AIR FILTER FOR REMOVING ALDEHYDE-LIKE VOCs FROM INDOOR AIR

Technical field

The present invention relates to an air filter, specifically an air filter cartridge for an air-purifying device, for reducing, substantially eliminating or cleansing contaminating agents from indoor air, suitable for use in residential areas, hospitals, leisure areas and work-space areas. The filter comprises a plurality of filament-like elements, one or more natural polyphenols and a catalytic agent.

The air filter relates specifically to a filter suitable for filtering aldehyde-like VOCs (Volatile Organic Compounds), wherein the main aldehyde-like VOC to be filtered from indoor air is formaldehyde (HCHO, methanal).

Furthermore, the disclosed air filter may also filter amine-like VOCs and ammonia, as well as imines and aminals as a result of their reaction with aldehydes, from indoor air.

Throughout this document, the acronym “VOC” or “VOCs” may be used instead of the term “Volatile Organic Compounds”.

The term “filament-like element” should be understood herein as a slender thread-like object or fibre, also including narrow or thin strips of an elongated material, such as tape, acting as a support or carrier for particles.

State of the Art

In the present state of the art, there exists a wide range of air filters available in the market capable of filtering VOCs containing an aldehyde functional group, mainly formaldehyde, such as those disclosed in patent nº US 2016/228811, in which the filter comprises a solid amine filtering medium made up of a liquid amine and one or more granular solid support materials (including silica, clay, alumina, carbon, polymer, fibre or combinations thereof) arranged in one or more filter sheets, providing an interaction between the amines present in the filter and the formaldehyde present in the air, flowing through said filter medium.

Patent nº WO 2017/114687 discloses a multi-layer air filter made up of a first layer including a catalyst, a second layer including amines and an optional third layer, which is made up of a water absorbing and releasing material, such as a hygroscopic material.

Polyphenols, such as resorcinol, are known to react with aqueous solutions of formaldehyde (known as formol or formalin) by heating under basic conditions, since the beginnings of the

polymer chemistry. Phenol–formaldehyde (PF) polymers also known as phenolic resins, in general, and resorcinol–formaldehyde resins in particular (known as resoles), are found in many industrial products and are formed by step-growth polymerization. This known reaction conveniently and innovatively developed, allows one to trap formaldehyde gas at ppm (parts per million) concentrations.

The present invention aims to improve and solve certain issues regarding the air filters available in the market by adding one or more polyphenolic substances and a catalytic agent or catalyst, both in a solid state, to a plurality of filament-like elements, which then react with aldehyde-like VOCs, mainly formaldehyde, and amine-like VOCs present in an air-flow going through the filter, to substantially reduce the quantity of aldehyde-like and amine-like VOCs present through absorption, therefore improving the quality of indoor air.

Document US 2013/052113 discloses a method for purifying air and an air purifier used for purifying air, wherein the air purifier comprises a filter with a fibrous structure having a photocatalytic action coating, and a fan or turbine which forces air to pass through the filter, removing VOCs. The air purifier may further comprise a scavenger media, such as phenolic compounds (e.g. resorcinol).

Document CN 107321328 discloses a VOC adsorbent and a process for obtaining such a VOC adsorbent, whose composition includes polyvinyl alcohol, water, plant polyphenols, kaolin, acidic liquid and a defoamer.

Document JPH11-226100 discloses a filter containing a formaldehyde absorbent, i.e. plant polyphenols, specifically tea-derived polyphenols such as flavan-3-ol and/or flavan-3-ol derivatives.

Brief description of the invention

The present invention, as stated previously, aims to improve the air quality of indoor air by reducing, filtering or cleaning aldehyde-like VOCs, and in certain embodiments amine-like VOCs and ammonia, from said indoor air, specifically formaldehyde which is a critical indoor pollutant, as well as certain odours caused by other aldehyde-like VOCs, as well as ammonia and amine-like VOCs.

Formaldehyde is highly reactive as well as toxic, allergenic and carcinogenic, and when it is present in indoor air at certain levels it poses a significant safety hazard, leading to multiple health problems. Formaldehyde in concentrations above 0.1 ppm (parts per million) can cause eye irritation while inhaling said air can also cause headaches, throat irritation and shortness

of breath (SOB). Formaldehyde present in indoor air in a concentration equal to or above 5 ppm may lead to death.

Even though formaldehyde and its derivatives pose all these health risks, they are still widely used in a wide variety of applications such as: i) resins and adhesives, used during flooring and carpeting or in the process of fabricating furniture, among others, wherein these industrially prepared resins and adhesives, once applied to a building, may gradually release traces of formaldehyde into the indoor air, a process which is accelerated when exposed to heat, e.g. owing to a heating source (radiator) located nearby or to sunlight, ii) a disinfectant, when present in an aqueous solution used mainly in operating theatres and other rooms in hospitals or cleanrooms, as it kills most bacteria and fungi that may be present, or iii) preservation aqueous solutions, known as formalin or formol, for biological organs in hospitals, museums and biology laboratories.

Therefore, the present invention discloses an air filter cartridge for use in an air-purifying device for removing aldehyde-like VOCs, and in certain embodiments amine-like VOCs, from indoor air. The air filter cartridge is made up of a casing containing a plurality of filament-like elements as a support; integrated in said support there is a mixture of one or more natural polyphenols and a catalyst or catalytic agent, present in the support as a finely divided powder, forming a sponge-like mesh.

The air filter acts as an absorption filter, the sponge-like mesh reacting irreversibly with the aldehyde-like VOCs of the indoor air, generating a polymer in the form of a polyphenol–aldehyde resin inside the air filter. Due to this irreversible reaction, the air filter is capable of absorbing very rapidly any trace of formaldehyde present in indoor air, as well as (although not so rapidly) other aldehyde-like VOCs, which are less reactive, volatile and toxic than formaldehyde.

In certain embodiments of the air filter, by increasing the proportion of the sulfonic acid component that is added to the sponge-like mesh, the filter is also capable of absorbing significant amounts of amine-like VOCs and ammonia, as well as of imines and aminals arising from the reaction of amines with aldehydes.

The filament-like elements, used as the support for the mixture of the finely divided powder of one or more natural polyphenols and the catalyst, are of organic or inorganic nature, selected from amongst a group comprising inorganic fibres, synthetic and semisynthetic fibres, natural fibres and mixtures thereof.

The inorganic fibres are preferably selected from a group comprising glass wool, rock wool, ceramic wool, and mixtures thereof.

On the other hand, synthetic and semisynthetic fibres are preferably selected from a group comprising polyesters, polyamides such as nylons, polypropylenes, their copolymers and mixtures thereof.

Finally, the natural fibres are preferably selected from a group comprising wool, silk, hair, collagen, keratin, cellulose, lignin, and mixtures thereof.

Regarding the one or more natural polyphenols which may be added to the support of the filter, as a finely divided powder, they are preferably selected from a group comprising resveratrol (3,4',5-trihydroxystilbene, a natural product very abundant in the peel of red grapes and in red wine), resorcinol (1,3-benzenediol), pyrogallol (1,2,3-benzenetriol), phloroglucinol (1,3,5-benzenetriol), and hydroquinone (1,4-benzenediol) or a combination thereof.

The polyphenols included in the support of the filter, inside the cartridge, are selected for removing aldehyde-like VOCs present in the indoor air, including but not limited to: acetaldehyde or ethanal, glyoxal or ethanedial, propionaldehyde or propanal, acrolein or propenal, propargyl aldehyde or propynal, methylglyoxal or 2-oxopropanal, glyoxylic acid and their alkyl esters, butyraldehyde or butanal, isobutyraldehyde or 2-methylpropanal, methylacrolein or 2-methylpropenal, both isomers of crotonaldehyde or 2-butenal, valeraldehyde or pentanal and isovaleraldehyde or 3-methylbutanal, and particularly formaldehyde (HCHO, methanal). Longer-chain aldehydes (i.e., aldehydes of higher molecular weight than those mentioned), such as those used in the cosmetic industry, may also be slowly absorbed, but they have a pleasant odour and their capture is not deemed as necessary.

These aldehyde-like VOCs may contaminate indoor air as a consequence of the reaction of olefins, such as unsaturated hydrocarbons from the petroleum industry and several essential oils and natural perfumes used as air fresheners, with ozone, coming from the outside or generated internally by laser printers, photocopiers, ionic air purifiers, arc welders or motor brushes. Atmospheric oxidation of ethanol vapour (from alcoholic drinks) and other short-chain primary alcohols (from perfumes) may also produce aldehyde-like VOCs.

In a preferred embodiment the preferred polyphenol is resveratrol, whereas in another preferred embodiment the word "polyphenol" refers to a mixture of resveratrol and resorcinol in a proportion between 10% and 90% of resveratrol and between 10% and 90% of resorcinol.

In addition to the powder of one or more polyphenols included in the filter support, made up of a plurality of filament-like elements, the support also includes an acid catalytic agent, preferably

a solid sulphonic acid also added as a finely divided powder. The sulphonic acid used in the filter is preferably selected from a group comprising:

- any arenesulphonic acid (Ar-SO_3H), such as *p*-toluenesulphonic acid ($TsOH \cdot H_2O$ or TsOH), or benzenesulphonic acid (BsOH),
- any alkanesulphonic acid (R–SO_3H), such as 10-camphorsulphonic acid (CSA),
- any sulphonic acid resin, such as strong-acid ion-exchange resins, or sulphonic acid polymers, added as a finely divided powder,
- thymol blue in its acidic form and related triphenylmethane-sulfonic dyes,
- food dyes (such as Allura Red, E129, or Ponceau 4R, E124, and related azo derivatives) in their acidic form, or
- any long-chain alkyl hydrogen sulphate or aryl hydrogen sulphate, RO–SO_3H or ArO–SO_3H, respectively, such as common detergents and surfactants in their acidic form.

The present invention further discloses a method describing how the contaminating agents present in the indoor air react with the air filter disclosed previously, wherein the method for removing aldehyde-like VOCs, and in certain embodiments amine-like VOCs and ammonia, from indoor air is the result of establishing a controlled airflow of the contaminated indoor air to be passed through the air filter so that the aldehyde-like VOCs react with powdered natural polyphenols, preferably resveratrol or mixture of resorcinol and resveratrol, in the presence of the catalytic agent, preferably a solid sulphonic acid, both supported on a plurality of filament-like elements, and through said reaction generating a polymer (a polyphenol–aldehyde resin), which is retained in the filter. In those embodiments where the air filter is also capable of filtering amine-like VOCs and ammonia, as well as imines and aminals due to their reaction with aldehyde-like VOCs, these are captured in the filter due to the presence of the sulphonic acid added as a powder.

The reactions that take place in the filter, owing to the aldehyde-like VOCs present in the indoor air, will at some point reach a saturation level and will therefore be ineffective, and therefore the method further includes an additional step in which the air filter is removed and exchanged once the saturation level has been reached, as indicated by a visual indicator, in which the sponge-like mesh of the filter undergoes a colour change, in certain embodiments from white to a reddish-brown colour, depending on the level of saturation at any moment.

In certain embodiments of the air filter, the indicator is the result of the sulphonic acid-catalysed reaction of formaldehyde and related VOCs with the polyphenol or the polyphenol mixture included in the filter.

However, in other embodiments of the filter, the indicator may be the result of the change of colour of thymol blue and related dyes or of food dyes and related azo dyes, all of them used in their sulphonic acid forms, when the reaction of formaldehyde and related VOCs with the polyphenol or the polyphenol mixture used in the filter is completed, i.e. when the sulfonic groups had been neutralized by ammonia, amine-like VOCs, and/or derivatives of aldehydes and ammonia or amines

It will be understood that any range of values given may not be optimal in extreme values and may require adaptations of the invention to these extreme values, such adaptations being within reach of a skilled person.

Other features of the invention appear from the following detailed description of an embodiment.

Brief description of the Figures

The foregoing and other advantages and features will be more fully understood from the following detailed description of an embodiment with reference to the accompanying drawings, to be taken in an illustrative and non-limitative manner, in which:

- **FIG. 1** shows a first graph, which compares the quantity of formaldehyde (HCHO or CH_2O, methanal) absorbed by the filter (Y-axis) in relation to the quantity of HCHO that flows through said filter (X-axis); and
- **FIG. 2** shows a second graph that indicates the amount of HCHO absorbed by the filter during a first cycle (Y-axis) in relation to the time that has passed (X-axis) in which a flow of contaminated air with a concentration of 4.360 ppm of HCHO is introduced, equivalent to the graph shown in Fig. 1.

Detailed description of an embodiment

The graphs shown in **FIG. 1** and **FIG. 2**, discussed herein, show results obtained in a laboratory experiment performed under certain predetermined conditions, with an increasing exposure to a concentration of formaldehyde of up to approximately 4.4 ppm which far exceeds the recommended permissible exposure limit for humans during a prolonged period of time, usually set between 0.75 ppm and 1.0 ppm (taking into account that concentrations of approximately 0.1 ppm of formaldehyde can also be perceived as well as affect humans, e.g.

odours and/or irritation), and at a much smaller scale. These graphs only show the absorption of HCHO during a first cycle.

To simulate an air filter according to one embodiment of this invention, such as those used in an air-purifying device, which includes a sponge-like mesh made up of a plurality of filament-like elements and a mixture of polyphenols and a solid sulphonic acid used, respectively, in a w/w ratio of 2.0 : 1.0 : 0.2, a smaller sponge-like mesh with the same proportion is inserted in a sealed test tube and a flow of air containing an increased concentration, measured in ppm, of formaldehyde is passed through said sponge-like mesh within the test tube.

The conditions used to test the effectiveness of the air filter with regards to its ability to absorb HCHO was performed in an air column with a diameter of 1.0 cm filled with 120 mg of glass wool, 30 mg of resveratrol, 30 mg of resorcinol (60 mg of polyphenols) and 12 mg of $TsOH \cdot H_2O$, with a thickness of 4.0 cm, to simulate the sponge-like mesh of the air filter in a w/w ratio of 2.0 : 1.0 : 0.2.

Contaminated air is passed through the sponge-like mesh inserted in the column at an air flow rate of approximately 2.0 L/min and a speed of 0.42 m/s. The contaminated air that flows through the column has a concentration of formaldehyde of 4.360 ppm.

As shown in the graphs, for these conditions, the amount of HCHO absorbed or captured by the filter (sponge-like mesh) inside the column is at its most effective during the first 100-150 min, wherein the filter absorbs approximately 80% of HCHO present in the flow of air flowing through the filter. However, once this maximum percentage has been reached, the tendency of the slope shown in these graphs with regards to the amount of HCHO absorbed is gradually slower, indicating that the sponge-like mesh inside the column simulating the air filter is becoming saturated and therefore cannot absorb HCHO effectively.

In a preferred embodiment the casing of the air filter cartridge has the following dimensions: a length of 40 cm, a width of 40 cm and a height of 5 cm. However, these dimensions are non-limitative and may vary accordingly, so that the air filter cartridge can be inserted adequately into all kinds of air-purifying devices. Modifying the dimensions of the cartridge may vary the w/w ratio of the filament-like elements, the polyphenols, and the sulphonic acid in order for the air filter to function under optimum filtering conditions. In general, the different components that form the sponge-like mesh are used in a 2.0–8.0 : 1.0–2.0 : 0.2–2.0 w/w ratio, respectively. In a preferred embodiment the filament-like elements, the polyphenols, and the sulphonic acid are used in a 2.0 : 1.0 : 0.2 w/w ratio, respectively.

It has been proved through different tests that the proposed air filter can also be highly effective for the elimination of malodours caused by amines or NH_3 derivatives (urine smell). To this end it has also been noted that it would be favourable to increase the proportion of sulphonic acid used to the detriment of resorcinol. Thus, considering a typical composition as the one previously referred with the following proportions: 80% resveratrol, 10% resorcinol and 10% of sulphonic acid, the latter could be increased up to 20% when resorcinol being limited to 2% and resveratrol kept to 78%, only as an example, not limitative, providing an air filter highly effective for filtering aldehyde-like VOCs (Volatile Organic Compounds) and ad the same time removing referred malodours.

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It will be understood that various parts of one embodiment of the invention can be freely combined with parts described in other embodiments, even being said combination not explicitly described, provided there is no harm in such a combination.

CLAIMS

1. A filter cartridge for an air-purifying device for removing aldehyde-like VOCs from indoor air, comprising a casing containing a plurality of filament-like elements as a support, with one or more natural polyphenols and a catalyst or catalytic agent integrated in said support, wherein said plurality of filament-like elements are of an organic or inorganic nature, and wherein the filter acts as an absorption filter, reacting irreversibly with the aldehyde-like VOCs of the indoor air **characterised in that:**

- said one or more natural polyphenols are selected from a group comprising: resveratrol (3,4',5-trihydroxystilbene), resorcinol (1,3-benzenediol), pyrogallol (1,2,3-benzenetriol), phloroglucinol (1,3,5-benzenetriol), and hydroquinone (1,4-benzenediol) or a combination thereof, and
- said catalyst or catalytic agent is a solid sulphonic acid,

wherein a mixture of said one or more natural polyphenols and said catalytic agent are present, in the support, as a finely divided powder, forming a sponge-like mesh.

2. Filter according to claim 1, wherein the polyphenols included in the filter are selected for removing formaldehyde (HCHO, methanal) present in the indoor air.

3. Filter according to claim 1, wherein said solid sulphonic acid used as the catalytic agent is selected from a group comprising:

- any arenesulphonic acid (Ar-SO_3H), such as *p*-toluenesulphonic acid ($TsOH \cdot H_2O$ or TsOH), or benzenesulphonic acid (BsOH),
- any alkanesulphonic acid (R–SO_3H), such as 10-camphorsulphonic acid (CSA),
- any sulphonic acid resin, such as strong-acid ion-exchange resins, or sulphonic acid polymers, added as a finely divided powder,
- thymol blue in its acidic form and related triphenylmethane-sulfonic dyes,
- food dyes (such as Allura Red, E129, or Ponceau 4R, E124, and related azo derivatives) in their acidic form, or
- any long-chain alkyl hydrogen sulphate or aryl hydrogen sulphate, RO–SO_3H or ArO–SO_3H, respectively, such as common detergents and surfactants in their acidic form.

4. Filter according to claim 1, wherein the filament-supporting elements are:

- inorganic fibres selected from a group comprising glass wool, rock wool, ceramic wool, and mixtures thereof,

- synthetic and semisynthetic fibres selected from a group comprising polyesters, polyamides (nylons), polypropylenes, their copolymers and mixtures thereof,
- natural fibres selected from a group comprising wool, silk, hair, collagen, keratin, cellulose, lignin, and mixtures thereof, or
- mixtures thereof.

5. Filter according to any of the previous claims, wherein the filament-like elements, the polyphenols, and the sulphonic acid are used, respectively, in a 2.0–8.0 : 1.0–2.0 : 0.2–2.0 w/w ratio.

6. Filter according to claim 1 or 5, wherein the filament-like elements, the polyphenols, and the sulphonic acid are used, respectively, in a 2.0 : 1.0 : 0.2 w/w ratio.

7. Filter according to claim 1, 2, 5 or 6, wherein the polyphenol used is resveratrol.

8. Filter according to claim 1, 2, 5 or 6, wherein the polyphenols used are a mixture of resveratrol and resorcinol in a proportion between 10% and 90% of resveratrol and between 10% and 90% of resorcinol.

9. Filter according to claim 1, wherein the casing has the following dimensions:

- a length of 40 cm,
- a width of 40 cm, and
- a height of 5 cm.

10. Filter according to claim 2, wherein the filter for removing formaldehyde present in the indoor air, further removes aldehydes selected from a group comprising: acetaldehyde or ethanal, glyoxal or ethanedial, propionaldehyde or propanal, acrolein or propenal, propargyl aldehyde or propynal, methylglyoxal or 2-oxopropanal, glyoxylic acid and their alkyl esters, butyraldehyde or butanal, isobutyraldehyde or 2-methylpropanal, methylacrolein or 2-methylpropenal, both isomers of crotonaldehyde or 2-butenal, valeraldehyde or pentanal and isovaleraldehyde or 3-methylbutanal, as well as amine-like VOCs and ammonia.

11. A method for removing aldehyde-like VOCs, amine-like VOCs and ammonia from indoor air, wherein a controlled airflow of the contaminated indoor air is passed through an air filter comprising a plurality of filament-like elements as a support, with one or more natural polyphenols and a catalyst or catalytic agent integrated in said support, wherein said plurality of filament-like elements are of an organic or inorganic nature, **characterised in that** said aldehyde-like VOCs react with said one or more natural polyphenols in the presence of said catalytic agent, generating a polymer in the form of a polyphenol-aldehyde resin, which is

retained in the filter, and **in that** said amine-like VOCs and ammonia, as well as imines and aminals arising from their reaction with aldehyde-like VOCs, are captured in a sponge like-mesh of the filter due to the catalytic agent present in the filter,

wherein the one or more natural polyphenols are selected from a group comprising: resveratrol (3,4',5-trihydroxystilbene), resorcinol (1,3-benzenediol), pyrogallol (1,2,3-benzene¬triol), phloroglucinol (1,3,5-benzenetriol), and hydroquinone (1,4-benzenediol) or a combination thereof,

wherein said catalyst or catalytic agent is a solid sulphonic acid, and

wherein a mixture of said natural polyphenols and said catalytic agent are present, in the support made up of said plurality of filament-like elements, as a finely divided powder, forming said sponge-like mesh.

12. Method for removing aldehyde-like VOCS according to claim 11, wherein the aldehyde-like VOCs are selected from a group comprising: formaldehyde (HCHO, methanal), acetaldehyde or ethanal, glyoxal or ethanedial, propionaldehyde or propanal, acrolein or propenal, propargyl aldehyde or propynal, methylglyoxal or 2-oxopropanal, glyoxylic acid and their alkyl esters, butyraldehyde or butanal, isobutyraldehyde or 2-methylpropanal, methylacrolein or 2-methylpropenal, both isomers of crotonaldehyde or 2-butenal, valeraldehyde or pentanal and isovaleraldehyde or 3-methylbutanal.

13. Method for removing aldehyde-like VOCs, amine-like VOCs and ammonia according to claim 11, wherein the air filter is removed and substituted once an indicator of the filter has reached a saturation level.

14. Method for removing aldehyde-like VOCs, amine-like VOCs and ammonia according to claim 13, wherein said indicator operates as a result of the sulphonic acid-catalysed reaction of formaldehyde and related VOCs with the polyphenol or the polyphenol mixture included in the filter.

15. Method for removing aldehyde-like VOCs, amine-like VOCs and ammonia according to claim 13, wherein said indicator operates as a result of the change of colour of thymol blue and related dyes or of food dyes and related azo dyes, all of them used in their sulphonic acid forms, when the reaction of formaldehyde and related VOCs with the polyphenol or the polyphenol mixture used in the filter is completed.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2019/068116

A. CLASSIFICATION OF SUBJECT MATTER
INV. B01D53/04 F24F3/16
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
B01D F24F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2013/052113 A1 (MOLINS LAURENT [FR] ET AL) 28 February 2013 (2013-02-28) figure 1 paragraphs [0001], [0003], [0020], [0036] - [0038], [0056], [0060]	1-15
A	CN 107 321 328 A (DONGGUAN TASEN ENV ENGINEERING CO LTD) 7 November 2017 (2017-11-07) abstract claim 1	1-15
A	JP H11 226100 A (MITSUI NORIN KK) 24 August 1999 (1999-08-24) paragraphs [0006], [0013] claim 5	1-15

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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Date of the actual completion of the international search	Date of mailing of the international search report
2 October 2019	11/10/2019

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2019/068116

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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