



Rasa Vaiškūnaitė

**RESEARCH ON AND APPLICATION OF
THE BIOFILTER WITH ACTIVATED
PINE BARK CHARGE FOR AIR
PURIFICATION**

Summary of Doctoral Dissertation

Technological Sciences, Environmental Engineering
and Landscape Management (04T)

Vilnius „Technika“ 2004

**VILNIUS GEDIMINAS TECHNICAL
UNIVERSITY**

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TAIKYMAS ORUI VALYTI**

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The Main Characteristics

The Problem

Atmosphere pollution is one of the greatest problems that industries of developed and developing countries have been recently encountering with. A lot of volatile compounds of organic nature, as butanol, butyl acetate, ethyl acetate, xylene, etc. is used in many branches of industry – wood processing, paint, enamel, plastic, paper and wood-pulp, food industries, and is released to atmosphere. Usually, the hazardousness of these chemical materials is determined by their chemical nature and the harmfulness of these chemical substances is determined mostly by their chemical nature and overrun of the Maximal Allowable Concentration (MAC) in the air of the living and working environment. Basing on the statistic data of 2003 presented by the Ministry of Environment of the Republic of Lithuania, the following amounts of pollutants are emitted into the air by various enterprises (SC „Klaipėdos Baldai“, SC „Mažeikių Nafta“, SC „Naftos Terminalas“, etc.): up to 230 tons of butyl acetate, 63 – ethyl acetate, 123 – butanol, 412 – xylene, 276 – toluene, etc. In order to neutralise these pollutants, different air purification methods are applied: absorption, adsorption, catalytic oxidation, etc. Nevertheless, when applying these methods, we encounter their expensiveness, generation of harmful products of their decomposition, etc. Besides these traditional air purification methods, more up-to-date, cost-efficient and powerful biological air purification technologies of unsophisticated design are available.

Relevance of the Work

All over the world, besides traditional air purification methods biological air purification technologies that generate no waste are used cultivating certain microorganism cultures. Basing on this method, inexpensive waste generated in the course of different production processes (bark, bulrush, straw, active sludge, etc.) are used as a charge in the developed devices. Later, the spent charge of natural materials may be used as fertilisers. Up till now in Lithuania no particular attention has been paid to scientific researches related to biological air purification method, development, installation and use of devices using this methods in production enterprises has not been popular. According to statistic data, in Lithuania only one enterprise – SC „Klaipėdos Baldai“ – is known to be using a biofilter with straw charge for air cleaning from formaldehyde. To fill in this niche and to make the development of ecological biotechnologies more intense in Lithuania, a biofilter with the activated pine-bark charge has been designed at the Department of Environment Protection of Vilnius Gediminas Technical University; besides, complex

research on biological air purification processes have been carried out with the help of this biofilter.

The Goal of the Work

Using biologically activated pine-bark charge in the developed filter, to carry out biological air purification research and to evaluate the efficiency of the theoretical and practical methods and its application. The **main tasks** necessary to achieve this goal:

1. To evaluate scientific researches carried out on the base of biological air purification method on devices developed and installed in Lithuania and in other countries.
2. To define and find a quantitative expression of the dynamics of natural microorganism associations (bacteria and micromycetes) in biologically non-activated and biologically activated pine-bark.
3. To define and find a quantitative expression of the ability of natural microorganisms to dissolve pollutants (butyl acetate, butanol and xylene) of different concentrations.
4. To carry out the research on the porosity of the pine-bark of different fraction (1–10, 10–20, 20–50 and 50–70 mm).
5. To examine the ability of the biologically activated pine-bark charge to sorb organic pollutants (butyl acetate, butanol and xylene) of different concentrations.
6. To evaluate the efficiency of the developed biofilter, depending on chemical nature, concentrations and mixture composition of pollutants.
7. To determine the main technical characteristics of the biofilter and to evaluate the efficiency of the device by changing its technical characteristics: the flow velocity (from 2,0 to 0,8 m/s) and volume (from 145 to 58 m³/h), height of the charge (from 150 to 750 mm), the number of layers (from 1 to 5), the fraction size (10–20 and 20–50 mm).
8. To carry out mathematical modelling of aerodynamic processes (velocity of the air fed, aerodynamic resistance of the biocharge) occurring in the biofilter.
9. Basing on research results and after full evaluation of physical, chemical and biological processes occurring in the course of air purification, to find out biodegradation constant α and absorption ability constant β that determine the device efficiency and to reveal their dependence on the nature of pollutants and technical parameters of the filter.
10. After complex researches are carried out, to evaluate the potential to employ the developed biofilter in different industry branches of Lithuania

(chemical, oil, furniture, food, etc.) for air purification from compound pollutants.

Novelty of the Work. The fractionated charge of coniferous bark has been used in the special equipment with natural microflora cultivated for air purification from volatile organic compounds; and the potential of the employment of the developed equipment has been fully evaluated after theoretical and experimental researches and mathematical modelling.

1. Definition and quantitative examination of the dynamics of natural microorganism associations (bacteria and micromicetes) in biologically non-activated and biologically activated pine-bark; also the evaluation of their impact on microbiological dissolution of organic pollutants.
2. The research on different fraction (1–10, 10–20, 20–50 and 50–70 mm) pine-bark porosity and the examination of the ability of biologically activated pine-bark to sorb organic pollutants (butyl acetate, butanol and xylene) of different concentrations.
3. Evaluation of the efficiency of the developed filter with the help of experimental research, depending on different parameters of the operation mode of the device (velocity and volume of the air fed, height of the charge, etc.), concentrations of pollutants, their mixture composition, biological activation of the charge and size of its fractions.
4. Theoretical and experimental determination of construction and technical characteristics of the biological air purification device.
5. Mathematical modelling of aerodynamic processes (velocity of the air fed, aerodynamic resistance of the biocharge) occurring in the biofilter.
6. Mathematical modelling of biofiltration and making use of the experimental data revealed the dependence of biodegradation constant α and absorption ability β of pollutants and their mixtures on the parameters of pollutants and technical parameters of the filter.
7. The development of the biofilter containing biologically activated pine-bark charge and theoretical and experimental researches enabling the evaluation of practical implementation of the filter.

Importance of the Work

The experimental researches and their results prove that development and use of biofilters for air purification from volatile organic compounds at various production enterprises of Lithuania is promising. Its implementation is especially promoted by comparatively low capital and operation costs of air

purification, high air purification efficiency and strict norms of pollutants emitted into the air set in the European Union.

Statements of the Doctoral Dissertation to be Maintained:

1. Up till now, no complex researches have been carried out in Lithuania on the application of the pine-bark charge for air purification.
2. The charge used in the filter is biologically active.
3. Biologically activated pine-bark charge of the filter has good sorption characteristics.
4. The developed filter containing the pine-bark charge is very efficient.
5. The results of mathematical modelling of aerodynamic and biofiltration processes validate the results received during experiments.
6. The developed biofilter containing the activated pine-bark charge may be used for air purification from different organic compounds at many production enterprises of Lithuania.

Approval of the Work

The total number of scientific publications on the subject of the doctoral dissertation is 16, including 6 of them published abroad. The results of the doctoral dissertation have been discussed in 8 international, 4 national scientific conferences, 12 scientific publications have been included into the compendia of conference reports. Basing on the material of the doctoral dissertation, 4 scientific articles have been published in reviewed periodicals and the patent „Biofilter“ has been obtained. The student was on three-month traineeship in Hamburg-Harburg Technical University, where scientific researches were introduced.

Extent and Structure of the Work

The doctoral dissertation consists of an introduction, six chapters, general conclusions, references, publications and reports on the subject of the doctoral dissertation. In total: 181 pages of text, 95 figures and 5 tables.

1. Biological Air Purification Method and Principles of Its Application

All over the world, the biological air purification method is widely used in many countries – Germany, Austria, Denmark, France, Sweden, etc. In practice, biological air purification equipment is divided into three main types: membranous bioreactor, biofilters and bioscrubbers. Such type air purification devices are simple and compact. Different waste generated in production processes – bark, bulrush, straw, active sludge, etc. – may be used as a charge

in such devices. Later, the spent charge of natural materials may be used as fertilisers.

2. Research on Biological Activation of Filter Charge

The goal of the research on biological activation of the charge is to define and find a quantitative expression of the dynamics of natural microorganism associations (bacteria and micromicetes) in biologically non-activated and biologically activated pine-bark; also to evaluate the ability of microbes to dissolve pollutants of different concentrations (up to 100 mg/m^3) under laboratory conditions.

In the pine-bark charge, activated during the investigation bacteria – *Pseudomonas sp.*, *Micrococcus sp.*, *Bacillus sp.*, *Flavobacterium sp.* etc. – prevail among natural microorganisms found at different periods of time during the equipment operation rather than micromicetes – *Penicillium sp.*, *Aspergillus sp.*, etc. Such distribution of microorganisms (bacteria and micromicetes) in the activated pine-bark charge has been determined by different humidity, temperature and other surrounding conditions. In deeper layers (50–150 mm deep), where concentration of hydrogen ions is close to a natural one ($\text{pH} = 7,2$), the optimal humidity (60 %), temperature ($25\text{--}30^\circ\text{C}$), and other conditions are more favourable for the development of oxidizing bacteria. While, at the surface of the charge (up to 50 mm), higher temperature (above 30°C) and speedier drying stimulated domination of micromicetes [1 A, 3 A].

Such a distribution of microorganisms in each layer of the biofilter charge was apparently depended on the duration of its biological activation with the help of organic compounds and on the depth of the charge layer (Fig 2. 1). At deeper layers (50–150 mm) of the charge, bacteria were dominated during the first 45 hours of filter maintenance. at concentration of hydrogen ions close to a neutral value ($\text{pH} = 7,2$), a sufficient humidity (60 %) and optimal temperature ($25\text{--}30^\circ\text{C}$) conditions were more favourable for the development of bacteria oxidizing pollutants, while on the charge surface (up to 5 cm) a higher temperature (over 30°C) and more rapid drying of the charge ensured the dominance of micromicetes. At the beginning of filter maintenance (after 4 days), at the surface of the charge (up to 50 mm) and in deeper layers (50–150 mm deep) the amount of bacteria was $2 \cdot 10^6$ and $8 \cdot 10^6$ cfu/g, respectively, after 25 days – $2 \cdot 10^7$ and $4 \cdot 10^7$ cfu/g, after 45 days – $5 \cdot 10^7$ and $8 \cdot 10^7$ cfu/g. Similarly, the number of micromicetes in the activated pine-bark charge was increasing depending on the duration of its activation [7 A-9 A]. The distribution of the number of microorganisms depending on the duration of filter maintenance was proved by high coefficients of statistic analysis

reliability ($R^2 = 0,83$, $R^2 = 0,85$, $R^2 = 0,93$, $R^2 = 0,95$). The number of microorganisms on the biomedium surface and its deeper layers became equal after 66 days of the filter operation, and the amount of bacteria was $1 \cdot 10^8$ cfu/g, that of micromicetes was $4 \cdot 10^5$ cfu/g.

According to the total amount of natural microorganisms, during 65 days of the filter operation, it was determined that the longer microorganisms are in contact with organic pollutants, the more intensively they grow and more efficiently oxidize organic pollutants.

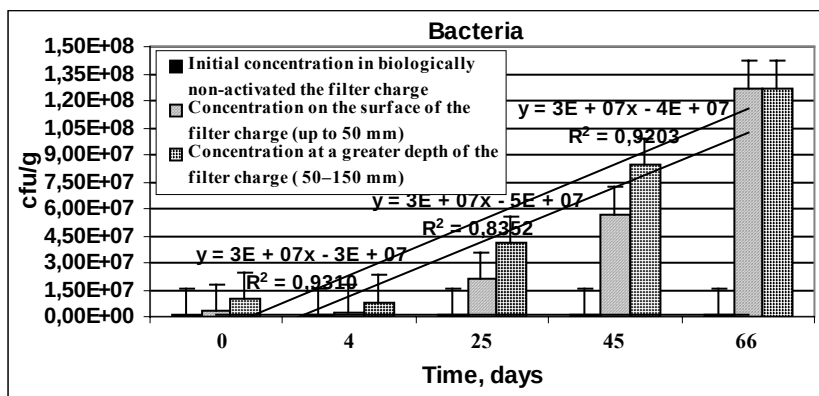


Fig 2. 1. Distribution of an average bacteria cell number at a different charge depth and different time of the equipment operation

For example, feeding butyl acetate through the filter when the initial concentration of butyl acetate is 63 mg/m^3 , after 480 hours from the beginning of the filter operation the amount of bacteria increased up to $4,1 \cdot 10^7$ cfu/g and the equipment efficiency reached 40 %, and correspondingly after 960 h bacteria increased up to $8,4 \cdot 10^7$ cfu/g; after 1 440 hours bacteria increased even up to $1,3 \cdot 10^8$ cfu/g and the equipment efficiency reached 65 % (Fig 2. 2). Similar results were also obtained with other pollutants of different concentrations (butanol and xylene). It shows that active cultures of microorganisms (bacteria and micromicetes) oxidizing organic pollutants were developed in the filter charge [8 A, 13 A]. These researched have been were validated by rather high coefficients of approximation reliability (R^2) reaching up to 0,98 and 0,99.

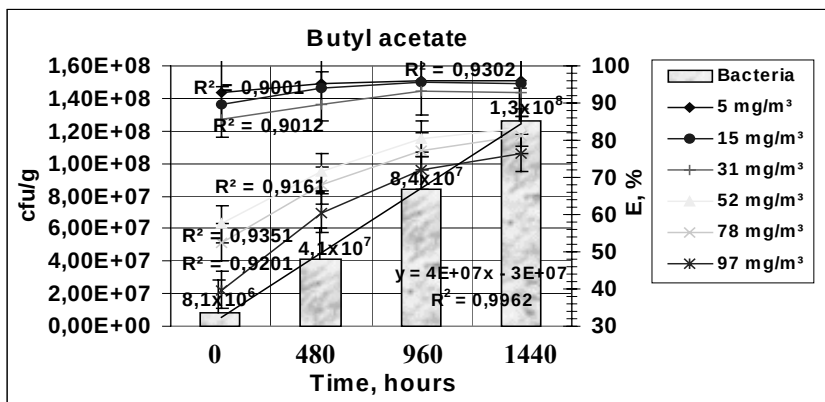


Fig 2. 2. Mean dynamics of the number of bacteria cells and the efficiency of the biofilter operation while purifying the air of butyl acetate depending on the operation time, when the flow rate is 2 m/s and the temperature is 20–55 °C

During filter operation, the microbiological oxidation of pollutants of different concentrations (up to 100 mg/m) was also determined in Petri plates. Among the investigated organic compounds (butyl acetate, butanol, xylene, ethyl acetate, ethanol, methanol) of the same concentration (30 mg/g), bacteria most intensively decompose compounds with a higher solubility in water – butanol and butyl acetate, and micromicetes – xylene.

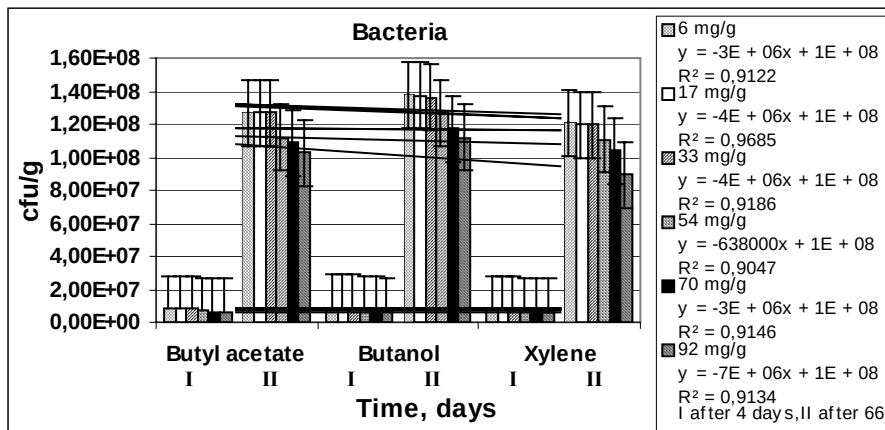


Fig 2. 3. Distribution of an average bacteria cell number in the samples of different biological charges affected by different pollutant concentrations

The research carried out under laboratory conditions showed the impact of different concentrations (6, 17, 33, 54, 70, 92 mg/g) of volatile organic compounds (butyl acetate, butanol, xylene) on the natural microorganism cultures living in the filter charge (Fig 2. 3). With comparatively low (up to 30 mg/g) pollutant concentrations, the number of natural microorganisms was hardly changing. The increase of pollutant concentrations (from 30 to 100 mg/g) resulted not only in a decreasing number of microorganisms but also in slower microbiological oxidation of pollutants [7 A, 13 A]. The research carried out under laboratory conditions is reliable, as rather high reliability coefficients (R^2) were received (0,90–0,96). Microbiological researches on the activated pine-bark charge of the biofilter were carried out using different fraction charge (10–20 and 20–50 mm). The above examples were received using finer pine-bark fraction (10–20 mm). The research results reveal that the number of microorganisms is 10 % higher in the finer pine-bark fraction (10–20 mm), than in the coarser one (20–50 mm), as the fine fraction form a bigger surface area.

3. Research on Porosity and Sorption of the Filter Charge

The goal of the research on the characteristics of the biofilter charge is to determine the porosity of the selected biocharge fractions (10–20 and 20–50 mm) and to evaluate the ability of pollutant sorption, by feeding organic compounds (butyl acetate, butanol and xylene) of different nature and concentrations (up to 100 mg/m³). After carrying out the research on different fraction (1–10, 10–20, 20–50 and 50–70 mm) pine-bark porosity, the use of the pine-bark of very fine fraction (1–10 mm) and low porosity (40 %) was refused. It is not efficient to use very fine fraction in the filter, as the finer is the fraction, the more it accumulates water and humidity, which causes a considerable aerodynamic resistance and the lack of oxygen for the microorganisms cultivated in the filter. The using of coarser fraction pine-bark (50–70 mm) with rather high porosity (85 %) was also refused, as the coarser is the fraction, the lower is its amount (only 30 kg in the filter) and the efficiency of the device. Neither too fine, nor too coarse pine-bark fraction (10–20 and 20–50 mm) was used in the research. The choice of this bark fraction was determined by the fact that rather high porosity is found among bark cells and other fraction cavities (75–80 %) and its amount in the filter is rather high (70–75 kg or 0,18 m³). For these reasons the contacting surface area of the biomedium is rather big and catches the organic pollutants fed through the

filter [10 A, 16 A]. The carried out research is reliable enough, ($R^2 = 0,95$). After determining the porosity of different pine-bark fractions, their ability to sorb organic compounds (butyl acetate, butanol and xylene) was evaluated in the course of 80 days of filter operation. (Fig 3. 1, 3, 2).

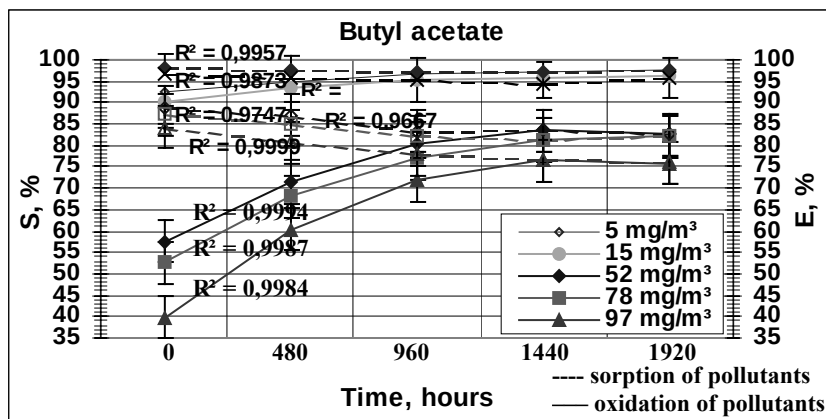


Fig 3. 1. The ability of the activated biofilter charge to sorb butyl acetate, when the velocity of the air fed is 2 m/s and the temperature is 20–55 °C

During air purification, pollutants are first of all sorbed into the biomedium, and only then they undergo treatment [15 A]. It was determined that the intensity of pollutant sorption largely depends on biological activation of the charge, i.e. on the ability of microorganisms cultivated in it to dissociate the selected organic compounds.

At the initial stage of filter operation (during the first 20 days), compounds with higher water-solubility – butanol and butyl acetate – feature better sorption in the biocharge (up to 75–100 %) if compared to xylene that has low water-solubility (up to 35–65 %) [10 A]. While, during the first 20 days the sorption of xylene was slower (35–65 %) (Fig 3. 3). In the course of further research (20–40 days later), with the charge getting further activated the process of biological air purification was markedly improving (35–45 %), while the sorption of butanol and butyl acetate into the biomedium slightly dropped (15 %). Such different sorption of pollutants and their oxidation in biocharge was determined by its low biological activation. With the increasing biological activation of the biomedium and efficiency of air purification process (45–80 %), the ability of the charge to absorb butanol and butyl acetate slightly decreased (from 100 to 70–75 %), as after 20–40 days of bark charge

activation the processes of pollutant oxidation became much faster than their sorption. Differently from the above-mentioned compounds (butanol and butyl acetate), the sorption of xylene featuring low water-solubility gradually increased (from 45 to 75 %) [15 A].

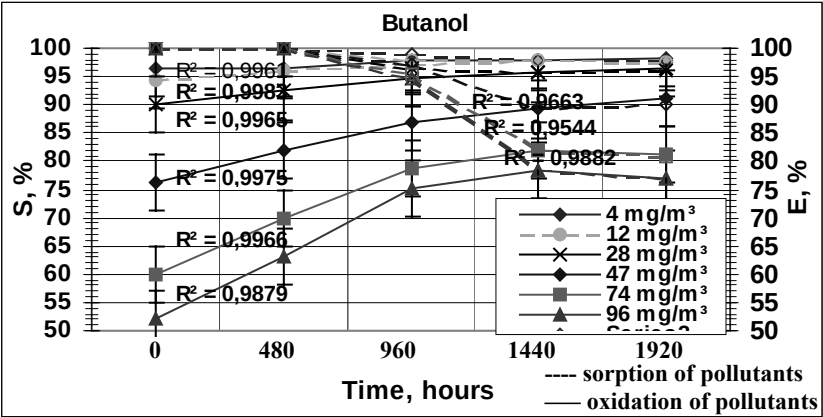


Fig 3. 2. The ability of the activated biofilter charge to sorb butanol, when the velocity of the air fed is 2 m/s and the temperature is 20–55 °C

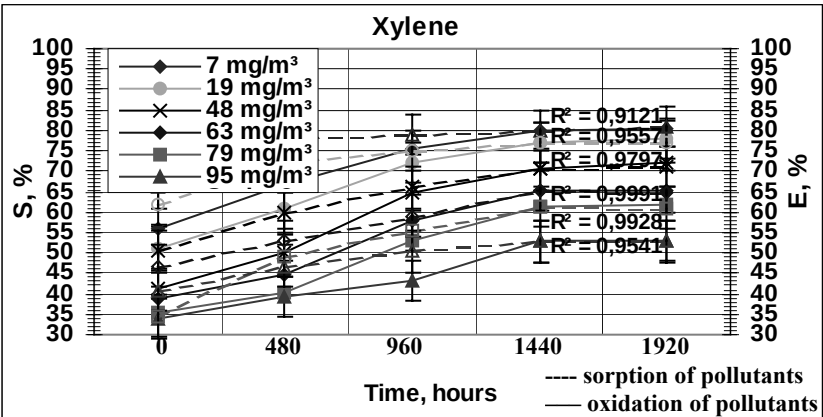


Fig 3. 3. The ability of the activated biofilter charge to sorb xylene, when the velocity of the air fed is 2 m/s and the temperature is 20–55 °C

After 65–80 days of filter operation, with the adaptation of microorganisms completed and with the highest and stabilized amount of

microorganisms as well as the effect of air purification (75–98 %) recorded in course of the research, the equilibrium of sorption and biochemical mechanisms was achieved. At that point the amount of all absorbed pollutants (butanol, butyl acetate and xylene) equalled to the amount of oxidized pollutants (60–95 %) (Fig 3. 3).

The result of the carried out research reveals that the sorption ability of the activated biofilter charge depends on its activation, which was proved by statistic analysis and by rather high approximation coefficients obtained (R^2) (from 0,91 to 0,99). During experimental researched it was found that replacement of the coarser bark fraction (20–50 mm) into the finer one (10–20 mm) in the filter resulted by 10–15 % improvement of the device efficiency and the sorption ability of the biologically activated pine-bark charge. For example, the amount of butanol (the initial concentration 96 mg/m³) absorbed into the biomedium of coarser fraction (20–50 mm) was 69 mg/g the amount sorbed into that of the finer fraction (10–20 mm) was 72 mg/g. Thus, the finer is the fraction (but not too fine) of pine-bark (1–10 mm), the bigger surface area is sorbed, and the greater number of microorganisms is found in it and, thus, the higher is the biological activity of the charge. The received reliability coefficients (R^2) (0,85 to 0,99) proved the dependence of pollutant sorption on the size of the charge fraction.

4. The Research on the Efficiency of the Biofilter Containing the Activated Pine-bark Charge

The goal of experimental researches on the filter efficiency is to determine the main technical characteristics of the designed device and to evaluate the efficiency of the devices by changing its technical characteristics (the flow rate and volume, concentrations of pollutants (butyl acetate, butanol, and xylene) (up to 100 mg/m³), height of the charge, the size if its fractions (10–20 and 20–50 mm). Determination of the main technical parameters (aerodynamic resistance, air load, etc. enabled the evaluation of filter efficiency. For example, the increase of the air load of the filter charge (from 49 to 125 m³/m²/h), the aerodynamic resistance (from 17 to 140 Pa) and the air flow rate (from 0,8 to 2,0 m/s) results in lower air purification efficiency (e.g., from 84 to 71 % in case of butyl acetate). Experimental investigation of filter efficiency revealed that, among substances investigated, natural microorganisms more easily decompose compounds, which are more easily dissolved in water: butanol and butyl acetate (Fig 4. 1) [2 A, 14 A].

When initial concentrations of compounds fed through the filter (butyl acetate, butanol and xylene) are low (below 30 mg/m³), air purification efficiency was 95–98 %. When concentrations of organic compounds go up (from 30 to 100 mg/m³), efficiency of biological air purification drops down, as higher concentrations inhibit activity of microorganisms – their reproduction, breathing and capability to oxidise organic compounds. However, biological decomposition of xylene, a hydrocarbon of benzene family, in the same time period is substantially slower, i.e. when concentration is up to 27 mg/m³, purification efficiency reaches 77–80 % [5 A-7 A, 11 A]. This is proved by a statistic analysis ($R^2 = 0,96$).

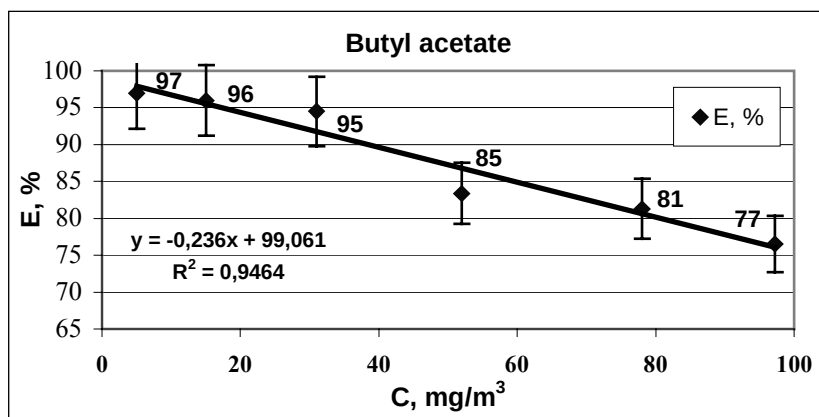


Fig 4. 1. Efficiency of air purification from butyl acetate after 64 days, when velocity of air flow injected was 2 m/s and temperature 20–55 °C

Biofilter efficiency depends on the amount of volatile organic compounds that are ingredients of a mixture. This is determined by quantity and quality of volatile organic compounds composing the mixture. When mixture rate is 1:1:1 (butyl acetate, butanol and xylene), purification efficiency is higher, up to 73–89 %, than that at rate 3:2:1, which is 41–71 % (Fig 4. 2). Capability of microorganisms to decompose volatile organic compounds depends also on quality composition of the mixture ($R^2 = 0,99$). When pollutants in the mixture are easily soluble in water and well biodegradable (butanol and butyl acetate), purification efficiency reaches 87 %. When the main component of the mixture is xylene, capability of microorganisms to oxidise pollutants chosen is lower and air purification efficiency is 81 % [8 A, 10 A, 12 A].

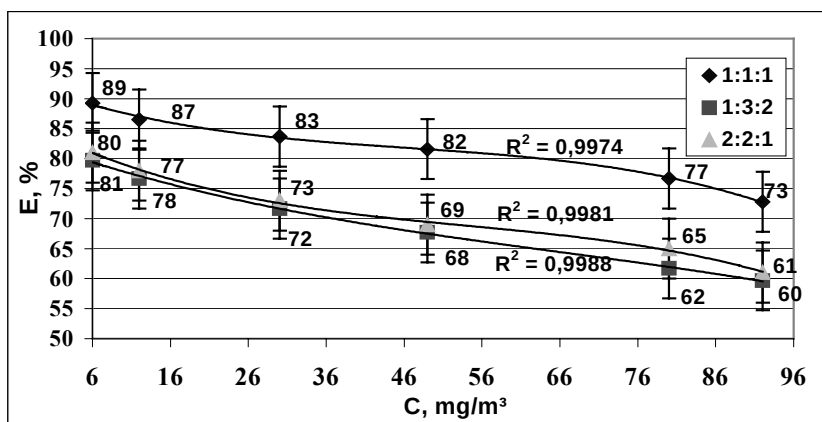


Fig 4. 2. Efficiency of air purification from mixtures of different ratio (1:1:1, 1:3:2 and 2:2:1; butyl acetate, butanol and xylene respectively) after 64 days, when velocity of air flow injected was 2 m/s and temperature 20–55 °C

Gas purification from volatile organic compounds (butyl acetate, butanol and xylene) is influenced by charge height and volume (Fig 4. 3). It is determined that increasing charge height (from 150 to 750 mm), its volume correspondingly (from 0,036 to 0,18 m³) and number of layers (from 1 to 5), area of surface in contact with pollutants becomes larger and efficiency of air purification becomes better. For example, it was determined that with one layer of the charge (150 mm high), purification efficiency was 70 % at initial concentration of butyl acetate equal to 52 mg/m³. Similarly, when number of layers was increased up to five (750 mm high), the efficiency of air purification at that same concentration has reached 85 % [6 A, 8 A, 11 A, 15 A]. Also, when increasing charge volume and number of its layers, efficiency of air purification was further improved by growing concentration of natural chemoorganotrophes in the biomedium. For example, in 65 days amount of bacteria has increased from $2 \cdot 10^6$ to $1,26 \cdot 10^8$ cfu/g and amount of micromycetes – from $3,8 \cdot 10^4$ to $4,5 \cdot 10^8$ cfu/g correspondingly. During the research it was found out that the filter efficiency also depends on the duration of the contact of the charge and pollutants. The approximation reliability coefficients of these researches were fluctuating within 0,93–0,99. Experimental investigation showed that the longer biological activation of the charge by gradually increasing concentrations of organic compounds (butyl acetate, butanol, xylene) injected, the better efficiency of biological air purification in the charge.

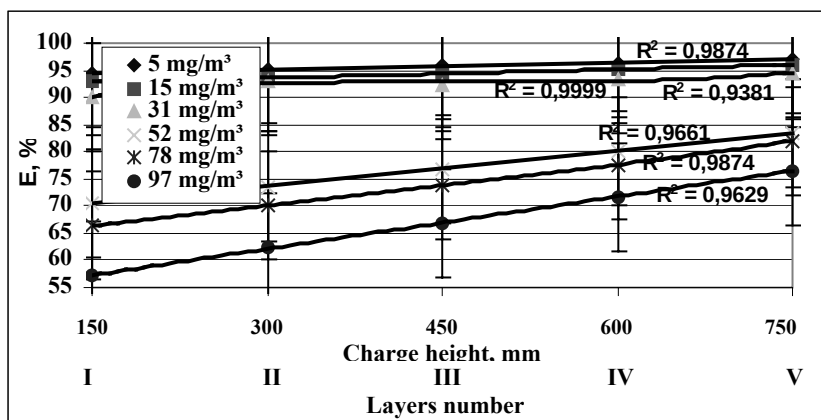


Fig 4. 3. Efficiency of air purification from butyl acetate after 64 days depending on the charge height (i.e. the number of layers), when velocity of air flow injected was 2 m/s and temperature 20–55 °C

The research on the biofilter efficiency was carried out using different fraction (10–20 and 20–50 mm) of pine-bark charge. The use of coarser pine-bark fraction in the filter resulted in 10–15 % lower efficiency than in case if finer fractions of pine-bark (10–20 mm). Thus, purifying the air from volatile organic compounds (butyl acetate, butanol and xylene) use of coarser fractions (10–20 mm) of pine-bark charge in the biofilter is more efficient. As it is obvious from regression equations, in all cases approximation reliability coefficients are higher than 0,9, which means that the air purification is efficient.

5. Mathematical Modelling of Aerodynamic and Biofiltration Processes in the Biofilter with Pine-Bark Charge

To project and to give a theoretical evaluation of aerodynamic processes going on in a biological air purification device, mathematical numeral modelling of a biofilter was performed with the help of versions 3.2 and 3.5 of PHOENICS. Aerodynamic processes in the filter – the velocity of the air fed and the aerodynamic resistance of the charge – were modelled and fully evaluated. Modelling was based on mathematical equations defining the physical laws. The distribution of the velocity of the air fed through the filter and the aerodynamic resistance depending on the number of charge layers was modelled. Similarly to the experimental researches, the increase of the biofilter charge layers (from 1 to 5) resulted in slower flow of the air being purified

(from 2,0 to 1,3 m/s). Its highest value was recorded at the opening of the filter (up to 1,7–2,0 m/s), and the lowest ones at the exit duct (up to 1,3 m/s) ($R^2 = 0,95$) (Fig 5. 1). Similarly to the results of experimental research, the highest aerodynamic resistance is formed at the bottom of the filter prior to the five layers of the filter charge. Reduction of the filter charge layers during modelling resulted in its lower aerodynamic resistance (from 156 to 10–20 Pa) ($R^2 = 0,91$) (Fig 5.1) [1 A].

Biochemical processes occurring in the biologically activated pine-bark charge were modelled. Basing on the results of experimental research and after full evaluation of the processes occurring during the research, the biofiltration processes were modelled using MAPLE 7, DATAFIT 6.1.10. Mathematical modelling of biofiltration and making use of the experimental data revealed the dependence of biodegradation constant α and absorption ability β of pollutants and their mixtures on the parameters of pollutants (initial concentration, molecular mass, number of carbon atoms in a molecule and their water-solubility) and technical parameters of the filter (charge height, velocity of the air fed).

In Equation (1) the coefficients are the function of the molecular mass (μ), the number of carbon atoms in a pollutant's molecule (N_C) and the solubility of materials (M):

$$K_i = f(N_C, \mu, M). \quad (1)$$

After the evaluation of the experimental results, a regression model was chosen for the above-mentioned coefficients:

$$K_i = a \cdot N_C + b \cdot \mu + c \cdot M + d, \quad (2)$$

where a , b , c – constants of the above mentioned parameters, such as the molecule mass, the number of carbon atoms in a pollutant's molecule and the solubility of materials, d is an ordinary constant.

If the process is stationary, i.e. $\frac{\partial C}{\partial t} = 0$ and due to sufficiently high velocity of the air fed we do not regard diffusion, then:

$$v \frac{\partial C}{\partial X} = -\varepsilon^2 \frac{\partial \beta}{\partial t}, \quad (3)$$

where C – pollutant concentration, mg/m^3 ; v – velocity of air flow in the filter, m/s ; X – biofilter charge height, m ; ε – filter charge porosity, β –

absorption ability of the filter charge, i.e. pollutant amount, absorbed by 1 m³ of the filter charge; t – time moment, during which pollutants are passing the biofilter, s.

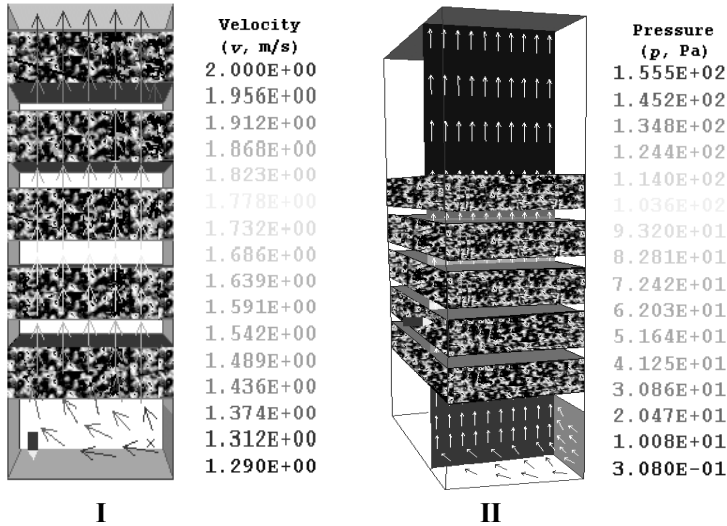


Fig 5. 1. Mathematical modelling of aerodynamic processes occurring in the filter with five charge layers, when the initial velocity of the fed air is 2 m/s:

I – dynamics of the velocity of the air fed (m/s), **II** – dynamics of the aerodynamic resistance of the charge (Pa)

Using this expression of the efficiency and the previous ones we obtain the formula for the calculation of the absorption capacity β :

$$\beta = (K_0 + K_1 \cdot C + K_2 \cdot X) C_0. \quad (4)$$

Biodegradation constant α has been estimated with the help of the following formula:

$$\alpha = \varepsilon^2 (K_0 + K_1 \cdot C + K_2 \cdot X) v / X. \quad (5)$$

Basing on the obtained relationship it is possible to calculate the concentration C of the pollutants emitted to the air:

$$C(X, \mu, \nu, N_c, M) = C_0 e^{-\varepsilon^2 (K_0 + K_1 \cdot C + K_2 \cdot X)} \quad (6)$$

Purifying the air from pollutant mixtures, the coefficients a , b , c of the Equation (1) $E = a + b \cdot X + c \cdot C_0$ slightly differ, thus the following expression may be used to determine the filter efficiency:

$$E = 74,78 + 0,11X - 0,31C_0. \quad (7)$$

In the course of modelling, bigger attention was paid to the regularities of mixture purification efficiency. As during various technological processes, production enterprises usually emit mixtures of pollutants rather than single pollutants. The dependence of the efficiency of each pollutant tested (butyl acetate, butanol and xylene) on the amount of other components and their ratio in the mixture was determined (Fig 5. 2) [4 A].

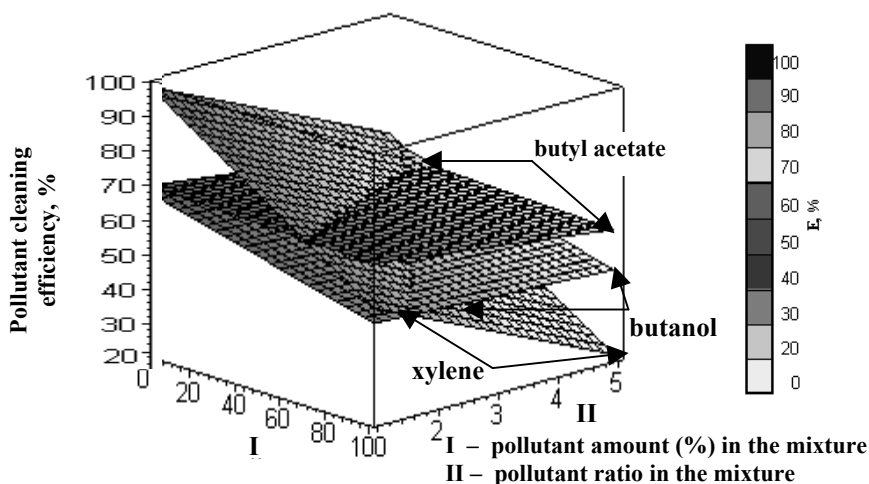


Fig 5. 2. The dependence of the tested pollutants (butyl acetate, butanol and xylene) on their amount and ratio in the mixture, when the initial concentration of the above-mentioned pollutants is 50 mg/m^3 and the height of the charge in the filter is 750 mm

This dependence is illustrated in a three-dimensional graph, where z – biofilter efficiency (%), x – amount of other materials tested (%) and y – their ratio in the mixture (from 1 to 5). Comparison of the received pollutant dependences on the amount of other components (%) and their ratio in the mixture reveals that the highest purification efficiency (from 50 to 100 %) is achieved in the case of butanol and butyl acetate, and the lowest one (from 20 to 70 %) in the case of xylene. Substantially, such different dependences are determined by different solubility of these materials and their biodegradation. Thus, the purification process of those pollutants (butanol and butyl acetate) that have easier biological dissolubility is less influenced by certain amount of other components (%) and their ratio in the mixture. In a similar way, the dependence of the efficiency of the pollutant tested (butyl acetate, butanol and xylene) on the amount of other components and their ratio in the mixture was determined for each pollutant. The results of mathematical modelling of biofiltration processes are reliable, as the received approximation coefficients are higher than 0,90. The results received during mathematical modelling revealed that modelling of processes occurring in the biofilter is much easier than performance of complicated, long and costly experimental researches. Mathematical modelling will enable the evaluation of filter efficiency both today and in future.

6. The Developed Equipment and Its Use

Review of biofilter application in different countries and of scientific-technical literature and the carried out researches revealed that biofilters are cheap and efficient. Basing on that, a filter with biologically activated pine-bark charge has been developed at the Department of Environmental Protection of Vilnius Gediminas technical University (Fig 6. 1). After carrying out the patent search in relation to biological air purification equipment, in November 2001 an application for patenting the developed biofilter was filed to the National Patent Bureau of the Republic of Lithuania. In May 2003, the National Patent Bureau decided that the patent application complies with the expertise requirements. In June 2003, the patent application was published in the official journal of the National Patent Bureau. In November 2003, the biofilter was patented. The number of the patent is LT 5061.

The biofilter (500·480·2 000 mm) with biologically activated charge contains five separate layers of biomedium (17), which mutually do not press each other, are separated by meshes (16) and ensure even distribution of air flow. When purifying air from volatile compounds of organic nature, flow of polluted air is blown through all five layers of biomedium by means of

ventilator (6). There are dampers (9) installed with control handles (8) in the inlet and outlet ducts of the filter to adjust air flow (58 to 145 m³/h) and flow velocity (0,8 to 2,0 m/s) (Fig 6.1). In order to increase amount of polluted air, inlet airduct (7) has funnel shape at the front end. Purified air is exhausted through the flexible duct (13) from the filter. There shall be maintained temperature of 30 °C in the filter charge, so the medium is heated by two heating elements (4) installed at the side walls of the filter. Prior starting the biofilter, the medium was moistened, biogenic elements were added and it was biologically activated by blowing organic pollutants through the medium. When air purifying process is in progress, there is consumed 20 l of water per day to maintain humidity of 60 % in the whole volume of the bark (0,18 m³). When the filter isn't operated, activity of microorganisms in the charge decreases, and water consumption goes down to only around 10 l.

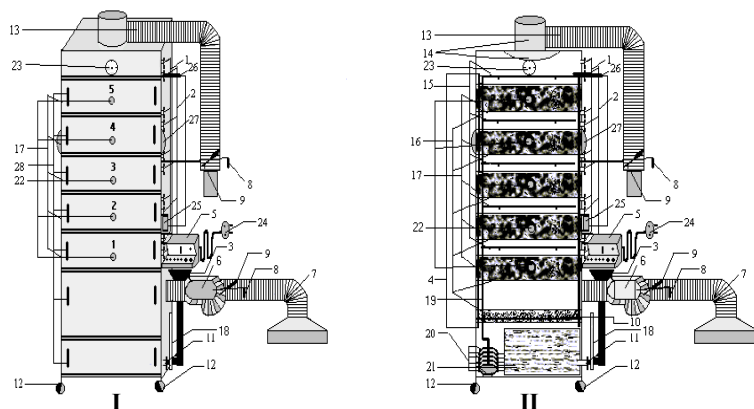


Fig 6. 1. Structure of biofilter with activated pine-bark charge: I – general front view, II – filter cross-section:

where 1, 2 – measurement openings; 3 – water tank; 4 – heating elements; 5 – control panel; 6 – ventilator; 7 – inlet airduct; 8 – damper control handle; 9 – damper; 10 – waste collector; 11 – water valve; 12 – castors; 13 – outlet airduct; 14 – condenser; 15 – nozzle; 16 – charge separating mesh; 17 – charge layer; 18 – measurement tube for water level in the tank; 19 – water supply pipes; 20 – pumps; 21 – water tank; 22 – pH measurement openings; 23 – thermometer; 24 – plug; 25 – temperature regulator; 26 – temperature sensor; 27 – filter handles; 28 – handles of layers

Water is supplied to each layer from the water tank (21) by means of five pumps (20) maintaining humidity in each layer at 60 %. Water is sprayed (15) above each layer and its excess is collected in the waste collector (10) together

with waste generated. The operation of the developed device is easy as it has four castors (12), seven handles or layers (28) and a pair of filter handles (27). The biofilter could also be used for experimental tests but in such case openings ($\varnothing$ 15 mm 1 and $\varnothing$ 20 mm 2, 22) for physical (pressure, velocity) and chemical (quality of the air being purified, concentration of hydrogen ions of the medium (pH)) should be made before and after each layer of the activated pine-bark layer. The device has a thermometer (23) and its control panel (5), used for connection to the network via the plug (24), in which necessary temperature is set and the water amount in the reservoir is monitored (Fig 6.1).

The efficiency of the filter with the biologically activated pine-bark charge reaches up to 90–95 %. The main technical parameters of the device: output – 145 m³/h, the amount (from 58 to 145 m³/h) and velocity (from 0,8 to 2,0 m/s) of the air flow fed through the filter, maximum temperature of the air being purified – 30 °C, aerodynamic resistance of the charge – 140 Pa (Fig 6. 1). Setting the filter with the activated pine-bark charge into an inactive status enable its operation in production enterprises working on discontinuous schedule. The research results have revealed that the developed biofilter could be used in all industry branches of Lithuania (chemical, oil, furniture, food, etc.) as it is cheap and effective.

7. General Conclusions

1. Evaluation of scientific researches carried out on the base of biological air purification method on devices developed and installed enable a conclusion that application of such devices is promoted in the world by considerable experience, their economy and efficiency, and in Lithuania – due to the sustainable development strategy – it is promoted by the need to replace ineffective technologies with the effective ones, not researched and applied in air purification from volatile organic compounds 25 % of which are annually emitted from different production enterprises.
2. In the pine-bark charge, activated during the investigation bacteria – *Pseudomonas sp.*, *Micrococcus sp.*, *Bacillus sp.*, *Flavobacterium sp.* etc – prevail among natural microorganisms found at different periods of time during the equipment operation rather than micromicetes – *Penicillium sp.*, *Aspergillus sp.* Depending on the biological activation of the charge and after the evaluation of the total amount of natural microorganisms it was determined that the longer microorganisms are adapted to organic pollutants the faster they develop (after 66 days of filter operation the amount of bacteria increased from $8,1 \cdot 10^6$ to $1,3 \cdot 10^8$ cfu/g, and that of micromicetes – from $3 \cdot 10^4$ to $4 \cdot 10^5$ cfu/g).

3. After the evaluation of the ability of microorganisms found in the charge of different biological activation under laboratory conditions, it could be stated that in case of low (up to 30 mg/g) concentrations, the number of microorganisms is hardly changing (bacteria in the case of butanol – from $1,30 \cdot 10^8$ to $1,31 \cdot 10^8$ cfu/g); with the increased pollutant concentrations (from 30 to 92 mg/g) the decrease in the number of microorganisms is more intense (bacteria in the case of butanol – from $1,1 \cdot 10^8$ to $1,0 \cdot 10^8$ cfu/g).
4. After the research on the porosity of the pine-bark fractions (1–10, 10–20, 20–50 and 50–70 mm), the pine-bark fraction (10–20 and 20–50 mm) of considerable porosity (75–80 %) and of considerable amount (70–75 kg or $0,18 \text{ m}^3$) was used. Using such fraction, a considerable contacting area is formed which catches the organic pollutants fed through the filter.
5. Depending on the biological activation of the charge, its sorption research proves that during the first 20 days of filter operation water-soluble butanol and butyl acetate (up to 75–100 %) are better sorbed into the charge if compared to xylene (up to 35–65 %). During the next 20–40 days, the sorption of butanol and butyl acetate, except xylene, into the biomedium was decreasing (15 %) with the increasing filter efficiency (35–45 %). After 65 days, the amount of pollutants sorbed into the biocharge became equal to the amount of the oxidized ones (60–95 %).
6. The research on the biofilter revealed that its efficiency largely depend on the chemical nature and concentrations of pollutants. When initial concentrations of butyl acetate and butanol are up to 30 mg/m^3 , and those of xylene are up to 19 mg/m^3 , the efficiency of purification from pollutants is 80–98 %; the increase of their concentrations results in lower efficiency of purification. When mixture rate is 1:1:1, the purification efficiency is higher, up to 73–89 %, than that at rate 3:2:1, which is 41–71 %.
7. After the evaluation of the main technical characteristics of the filter, it was determined that its efficiency is increasing (e. g., in the case of butyl acetate from 77 to 92 %) with the reduction of the velocity (from 2,0 to 0,8 m/s) and the volume (from 145 to $58 \text{ m}^3/\text{h}$) of the fed air, with the increase of the charge height (from 150 to 750 mm), and the number of layers (from 1 to 5). The filter efficiency also depends on the size of its charge fraction. Respectively, replacement of the coarser fraction (20–50 mm) with the finer one (10–20 mm) results in 10–15 % higher efficiency.
8. The received results of modelling of aerodynamic processes prove that the greatest aerodynamic resistance of the biocharge (up to 156 Pa) and the velocity of the air fed (up to 2,0 m/s) are formed before the fine layers of the charge. The reduction of the number of the biofilter layers (from 5 to 1)

results in lower velocity of the air fed and the aerodynamic resistance, respectively – up to 1,3 m/s and 10–20 Pa.

9. Basing on the research results and taking into consideration physical, chemical and biological processes, the dependence of biodegradation constant α and absorption ability β of pollutants and their mixtures on the parameters of pollutants (initial concentration, molecular mass, number of carbon atoms in a molecule and their water-solubility) and technical parameters of the filter (charge height, velocity of the air fed).
10. A biofilter containing the charge of the activated pine-bark has been developed and produced on the basis of the received patent and the potential of its application for air purification from volatile organic compounds in different production enterprises if Lithuania (chemical, oil, furniture, food, etc.) has been evaluated

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BIOFILTRO SU SUAKTYVINTA PUŠŲ ŽIEVIŲ ĮKROVA TYRIMAI IR TAIKYMAS ORUI VALYTI

SANTRAUKA

Sukūrus biofiltrą su biologiškai suaktyvinta pušų žievių įkrova, darbe atlikti oro valymo tyrimai, įvertintas teorinis ir praktinis metodo efektyvumas bei jo pritaikymas.

Pirmojoje darbo dalyje apžvelgtas biologinis oro valymo metodas ir jo taikymo galimybės. Aptarti šį oro valymo procesą lemiantys pagrindiniai aplinkos veiksniai: fizikiniai (temperatūra, drėgmė, hidrostatinis ir mechaninis slėgiai ir kt.), cheminiai (pH, įvairios cheminės medžiagos, deguonis ir kt.) ir biologiniai (mikroorganizmų aktyvumas, jų mityba ir kt.). Aprašyti biologinio oro valymo įrenginiai bei juose taikoma įkrova.

Antrojoje darbo dalyje apibūdintos bei kiekybiškai nustatytos pušų žievių mikroorganizmų (bakterijų ir mikromicetų) asociacijos, įvertintas jų biologinis

aktyvumas skaidyti tiriamus teršalus (butilacetatą, butanolį ir ksileną). Sukurto filtro pušų žievių įkrovoje dominuoja bakterijos – *Pseudomonas sp.*, *Micrococcus sp.*, *Bacillus sp.*, *Flavobacterium sp.* ir kt., o ne mikromicetai – *Aspergillus sp.*, *Penicillium sp.* Įvertinus mikrobų biologinį aktyvumą nustatyta, kad esant nedidelėms (iki 30 mg/g) teršalų koncentracijoms, jų skaičius kito nedaug (bakterijų su butanolio – nuo $1,30 \cdot 10^8$ iki $1,31 \cdot 10^8$ kfv/g), padidinus teršalų koncentracijas (nuo 30 iki 92 mg/g), mikrobų skaičius sumažėjo (bakterijų su butanolio – nuo $1,1 \cdot 10^8$ iki $1,0 \cdot 10^8$ kfv/g).

Trečioji disertacijos darbo dalis skirta nustatyti pagrindines bioįkrovos charakteristikas – porėtumą bei sorbciją. Atlikus žievių porėtumo tyrimus, filtro įkrovoje naudotos nemažą porėtumą (75–80 %) sudarančios frakcijos (10–20 ir 20–50 mm). Bioįkrovos sorbcijos tyrimai parodė, kad pirmąsias 20 filtro parų, į ją geriau sorbuojami vandenyje tirpstantys butanolis, butilacetatas (iki 75–100 %), o ne mažai vandenyje tirpus – ksilenas (iki 35–65 %). Per kitas 20–40 paras, didėjant filtro efektyvumui (35–45 %), mažėjo (15 %) butanolio ir butilacetato, išskyrus ksileną, sorbcija į bioterpę. Po 65 parų įkrovoje sorbuotų teršalų kiekis tapo lygus oksiduotų kiekiui (60–95 %).

Ketvirtoje darbo dalyje nustatytos pagrindinės įrenginio techninės charakteristikos bei įvertintas jo efektyvumas. Atlikti tyrimai parodė, kad kai pradinės butilacetato, butanolio koncentracijos iki 30 mg/m³, teršalai valomi 80–98 % efektyvumu. Didinant jų koncentracijas (nuo 30 iki 100 mg/m³), mažėjo (iki 60–70 %) filtro efektyvumas. Oro valymas taip pat priklausė nuo teršalų mišinių sudėties, atitinkamai, kai jų santykis 1:1:1 išvaloma efektyviau (iki 73–89 %), nei santykiui esant 3:2:1 (41–71 %). Filtro efektyvumas gerėja (pvz. butilacetato nuo 77 iki 92 %), mažinant per jį leidžiamo oro srauto greitį (nuo 2,0 iki 0,8 m/s), didinant įkrovos aukštį (nuo 150 iki 750 mm) ir pan.

Penktoje disertacijos dalyje PHOENICS 3.2 bei 3.5 programa įvertinti aerodinaminiai procesai biofiltre. Analogiškai gautiems tyrimų rezultatams, didžiausias aerodinaminis pasipriešinimas (iki 156 Pa) bei oro srauto greitis (iki 2,0 m/s) filtre susidaro prieš 5 jo įkrovos sluoksnius. Naudojant MAPLE 7, DATAFIT 6.1.10 programas, filtro biologiškai suaktyvintoje įkrovoje modeliuoti biocheminiai procesai. Remiantis tyrimų rezultatais bei įvertinus vykstančius fizikinius, cheminius ir biologinius procesus, nustatytos teršalų, jų mišinių biodegradacijos α ir absorbcijos gebos β konstantos bei gautos jų priklausomybės.

Gauto patento principu sukurtas bei pagamintas biofiltras su suaktyvinta pušų žievių įkrova bei įvertintos jo, kaip nebrangaus ir efektyvaus oro valymo įrenginio, panaudojimo galimybės orui valyti nuo lakiųjų organinių junginių įvairiose Lietuvos pramonės (chemijos, naftos, baldų, maisto ir kt.) įmonėse.

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