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Volatile organic compound emissions to the atmosphere and their reduction

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Every day thousands of tons of dangerous chemical pollutants are emitted to the environment and exert a negative effect on the ecosystems. The greenhouse effect is one of the dangerous effects on ecosystems. The increase of carbon dioxide, CH_4, N_2O, halogen-hydrocarbons and other volatile organic compounds (VOC) (except CH_4) has an influence on the greenhouse effect. The reduction of VOC emissions to the atmosphere is very complicated. VOC emissions the period 1996–2003 increased from 7.6 to 28.4 t/year. Since 2004 VOC emissions have been slowly decreasing. VOCs get into the atmosphere when different processes occur in the soil, water and in atmosphere. The biggest emission of VOC to the environment is related to atmospheric pollution. This article offers an analysis of the possibilities to reduce the emissions of popular VOCs such as acetone, toluene and butylacetate. These compounds are used in industry as solvents. Oil industry, chemical industry, small burning equipment, food industry, wastes and farms are also emitters of VOC. According to the European Parliament Resolution adopted on 16 November 2005, the quantity of the greenhouse gas must be reduced significantly (by 30 percent until the year 2020 and around 80 percent until 2050). There are two ways of reducing VOC emissions to the atmosphere: changing VOC to other compounds containing small quantities of VOC, and reducing VOC emissions with the help of air treatment equipment.

Adsorption air treatment equipment loaded with carbonaceous fibers was created for VOC removal from the air. Carbonaceous fibres are new materials that have many advantages, but they have not yet been comprehensively analysed. The present experimental research shows that these materials are very effective (VOC removal from air efficiency reaches 90 percent).

Key words: organic materials, adsorption air purification, carbon fibres, volatile organic compounds (VOC), acetone, toluene, buthylacetate

INTRODUCTION

Large amounts of hazardous chemical pollutants causing changes in the animal and plant populations get into ecosystems. One of the most relevant problems related to a major impact on ecosystems is global warming caused by the increasing amounts of carbon dioxide and CH_4, N_2O, halogen-hydrocarbons and NOx, CO as well as other volatile organic compounds (VOC) (except CH_4) in the atmosphere.

Researches into the influence of temperature on the growing capacity of ash-trees and oaks carried out in Lithuania show that with an increase in the average annual or monthly air temperature the radial increment of oaks and ashes also grows (with some exceptions). In this case, a positive impact of the greenhouse effect on some species of Lithuanian plants was recorded (Karpavičius et al., 2006; Ruseckas et al., 2006), but due to the greenhouse effect and changed distribution of precipitation on the earth the specific composition of ecosystems changes.

VOCs are discharged to the atmosphere during various processes occurring in the soil, water and air. However, the largest emissions thereof are directly related to human economic activities (when butanol, butylacetate, methanol, formaldehyde, phenol, benzene, toluene, xylene and other pollutants are discharged) (Baltrėnas et al., 2003). VOC emissions into the air can cause different allergies, malignant tumours, acute diseases of the respiratory and cardiovascular systems, children's neurological development disorders and other diseases. VOC can react with nitrogen oxides (NOx) and convert into underground (tropospheric) ozone and afterwards into gas causing the greenhouse effect. The reduction of VOC emissions to the atmosphere would allow reducing the greenhouse effect. The dynamics of pollutant emissions to the atmosphere is presented by the Department of Statistics under the Government of the Republic of Lithuania (Table 1) (Emissions..., 2006).

In the global practice, different equipment is used for removing gaseous pollutants from the air, such as scrubbers, adsobers, absorbers, catalysers, etc. A research on VOC removal from the air by a wet technique using water as an absorbent was carried out in Lithuania (Bakienė et al., 2003). During painting, lacquering and similar operations, depending on the composition of the applied paint or lacquer, different pollutants may be emitted into the air, such as acetone, toluene, butylacetate and others. Complex air

Table 1. **Pollutant emissions into the atmosphere in Lithuania, thousand tons**

Year	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Total amount	152.1	148.9	137.6	147.8	120.0	91.2	98.3	94.2	88.2	91.2	88.3
Solid pollutants	11.8	10.3	9.7	7.3	5.8	4.8	5.4	4.7	4.7	4.8	4.7
Gases and liquids	140.3	138.6	127.9	140.5	114.2	86.4	92.9	89.5	83.5	86.5	83.6
Sulphur dioxide	64.7	63.0	60.9	74.5	55.3	31.0	35.4	29.7	21.1	24.0	24.8
Carbon monoxide	14.6	26.2	14.8	25.9	23.8	21.2	20.0	21.0	21.5	22.4	20.7
Nitric oxides	13.3	13.9	24.5	14.8	13.4	11.1	10.4	11.0	11.6	12.1	12.6
VOC, t	7.6	8.4	6.7	5.9	13.3	15.3	26.2	26.7	28.4	26.9	24.6
Fluorides, t	57.6	47.9	35.6	49.6	28.1	21.0	18.0	22.2	24.3	17.1	16.8
Other, t	40.1	27.1	21.0	19.4	8.4	7.8	0.9	1.0	0.9	1.0	0.9

treatment techniques may be employed for the removal of gaseous pollutants from the air, e. g., electrostatic in combination with adsorptive ones (Baltrėnas et al., 2003). The adsorptive technique is the most suitable for the removal of small and medium gaseous pollutant concentrations from the air (Aranovich et al., 2000). It can be used for cleaning polluted air at work places by employing filters operating under recirculation mode (Baltrėnas et al., 2002). Among the most widely used adsorbents is activated carbon obtained from different organic raw materials, solid fuel (peat, stone, brown coal, anthracite), wood (trees, wooden off-cuts, cellulose industry waste) and leather industry waste (Stoeckli et al., 2002; Wang et al., 1999). Recently, new filtration materials – carboneous fibres – have been introduced for the removal of gaseous pollutants and aerosols from the air. Carboneous fibres represent a wide range of carbon-impregnated polyesters, graphite fibres and polymerized carboneous fibres. They are distinguished by a high air cleaning efficiency, the option to combine different materials in order to expand the scope of application, the possibility to be used in standard air treatment filters. By the sorption and de-sorption speed they supersede activated carbon 10–100 times; they are resistant to aggressive and organic media and preserve a temperature of up to 1,000 °C in an inert atmosphere. They can be regenerated by using electric heating (due to electric conductivity). But carboneous fibres have also some shortcomings: smaller regeneration possibilities compared to activated carbon, different types are characterised by a different scope of application, a smaller filtration capacity volume compared to that of adsorbents. There are some papers analysing the application of carbonaceous fibres to remove formaldehyde from the air. Apart from that, carbonaceous fibres can be used for the removal of different odours from the air (deodorization) (Changsub et al., 2001, Haiqin et al., 2002). Thermal treatment of the fibres reduces the size and volume of micro-pores because surface groups can be fully removed from them (Carrot et al., 2001). The adsorption of gas and vapours is researched by static and dynamic methods (Paliulis et al., 2000).

The aim of this research was to analyse the suitability of the filtration material, АУТ-М2, for the removal of VOC (acetone, butylacetate, toluene) from the air and the dependence of air treatment efficiency on the number of filtration material layers, airflow velocity and the concentration of pollutants in the air.

METHODS

Materials, devices and instruments. The experimental study of the suitability of the filtration material АУТ-М-2 to treat air polluted with VOC (acetone, toluene, butylacetate) was carried out, the ability of this material to sorp water by a static desiccatory technique was evaluated, and the thermal resistance of this material was examined. The experimental scheme is shown in Fig. 1.

MATERIALS

1) АУТ-М2 – elastic black material (carbonic sorbent), practically insoluble in water, alcohol and acetone. Its surface density is 200 g/m^2, and a force of 700 N is necessary for it to break. Its sorption pore volume according to benzene is no less than 0.5 cm^3/g.

2) Acetone (analytical grade).

3) Toluene (analytical grade).

4) Butilacetate (analytical grade).

Devices and instruments

1) thermometer, temperature measuring range 20–200 °C, error 0.5 °C;

2) psychrometer, relative air humidity measuring range 10–100%, error 10%;

3) barometer, pressure measuring range 610–790 mm Hg, error 0.8 mm Hg;

4) velocity meter TESTO-452, airflow velocity measuring range 0.4–60 m/s, error 0.1 m/s;

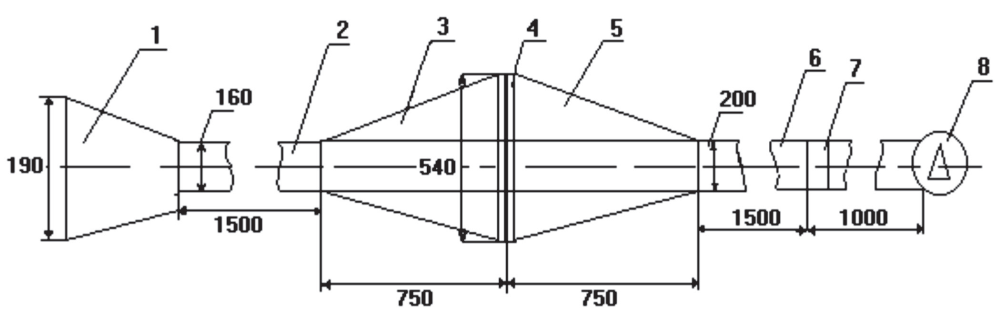

Fig. 1. Scheme of experimental air treatment equipment: *1* – suction hood; *2* and *6* – air ducts; *3* and *5* – transitions; *4* – filtration material; *7* – valve; *8* – ventilator

5) pressure meter DSM-1, pressure measuring range 0–2,000 and from 0 to 10,000 Pa, error 30 Pa;

6) analytical balance ВЛР-200, measuring range 0–100 g, error $5 \cdot 10^{-5}$ g;

7) gas chromatographer ЦВЕТ-500 with a flame ionizing detector, detector measuring limit $1.8 \cdot 10^{-12}$ g/cm^3 (tested with C9 alkane).

The study was carried out in the following stages:

1. Analysis of water adsorption by the filtration material.
2. Analysis of the thermal resistance of the filtration material.
3. Analysis of the suitability of the filtration material to remove VOC from the air.

Analysis of water adsorption by the filtration material. After roasting for 4 h at 100–105 °C, the filtration material was cooled in a desiccator to room temperature and after 24 h weighed with an analytical balance. From 0.5 to 3 g of the filtration material was used. To study the equilibrium of adsorption under conditions of 100% relative humidity the desiccator technique was applied. A plate containing deionized water was placed at the bottom of the desiccator. The desiccator produces a vapour concentration of 100% relative air humidity and the vapour gets in contact with the filtration material placed in the desiccator in an open vessel (weighing bottle). Under experimental conditions, at a temperature of around 20 °C, adsorption reaches equilibrium in the desiccator in 24 h. The amount of adsorbate is determined by weighing the filtration material. Every measurement was repeated five times and the average value was set afterwards.

Analysis of the thermal resistance of the filtration material. The thermal resistance of the fibrous material was determined by heating the material in a muffle furnace. The temperature in the muffle furnace was raised gradually from 20 to 550 °C and by keeping the carbonic fibre at this temperature 5 to 10 min. The test was repeated five times, each time using a new filtration material and setting the average value of the destructive temperature afterwards.

Analysis of the suitability of the filtration material to remove VOC from the air. The test of the suitability of the fibrous material AYT-M2 used to remove VOC from the air was performed by employing the experimental air treatment equipment designed at the Vilnius Gediminas Technical University (VGTU) Department of Environment Protection (see Fig. 1). It operates as follows: the air drawn by a ventilator (8) gets into the air treatment equipment via the intake hood (1), then moves within an air duct (2), transition (3), passes through filtration material (4), transition (5), an air duct (6) and escapes through the end of the air treatment equipment. The air treatment equipment air ducts (2 and 6) have openings where airflow velocity, aerodynamic resistance and the concentration of pollutants are recorded. Airflow velocity and aerodynamic resistance measuring places and sampling points are selected according to applicable standard documents (Paliulis et al., 1999; LAND 26-98/M-08, 1998).

The suitability of the fibrous material to remove VOC from the air was analysed in the following sequence:

• A mesh with the fibrous material was mounted in the filter.

• Formation of VOC source. The source was formed by heating acetone, butylacetate or toluene on an electric stove.

• Change of airflow velocity (see Fig. 1). The airflow velocity was changed by changing the position of the valve (7) from 1 to 8.

• Change of pollutant concentration. The pollutant concentration was changed by regulating the heating of separate pollutants (acetone, toluene and butylacetate) contained in the glass vessel on the electric stove.

• Change of the number of filtration material layers. The test employed the filtration material AYT-M2 of the following measurements: width 0.5 m, length 1 m, thickness 0.001 m. The notion of the "filtration material layer" means 1 mm thick filtration layer in the direction of airflow movement. When using 2 or 3 layers between two filtration materials mounted in the air treatment equipment, 0.5–1 cm thick air gaps were allowed between them.

• A piece of 1 mm wide filtration cloth was used. 0.5–1 cm gaps were allowed between the filtration layers.

• Measurements of velocity and aerodynamic resistance. Velocity was measured with a TESTO- 452 wing-type meter and the aerodynamic resistance with a DSM-1.

• Setting of VOC concentration. VOC concentration in the air duct was set by sucking gaseous samples into gaseous pipettes and afterwards analysing them with a gas chromatographer.

• Determination of cleaning efficiency. After recording the pollutant concentration before and after filtration, the air cleaning efficiency was calculated. The VOC concentration in the air duct was determined by the method of gas chromatography.

• Recording of ambient meteorological conditions. Since the ambient meteorological conditions influence the air treatment efficiency, they were recorded. The experiment was performed under the following meteorological conditions: 748–767 mm Hg pressure, 20–23.5 °C temperature, 48–59% relative humidity.

RESULTS AND DISCUSSION

The temperature and air humidity of air emissions from stationary sources of pollution vary within a very wide range. Therefore, it is very important to study the suitability of filtration materials used for the treatment of polluted air under high temperature conditions and analyse water adsorption. The temperature resistance research of the filtration material AYT-M2 was performed under oxidation conditions (in the air atmosphere).

Our findings are presented in Table 2.

The results show that AYT-M2 is a material resistant to temperature; its starts decomposing at around 540 °C. This material can be used for the treatment of polluted air in high-temperature processes. The carbonaceous fibre selected for our study is produced in Svetlogorsk (Belarus). It is made in the form of single-direction strips and woven strips with a different textile structure. This material is distinguished by a complex of valuable technical qualities and can be used as a reinforcing, electrically conductive and thermally as well as chemically resistant material, and as a heating element.

For the removal of VOC from polluted air, it is important to use materials distinguished by the highest efficiency of air treatment. In addition, it is important to analyse how airflow velocity influences the efficiency of air treatment. Therefore, we studied the efficiency of VOC removal from the air and evaluated changes in airflow velocity. The airflow velocity was 0.9–1.8 m/s.

Table 2. **Thermal resistance and water adsorption of fibre material**

Material type	Change in material mass, Δ m, g	Water adsorption, g/g
AYT-M2	0.73065 ± 0.0531	0.4849 ± 0.03524
	Temperature range of thermal destruction	Internal destruction temperature, °C
	535–545	540 ± 5

When the airflow velocity changes from 0.9 to 1.8 m/s and 1–3 layers of filtration material are used, the aerodynamic resistance varies in the range 60–180 Pa. The dependence of the efficiency of organic volatile compound removal from the air on the concentration of chemical pollutants in the air is shown Fig. 2. The axis of ordinates shows the cleaning efficiency, E, %, and the axis of abscissas gives the pollutant concentration in the air. The findings show that the cleaning efficiency of the air polluted with acetone (81.0%) is the lowest using AYT-M2 when the pollutant concentration in the air is 15.4 mg/m^3. When it increases to 25.4 mg/m^3, the air treatment efficiency grows up to 84.5% and practically remains stable when the concentration of acetone increases within the range of the concentrations studied.

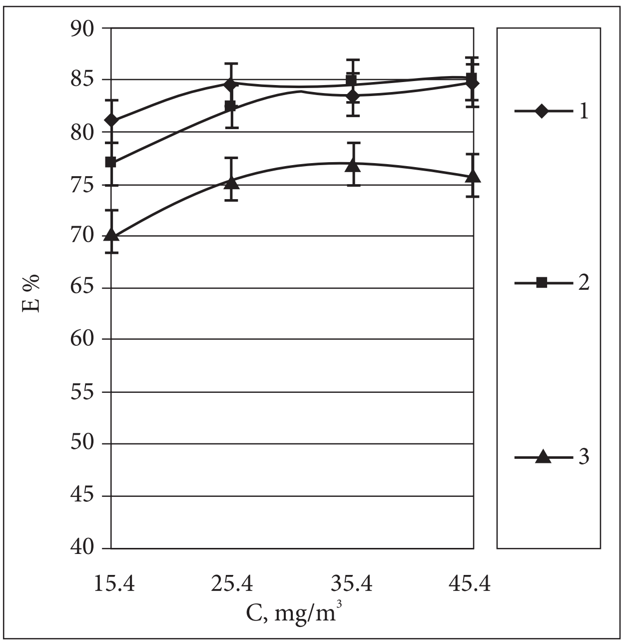

Fig. 2. Dependence of treatment efficiency of air polluted with VOC on pollutant concentration in the air when using one layer (1 mm thick filtration material cloth in the direction of airflow movement) at the airflow velocity of 0.9 m/s (*1* – acetone, *2* – toluene, *3* – butylacetate)

The lowest treatment efficiency (77.0%) of the air polluted with toluene within the range of the study concentrations was recorded using AYT-M2 at the pollutant concentration in the air 15.4 mg/m^3. When the pollutant concentration was increased to 25.4 mg/m^3, the air cleaning efficiency increased to 82.5% and later changed insignificantly with increasing the concentration. The lowest cleaning efficiency of the air polluted with butylacetate (70.3%) was recorded using AYT-M2 when the pollutant concentration in the air was 15.4 mg/m^3. With increasing the pollutant concentration to 25.4 mg/m^3, the air cleaning efficiency increased to 75.5%, while the subsequent concentration increase only insignificantly influenced the air treatment efficiency.

Analysis of the obtained dependence of air treatment efficiency on the concentration of the organic pollutants in the air allowed a conclusion that the concentration of the study VOC below 15.4 mg/m^3 would precondition an even lower air treatment efficiency.

The dependence of the treatment efficiency of the air polluted with VOC on the airflow velocity is presented in Fig. 3. The axis of ordinates shows the cleaning efficiency E (%), and the axis of abscissas indicates airflow velocity.

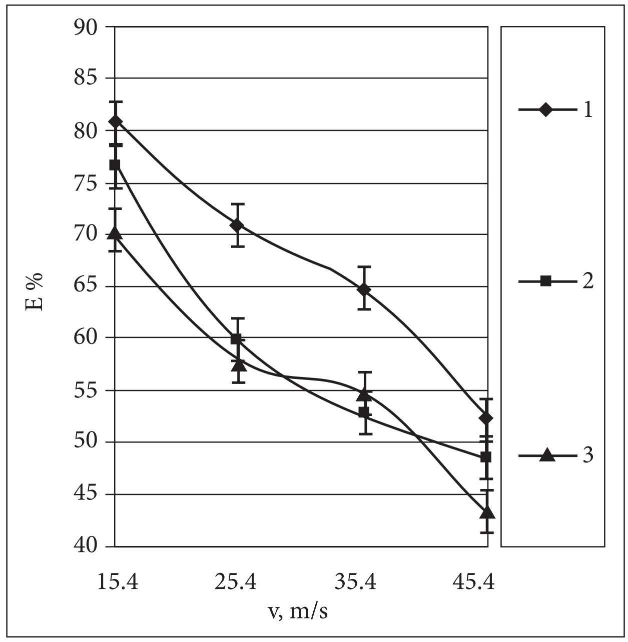

Fig. 3. Dependence of treatment efficiency of air polluted with VOC on airflow velocity when using one layer (1 mm thick filtration material cloth in the direction of airflow movement) at the pollutant concentration in the air 15.4 mg/m^3 (*1* – acetone, *2* – toluene, *3* – butylacetate)

The highest cleaning efficiency of the air polluted with acetone (81%) was recorded using AYT-M2 at the airflow velocity 0.9 m/s. When the airflow velocity was increased to 1.8 m/s, air treatment efficiency fell down to 52.7%. When removing toluene from the air using AYT-M2 at the airflow velocity 0.9 m/s, air treatment efficiency was 77%. When the airflow velocity increased to 1.8 m/s, the air treatment efficiency decreased to 48.8% (Fig. 3).

To treat the air polluted with butylacetate, AYT-M2 was used. When the airflow velocity was 0.9 m/s, air cleaning efficiency reached 70.3%, and when airflow velocity increased to 1.8 m/s, the air treatment efficiency fell down to 43.8%. Thus, with increasing the airflow velocity, air treatment efficiency decreases due to a decreasing time of contact between filtration material and VOC. The dependence of the treatment efficiency of the air polluted with VOC on the number of filtration material layers is given in Fig. 4. The axis of ordinates shows the treatment efficiency E(%), and the axis of abscissas indicates the number of filtration material layers.

The highest treatment efficiency of the air polluted with acetone (88.4%) was achieved using AYT-M2 and applying three

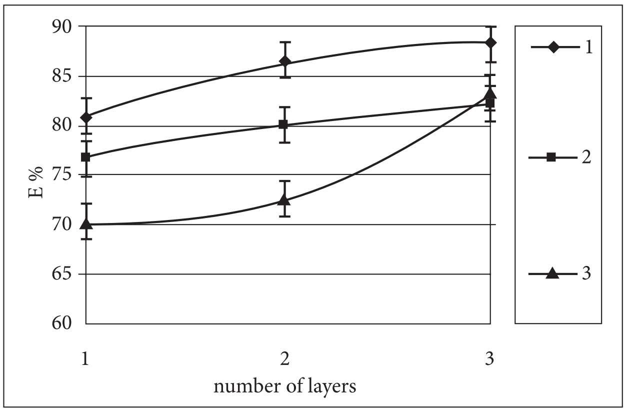

Fig. 4. Dependence of VOC removal from the air on the number of filtration material layers (one layer means 1 mm thick filtration material cloth in the direction of airflow movement; 0.5–1.0 cm air gaps are allowed between two cloths of filtration material when 2 or 3 layers are used) at the airflow velocity of 0.9 m/s and pollutant concentration in the air 15.4 mg/m^3 (*1* – acetone, *2* – toluene, *3* – butylacetate)

layers of filtration material at a 0.9 m/s airflow velocity. With one layer of filtration material, the air treatment efficiency decreased to 81%. A 82.0% treatment efficiency of the air polluted with toluene was achieved applying АУТ-M2 and using three layers of filtration material at a 0.9 m/s airflow velocity. With one layer of filtration material, the air treatment efficiency fell to 77%. A 82.9% treatment efficiency of air polluted with butylacetate was achieved applying АУТ-M2 and using three layers of filtration material at a 0.9 m/s airflow velocity. When using one layer of filtration material, the air treatment efficiency decreased to 70.3% (Fig. 4).

In the literature, data on the application of the carbonaceous fibre АУТ-M2 for removing acetone, toluene or butylacetate from the air are rather searce. Our study of the suitability of carbonaceous fibres for the removal of nitric oxides or welding aerosol from the air shows that, depending on the nature of fibre (актив карбопон, T-040 or АУТ-M2), air treatment efficiency varies in the range of 30 to 86.5% (Балтренас и др., 2004). Weschler presented data on how modified carbonaceous fibres removed VOC from the air when the airflow rate was 1 m^3/s, depending on the nature of the substance to be removed (the highest efficiency was achieved for perchlorethylene – 100%, toluene – 90%, m, p-xylene – 80%, o n-dodecane – 50% (Fisk, 2006). Our findings show the experimental data to be similar to the theoretical ones (when using one layer of filtration material, air treatment efficiency for toluene varies within 77 to 85%, depending on the pollutant concentration, see Fig. 2), even though the airflow rate was 0.45 m^3/s.

The highest cleaning efficiency of the filter was achieved when applying three layers of filtration material. This is related to the fact that with increasing the surface area of the filtration material in contact with pollutants, the cleaning efficiency also increases. As we see, when the number of filtration layers is gradually increased from 1 to 3 (Fig. 4), every new filtration layer increases the air treatment efficiency less; besides, with increasing the number of layers the aerodynamic resistance also proportionally increases and thus the energy costs also increase; therefore, the filtration material was limited to three layers. It is rational not to increase the number of layers but to change or regenerate the filtration material more often.

The expedience of applying filtration materials is obvious since they reduce the pollution of the ambient air and technosphere with VOC. This, in turn, will have a positive impact on human health and working capacity.

CONCLUSIONS

1. The experimental water adsorption research conducted by the desiccator method under static conditions shows that 1 g of the carbonaceous fibre AYT-M2 sorps 0.4849 ± 0.03524 g. The material AYT-M2 showed a rather high resistance to temperature and started decomposing at around 540 °C, therefore it can be used in high-temperature processes to treat the air polluted with VOC.

2. With increasing the airflow velocity from 0.9 m/s to 1.8 m/s the filtration material AYT-M2 removes acetone in the range 81–52.7%, toluene 77–48.8%, and butylacetate 70.3–43.8% from the air. With increasing the airflow velocity, the air treatment efficiency decreased because of the decreased time of contact between the filtration material and the pollutant.

3. The filter reached the highest efficiency when three layers of the filtration material were used. Applying the carbonaceous fibre AYT-M2 and increasing the number of filtration layers from 1 to 3, the treatment efficiency for acetone was 81–88.4%, for toluene 77–82% and for butylacetate 70.3–82.9%. When the surface area of the filtration material in contact with the pollutant increases, the cleaning efficiency also increases; however, every new filtration layer increases the cleaning efficiency less and less; in addition, with increasing the number of layers the aerodynamic resistance also proportionally grows and thus the energy costs also increase; therefore, the filtration material was limited to three layers. It is rational not to increase the number of layers but to change or regenerate the filtration material more often.

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LAKIŲJŲ ORGANINIŲ JUNGINIŲ EMISIJOS Į ATMOSFERĄ TYRIMAS IR MAŽINIMAS

Santrauka

Kiekvieną dieną tūkstančiai tonų pavojingų cheminių teršalų patenka į aplinką ir daro neigiamą poveikį ekosistemoms. Šiltnamio efektas yra vienas pavojingiausių efektų ekosistemoms. Anglies dioksidas, CH_4, N_2O, halogeniniai angliavandeniliai ir kt. (išskyrus CH_4) lakieji organiniai junginiai (LOJ) turi įtakos šiltnamio efektui.

Mažinti LOJ emisiją į atmosferą yra labai problemiškas. LOJ emisija 1996–2003 m. didėjo nuo 7,6 iki 28,4 t per metus, nuo 2004 m. – lėtai mažėja. LOJ patenka į atmosferą vykstant įvairiems procesams dirvožemyje, vandenyje ir atmosferoje.

Didžiausia LOJ emisija į aplinką yra susijusi su atmosferos tarša. Straipsnyje analizuojamos populiarių LOJ (acetono, tolueno ir butilacetato) emisijos į atmosferą mažinimo galimybės. Šie junginiai yra naudojami kaip tirpikliai. Naftos pramonė, chemijos pramonė, maži deginimo įrenginiai, maisto pramonė, atliekos ir kolektyviniai ūkiai taip pat yra LOJ emisijų šaltiniai. Pagal 2005 m. lapkričio 16 d. priimtą Europos parlamento rezoliuciją, šiltnamio dujų kiekiai turi būti ženkliai sumažinti (30% iki 2020 m. ir apie 80% iki 2050 m.). Yra du pagrindiniai keliai LOJ emisijai į atmosferą mažinti: LOJ pakeitimas kitais junginiais su mažesniu LOJ kiekiu, LOJ emisijos mažinimas panaudojant oro valymo įrangą.

Adsorbcinio oro valymo įrenginys su anglies pluošto įkrova buvo sukurtas LOJ šalinti iš oro. Anglies pluoštai yra naujos, pranašesnės medžiagos, bet dar nepakankamai ištirtos. Atlikti eksperimentiniai tyrimai parodė, kad šios medžiagos yra labai efektyvios (LOJ šalinimo iš oro efektyvumas siekia iki 90%).

Raktažodžiai: organiniai junginiai, adsorbcinis oro valymas, anglies pluoštai, lakieji organiniai junginiai (LOJ), acetonas, toluenas, butilacetatas