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Sorption materials for indoor environment cleaning from microorganisms

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Abstract. The article touches upon the problem of working area air pollution by pathogenic microorganisms. The problem's solution requires increased efficiency of filtration materials. Using thermally expanded graphite and Cribrol® polymer composite material, we analyzed air purification quality of multifunctional rooms in comparison with traditional activated carbon. The filtration materials properties were studied using a set of analytical methods. The air was pumped through tested materials in a volume of 500–2000 liters with the use of PU-1B sampling device depositing microorganisms on the nutrient medium. We showed that activated graphite and Cribrol® are effective in cleaning the air from bacteria (cell sizes do not exceed 1 micron), as well as larger microorganisms (from 3 microns or more). Activated graphite completely trapped microorganisms in all test variants. The filtration capacity of the new materials turned out to be higher than that of traditional activated carbon, which indicates the prospects for their further research and practical application.

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1. Introduction

The spread of microorganisms through microbial-dust aerosols can have a noticeable effect on the human environment. The particular attention in this regard is paid to the occurrence of the pathogenic or conditionally pathogenic microorganisms (macroscopic fungi – micromycetes and bacteria) in the indoor environment. This problem has been attracting the attention of specialists all over the world for the past decades [1–8]. Being in the premises with a high content of spores in the air for a long time, a person may experience deterioration of well-being, allergic reactions, etc. The contact with human respiratory organs is explained by the small size of the spores of many mold fungi, which allows them to reach even the alveoli of the lungs. Typically, the size of the fungal spores ranges from 2 to 10 microns, while the size of the bacterial cells transported by air is less than 1 micron. Most often, the accumulation of the fungi in the air of premises for various purposes is associated with the open growth and sporulation of colonies of micromycetes on building materials in areas of increased moisture [9]. The studies of air microbiota in Russia took place in residential and working buildings, museums, libraries, the subway, at polar stations [7, 10–13]. According to WHO recommendations [14], the content of indoor spores should not exceed 500 spores per 1 cubic meter. In many cases, these values are significantly higher, which requires special measures to reduce the number of the micromycetes in the indoor air. The first thing that is required in this regard is the regular monitoring of the indoor air environment. Such control is needed not only in residential and working premises, but also in engineering structures, such as the subway, where there are favorable conditions for accumulation and spread of microorganisms. For sampling and analysis of air samples sampling devices of various configurations are used. The typical samplers (aspirators) suck air containing various particles onto the collecting surface (nutrient medium or filters). The collection efficiency of such devices depends on a combination of factors, including the intake air speed, the shape and diameter of the suction nozzle, the distance between the nozzle and the collection surface, the diameter of the surface pores and the collection time, as well as the inertial properties of the microorganisms associated with their size and density. The filter samplers pump air through filters with different pore sizes. These samplers are most commonly used for collecting aerosols. In Russia, the certified device for sampling the air environment is the sampling device PU-1B (Manufacturer – "Khimko", Moscow). This device has been vastly tested, including in conditions of high latitudes (Arctic and Antarctic), where it has shown its sufficiently high efficiency.

The second important aspect is air purification. The improvement of indoor air quality is most often achieved through the use of air purifiers. The use of filters in such installations is the most commonly used approach to solving this problem. The filtration efficiency depends on the material used for this purpose. These studies are currently focused on finding effective, environmentally friendly and technologically accessible filters.

The purpose of the work is to evaluate the effectiveness of the new filtration materials for multipurpose cleaning of the air environment from microorganisms living indoors.

2. Methods

Analysis of the effectiveness of the known filtration materials. The main requirement for the sorbents for air and water purification is their high sorption capacity. The most important characteristic of the sorbents is porosity, which characterizes the degree of the surface development, as well as specific surface area (SD, m^2/g). The greater the specific surface value of the sorption material, the greater the number of active pores, presented in the volume of its structure, as a result of which the sorption capacity of the material increases proportionally [15–18].

All the methods (technologies) of increasing the surface area of materials are directed primarily to the development of the existing micropores and the formation of new ones by removing individual microstructural elements.

The various types of activated carbon have become the most widely used in the world practice as materials for indoor air sorption purification. The best samples of activated carbon are obtained by the activation method. The essence of the activation process consists in the opening of the pores in the carbon material in a closed state. This is achieved thermally: the material is pre-impregnated with a solution of zinc chloride, potassium carbonate and either heated to 400–600 °C without air access, or with superheated steam at a temperature of 700–900 °C under strictly controlled conditions. And, if conventional activated carbons have a specific surface area of 20–70 m^2/g, then modified ones already have 200–850 m^2/g.

The indirect and non-obvious analogues of carbon materials (UM) are sorbents based on dispersed natural flake graphite. This type of materials has a low specific surface area (up to 5 m^2/g), but is well regenerated, technologically advanced, inert to all media and numerous aggressive compounds [19].

In principle, it is possible to significantly increase the specific surface area of the UM by intercalating graphite-like blocks, followed by their stratification. Thermally expanded graphite (TEG) is obtained specifically by this method: by introducing acid anions into dispersed natural graphite, followed by heating. The natural UM processed in this way have a specific surface area already up to 80–200 m^2/g (according to some literature sources up to 600–m^2/g). Thus, TEG is a very promising adsorbent for purification of polluted water and gas media.

Thermally expanded graphite (TEG) is widely used for the manufacture of graphite seals, and technology for its production has been known since the 1960s. Some scientists and a number of enterprises [20] were trying to use the TEG properties for water purification, but for a number of reasons their attempts were unsuccessful. The resulting "technical" TEG cannot be used in filters, due to low quality and a large amount of impurities, as well as unstable results in the quality of purification.

To obtain their own carbon material (Fig. 1) based on thermally expanded graphite, as well as to identify its applicability as a sorption material for air and water purification, the authors conducted appropriate laboratory tests with varying synthesis parameters and parallel analysis of the properties and quality of the obtained products. The obtained material properties were controlled by the differential scanning calorimetry and X-ray phase analysis methods. Bulk density was measured according to the VNIIEM technique standard (OST 16-0689.031-74).

Figure 1. Carbonic sorbent based on TEG.

The developed technology for producing TEG [21], based on the short-term heat treatment of a graphite composition in the presence of the developed inhibitor, makes it possible to obtain a carbonic sorbent (activated graphite) based on TEG with high sorption characteristics (specific surface area Sud = 2000 m^2/g) many times higher than all known filter materials (Fig. 2). Thus, the specific surface area of absorbent carbon is Sud = 450–750 m^2/g. The chemical purity and environmental safety of the product allow it to be used for water treatment needs.

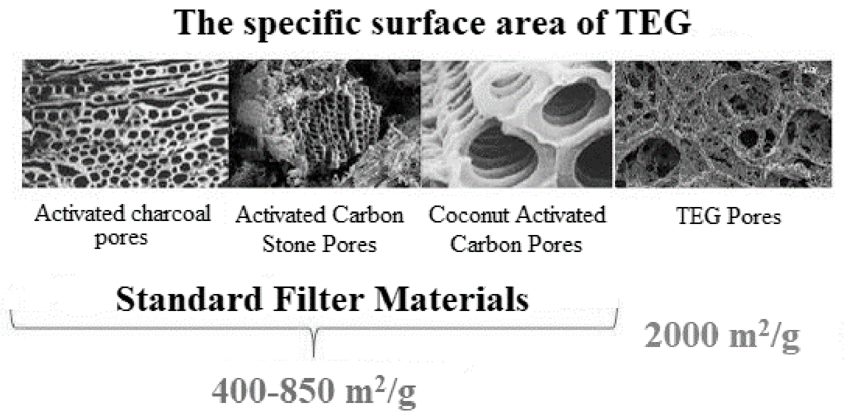
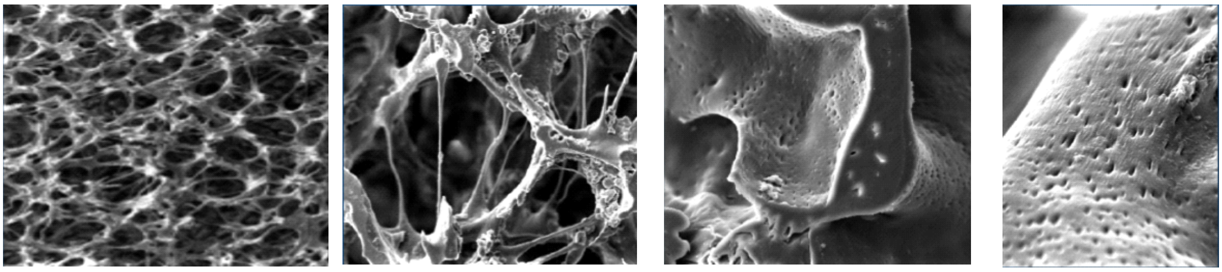


Figure 2. Comparison of the TEG specific surface area with Absorbent carbon.

When the thermally expanded graphite is compacted in the filter, a strong, porous structure is created, similar in its properties to a membrane. By adjusting the compaction degree, it is possible to achieve different porosity values of the filter load and set the required material throughput for certain polluting components, depending on the required purification degree.

The Cribrol® polymer composite is also a competitive material for the cleaning of gas-air mixtures. Its structure is formed according to the ratio of through and non-through pores of different diameters, uniformly distributed throughout the volume of the filtering partition, set in the polymerization process (Fig. 3) [22]. The technological process makes it possible to obtain the porous percolating material with a given pore system: micropores, mesopores and macropores. This ensures versatility in the production of materials with different performance characteristics for solving various tasks.



Pore size: **10–15 microns** | **Pore size:** **6–10 microns** | **Pore size:** **1–6 microns** | **The number of micropores in the walls of macropores**

Figure 3. Pore combination in Cribrol® materials.

For these meterials, there is no concept of surface area, since they are bulk, porous materials. One filter cartridge Cribrol® combines three technological functions – sorption, coalescence and inertial separation, which proceed sequentially in a stream of liquids and gases passing through a porous partition (Fig. 4).

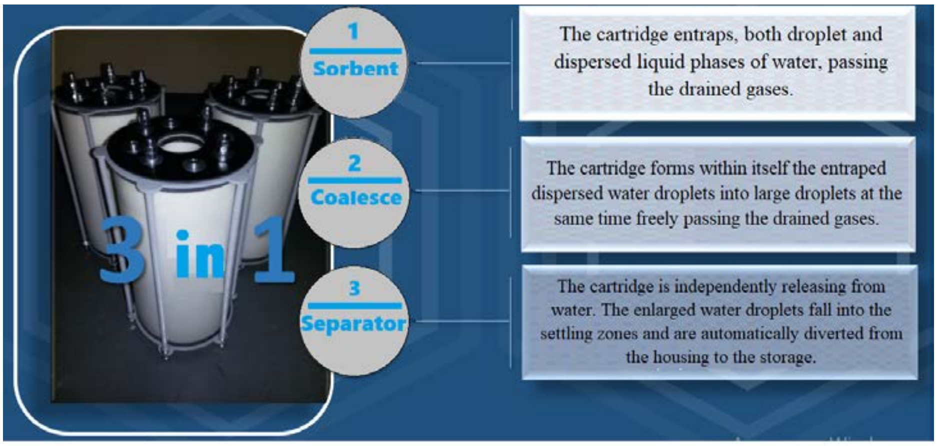


Figure 4. The principle of Cribrol® cartridges operation.

These unique properties of Cribrol® materials determine their effective use for solving various technological problems associated with the separation of dispersed phases of liquids contained in gaseous media, which, in particular, can transport infectious agents – bacteria, fungi and viruses [23].

Two rooms, differing in their purpose and conditions, were selected as objects for evaluating the effectiveness of the new sorption materials:

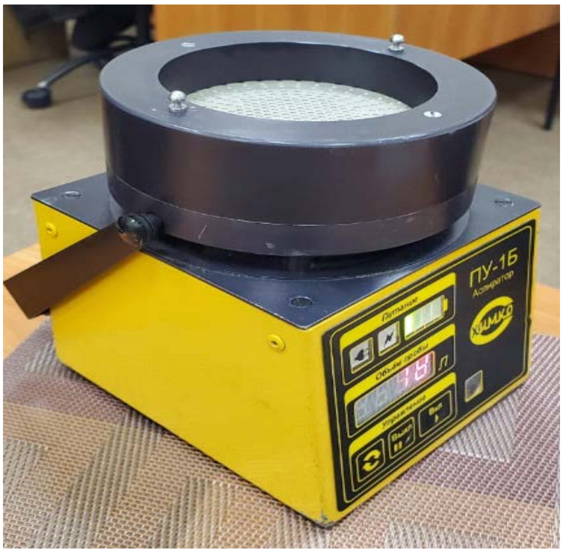
1) an office space, where people stay daily during working hours (without external signs of biological damage to materials and sufficient ventilation);

2) a library space, where books and archival documents are stored for a long time with weak ventilation.

The microbiological sampling of the air environment in the office and in the library spaces was carried out using a PU-1B aspirator (a Russian-certified sampling device for taking air samples), through which air was pumped in a volume of 500–2000 liters, depositing microorganisms in Petri dishes on the nutrient medium. When air was pumping, filtration materials were placed in its path. The control was conducted with the air samples without the use of filtration materials. When taking a sample, the device was positioned at a level of about 1 m from the surface. Each sample was taken three times for Czapek medium (to determine the number of fungi) and FMH (fermented meat hydrolysate) – to determine the total microbial number of organotrophic bacteria).

As tested substances, new filtration materials were used: Cribrol®, thermally expanded graphite (EG) and activated carbon (AC).

For the testing, the material was placed in the over-frame space of the sampling device PU-1B in such a way that the material covered the hopper above the nozzles of the device and air could be pumped through it (Fig. 5).

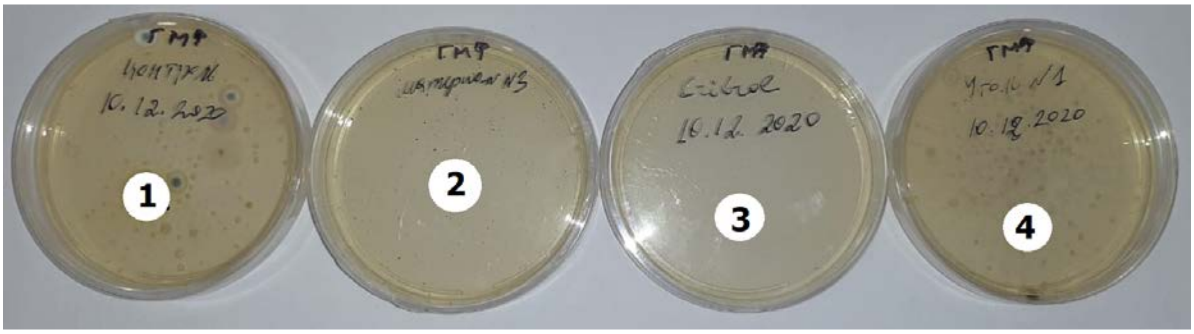


a b

c d

Figure 5. The experimental installation for the testing of new filtration compositions: a) control (sampling device with an unfilled hopper), b) thermally expanded graphite (filled hopper), c) Cribrol®, d) activated carbon

After pumping air in rooms for various purposes through the tested materials, Petri dishes were removed from the PU-1B device, maintaining sterility, and incubated in laboratory conditions at a temperature of +25 °C. After 10 days, the number of grown colonies was calculated (Fig. 6) and colony-forming units (CFU) were recalculated per 1 m^3 of the air (in accordance with the PU-1B device operating manual).



a

b

Figure 6. An example of counting colonies of bacteria and microscopic fungi after the sampling in different figure versions of the experiment: a) cleaning from bacteria, b) cleaning from fungi 1 – Control (120 CFU); 2 – Thermally expanded graphite (0 CFU); 3 – Cribrol® (2 CFU); 4 – Activated carbon (50 CFU); 5 – Control (17 CFU); 6 – Thermally expanded graphite (0 CFU); 7 – Cribrol® (1 CFU); 8 – Activated carbon (2 CFU).

3. Results and Discussion

The results of the experiments (Table 1) showed high efficiency of the used materials for the air purification in rooms for various purposes. The highest efficiency was demonstrated by the EG composition, which completely detained microorganisms (fungi and bacteria) in all test variants. The Cribrol® composition in various modifications also showed significant effectiveness (a decrease in the number of cells by an order of magnitude and higher in comparison with the control). The AC composition showed high efficiency against fungi (micromycetes), while the effect of air purification from bacteria was lower. It must be noted, that the number of bacteria in the air of the office and library was several times higher than of micromycetes in control samples.

Table 1. Determination of the number of bacteria and fungi in rooms for various purposes with different test variants of sorption materials

Sample No.	The filter material	The filter material mass	Nutrient medium	Air volume	The number of the grown colonies	CFU per 1 cubic meter of air
The office space						
1	Activated carbon		Czapek	500	2	4
2	Activated carbon		FMH	500	50	110
3	Cribrol®		Czapek	500	1	2
4	Cribrol®		FMH	500	2	4
5	EG		Czapek	500	0	0
6	EG		FMH	500	0	0
7	Control		Czapek	500	17	34
8	Control		FMH	500	120	290
9	Cribrol	10,570	Czapek	500	0	0
10	Cribrol	9,910	FMH	500	3	6
11	Cribrol	9,010	Czapek	1000	1	1
12	Cribrol	10,670	FMH	1000	0	0
13	Cribrol	9,210	Czapek	2000	0	0
14	Cribrol	9,310	FMH	2000	10	5
15	Cribrol®+H20	16,300	Czapek	1000	2	2
16	Cribrol®+H20	22,870	FMH	1000	0	0
17	Control		Czapek	1000	31	35
18	Control		FMH	1000	54	58
19	Control		Czapek	2000	26	14
20	Control		FMH	2000	128	79
21	EG	2,280	Czapek	1000	0	0
22	EG	2,650	FMH	1000	0	0
23	EG	2,570	Czapek	2000	0	0
24	EG	2,290	FMH	2000	0	0

Sample No.	The filter material	The filter material mass	Nutrient medium	Air volume	The number of the grown colonies	CFU per 1 cubic meter of air
The library space						
25	Control		FMH	1000	42	45
26	Control		FMH	2000	52	28
27	Control		Czapek	1000	6	6
28	Control		Czapek	2000	10	5
29	EG	3,030	FMH	1000	0	0
30	EG	2,810	FMH	2000	0	0
31	EG	2,400	Czapek	1000	0	0
32	EG	2,390	Czapek	2000	0	0
33	EG	37,640	FMH	1000	100	100
34	EG	37,870	Czapek	1000	2	2

Thus, all the compounds included in the tests demonstrated different effectiveness in cleaning the air from microorganisms. The tested materials purified the air from small bacterial cells (not exceeding 1 micron in size), as well as larger micromycete spores (from 3 microns or more). Whereas in all control variants, dozens of colonies of micromycetes (from the genera Cladosporium, Penicillium, Aspergillus) and organotrophic bacteria colonies germinated on nutrient media. Some of the identified microorganisms were typical for the indoor environment of buildings with different microclimates [24–27]. They are also commonly found on various building materials and can spread through the air [28, 29]. In variants with sorption materials, either single colonies were noted, or they were absent altogether. If, as a comparison of the air purification quality indicators of the working space, we could draw an analogy between the efficiency of activated carbon and activated graphite, then the result directly depends on the value of the specific surface area: the higher it is, the more microorganisms can be sorbed from the air. There is no doubt that EG has an advantage here.

Since the studied Cribrol® material has a mechanism fundamentally different from the sorption processes occurring in activated graphite and carbon, a direct comparison with its analogues is not entirely correct. However, the obtained efficiency in air purification makes it possible to consider it, as well as EG, as materials analogous to traditional activated carbons. It is also worth considering that according to the manufacturer's statements, the greatest efficiency of this material is observed during the separation of aerosols, since Cribrol® works better in the "wetted" state.

In addition, despite the fact that during the tests the difference in cleaning efficiency was observed after 2000 liters of filtered air, the question of the compared materials resource remains open.

These experiments, as well as the Cribrol® material effectiveness studies in the "wetted" state, are planned to become a part of the subsequent studies.

4. Conclusion

1. Activated carbon (AC) widely used in world practice has low efficiency as a sorption material for air purification from microorganisms.

2. The best effect is shown by filtration materials Cribrol® and thermally expanded graphite (EG). They clean the air in rooms for various purposes from small bacterial cells, as well as large micromycetes.

3. The air purification efficiency obtained during the tests allows considering Cribrol®, as well as EG, as analogues to traditional activated carbons.

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