of distribution of aerosol particles penetrated through porous base coat and filtering element shows that the degree of aerosol particles capture by the nanostructured membranes applied in the similar mode varies within the wide range depending on the modes of porous base coat fabrication. This can be evaluated using integral coefficient of aerosol particles capture by nanostructured membrane (κ'). In Table 2 presented are calculated κ' values

As follows from Table 2, values of integral coefficient of aerosol particles capture by nanostructured membrane are between κ'_{min} = 24,2 % (No. 33 specimen) and κ'_{max} = 94,3 % (No. 12 specimen). For specimens 23, 27, 18, 20, 21 and 26 selected on the basis of integral permeability, coefficient κ' value is ranging from 45,5 % to 86,5 %. Only one out of six specimens under consideration provides κ' value beyond 10% deviation from the averaged value.

Proceeding from stated filtration rates by porous base coat (~ 3 µm) and filtering element (~ 0,2 µm), ~ 93,3 % average value of nanostructured membrane contribution to the effectiveness of liquids purification is obtained by calculation. Therefore, method of filtering elements quality evaluation on the basis of aerosol particles permeability requires integral capture coefficient κ' to be equal to at least 93,3 %. Out of all fabricated specimens, only No. 12 meets this criterion. Hence, No.12 specimen porous base coat fabrication mode is most preferable for the stated mode of nanostructured membrane application.

Comparison of capture coefficients (water and air)

In Table 3, presented are min, medium and max values of coefficient of air purification by the filtering element $\kappa_{air} = (100 - \kappa_{mem})$ and coefficients of iron removal from water (κ_{Fe}) obtained experimentally for the corresponding specimens.

Fig. 1: UNM vacuum facility for application of Ti-membranes:
a – control rack, b – working chamber with vacuum system

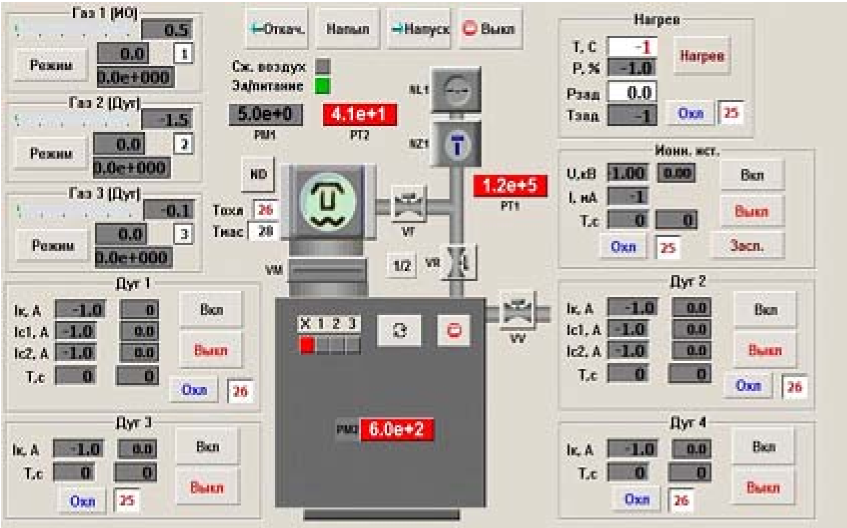

Fig. 2. Mnemonic diagram of control of parameters of
Ti-membrane application on porous polymer base coat

Fig. 3: Equipment for determining aerosol particles permeability through the filtering element (1 – porous base coat or filtering element with Ti-nano-membrane, 2 – aspirator UPRV 1-20, 3-electrostatic sorting machine, version 3080, 4 – aerosol particles counter N-WCPC, version 3788 Nano, 5 – computer with measurement data processing software)

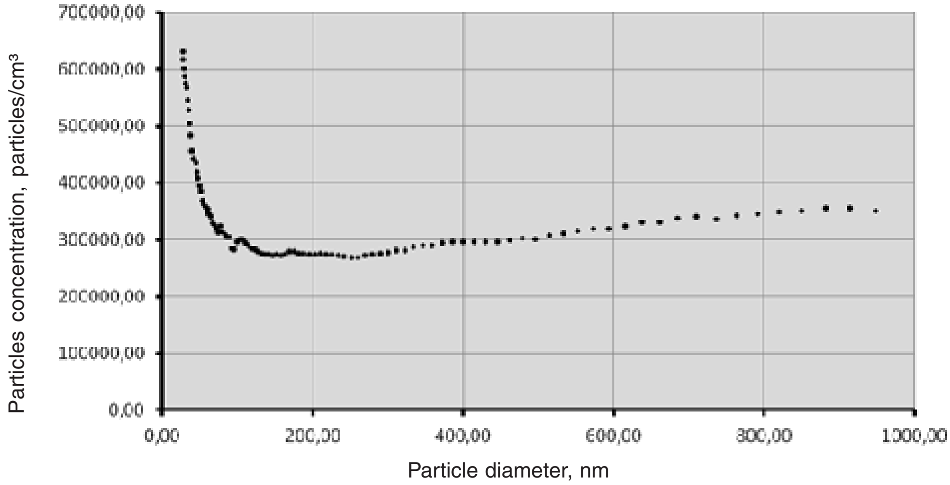

Fig. 4: Spectrum of aerosol particles distribution in the air

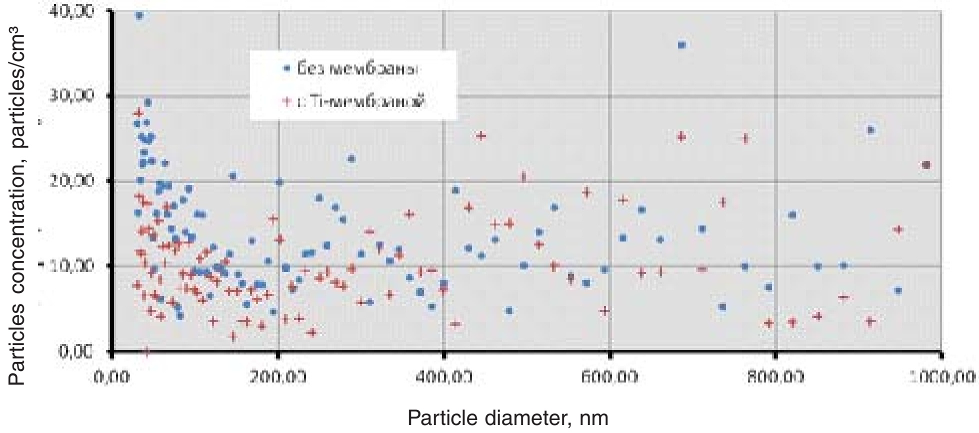

Fig. 5: Spectrum of aerosol particles distribution (No. 33 specimen)

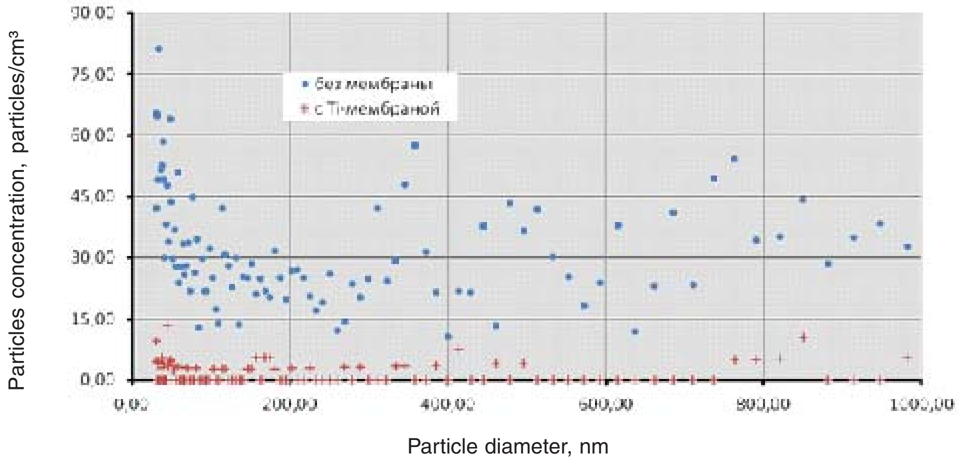

Fig. 6: Spectrum of aerosol particles distribution (No. 12 specimen)

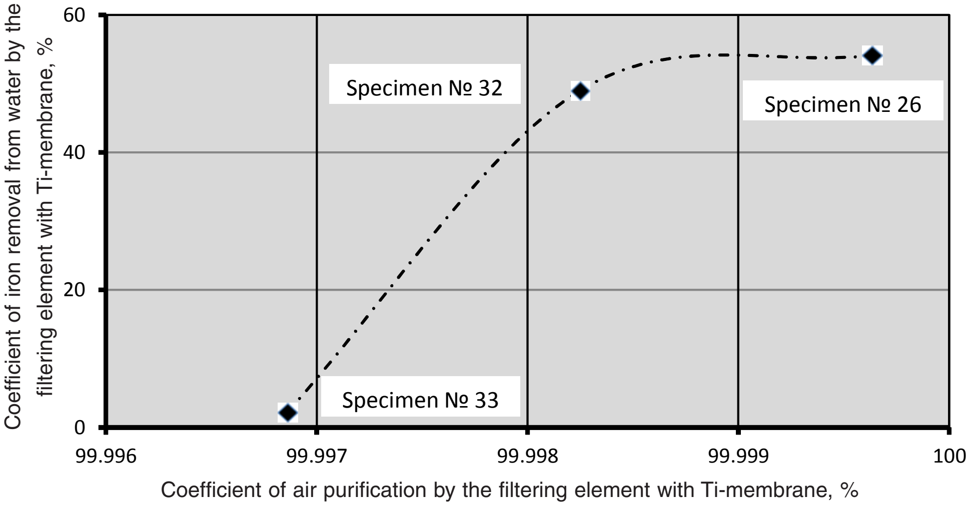

Fig. 7: Relationship between between the coefficients of air purification and those of iron removal from water

Fig. 7 shows the relationship between the coefficients of air purification and those of iron removal from water by the filtering elements. As it follows from Table 3 and Figure 7, there is correlation dependence between κ_{air} and κ_{Fe} coefficients. Therefore, with a knowledge of air purification coefficient value, one can estimate with a reasonable degree of accuracy the quality of filtering elements used for liquids purification, as well as detect filtering element fabrication stage in which fabrication technology failure occurred.

On the basis of analysis of obtained results the following technological procedures required for the further improvement of membrane filtering elements should be noticed:

´ Optimization of modes of nanostructured membrane application on the porous base coat in order to increase integral coefficient of aerosol particles capture by the membrane filtering element up to at least 93,3 % value;

´ Optimization of porous base coat fabrication technology within determined 40 % range of this technology parameters.

CONCLUSION

1. Method based on aerosol particles penetrability in porous materials can be adequately applied to incoming control of filtering elements with nanostructured membranes designed for liquids purification
2. Method of evaluation of membrane filtering element quality by measuring aerosol particles penetrability through this element includes recording and correction of all features of technological cycle of production of filtering element with nanostructured membrane to be used for purification of liquids including those containing uranium.
3. Ranges of key parameters of porous base coat fabrication technology (time of polymer powder vibropacking in the mold, and temperature and duration of agglomeration procedure) were determined with dispersion not exceeding 5 %.
4. Incoming quality control of membrane filtering element on the basis of aerosol particles capture coefficient of nanostructured membrane revealed the necessity for optimization of technology of nanostructured membrane formation on porous base coat.
5. Method of estimation of aerosol particles penetrability in porous materials as incoming control of filtering elements with nanostructured membranes makes it possible to evaluate filtering element quality and also detect any failure of filtering element fabrication technology and the stage of fabrication, in which this failure occurred.

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