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(54) Title: TREATMENT OF TOBACCO MATERIAL

Figure 1

Polyphenol	Chemical Structure
Scopoletin	
Caffeic acid	
Chlorogenic acid	
Rutin	

(57) Abstract: A method is provided for treating a liquid tobacco extract, wherein the method comprises contacting the tobacco extract with an ion exchange resin to reduce the polyphenol content. Also provided is a tobacco extract which has been treated by this method; a method for reducing the polyphenol content of tobacco material; a tobacco material which has been treated by this method, or a derivative thereof; and a smoking article which comprises this tobacco material.

Treatment of Tobacco Material

Field of the Invention

The present invention relates to a method for the treatment of tobacco material.

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Background

In some circumstances, it may be desirable to reduce the content of certain constituents from tobacco material before incorporating the tobacco material into a smoking article such as a cigarette.

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Summary

According to a first aspect, there is provided a method for treating a liquid tobacco extract, wherein the method comprises contacting the tobacco extract with an ion exchange resin to reduce the polyphenol content.

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In some embodiments, the method comprises contacting the tobacco extract with an ion exchange resin to reduce the chlorogenic acid content.

In some embodiments, the method removes less than 50% of nicotine from the tobacco extract. In some embodiments, the method removes less than 50% of sugars from the tobacco extract.

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In some embodiments, the method further comprises membrane filtration to reduce the protein content of the tobacco extract. In some embodiments, membrane filtration is carried out with a Molecular Weight Cut-Off of not more than 500 kDa. In some embodiments, membrane filtration is ultrafiltration.

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In some embodiments, the method further comprises: treating the tobacco extract with one or more enzymes; treating the tobacco extract with one or more surfactants; and/or treating the tobacco extract with one or more adsorbents.

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According to a second aspect, there is provided a tobacco extract which has been treated by a method according to any one of the preceding claims.

According to a third aspect, there is provided a method for reducing the polyphenol content of tobacco material, the method comprising: extracting components from

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tobacco material with a solvent to form a liquid extract and a tobacco residue; contacting the liquid extract with an ion exchange resin to form a treated extract; and, combining the treated extract with the tobacco residue.

- 5 According to a fourth aspect, there is provided a method for reducing the polyphenol content of tobacco material, the method comprising: extracting components from tobacco material with a solvent to form a liquid extract and a tobacco residue; treating the liquid extract by a method according to the first aspect; and combining the treated extract with the tobacco residue.

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According to a fifth aspect, there is provided a tobacco material which has been treated by a method according to the third or fourth aspects, or a derivative thereof.

- 15 According to a sixth aspect, there is provided a smoking article which comprises a tobacco material according to the fifth aspect, or a derivative thereof.

According to a seventh aspect, there is provided the use of an ion exchange resin for removing one or more polyphenols from a liquid tobacco extract.

20 **Brief Description of the Drawings**

Embodiments of the invention will now be described, by way of example only, with reference to the accompanying drawings, in which:

- 25 Figure 1 shows the chemical structure of some polyphenol compounds which may be removed from tobacco material.

Figure 2 shows the apparatus used in the ultrafiltration experiments.

Figure 3 shows a graph of protein retention, and flow rate, against Molecular Weight Cut-Off during membrane filtration.

- 30 Figure 4 shows how the specific flow rate varied over time during ultrafiltration experiments with a 50 kDa membrane and a 15 kDa membrane.

Figure 5 shows the apparatus used in the ion exchange experiments.

Figure 6 shows how the effluent concentrations of polyphenols and chlorogenic acid varied with the number of Bed Volumes during ion exchange experiments with the first feed tobacco extract.

Figure 7 shows how the effluent concentrations of polyphenols and chlorogenic acid varied with the number of Bed Volumes during ion exchange experiments with the second feed tobacco extract.

Figure 8 is a schematic side view of a smoking article including treated tobacco material according to embodiments of the invention.

Detailed Description

Tobacco material often undergoes treatment for the purpose of removing polyphenol compounds from the material. Methods which are known to effectively remove polyphenols are, however, not particularly selective in their removal. The present invention, however, comprises the use of an ion exchange resin which may, advantageously, facilitate the effective and selective removal of polyphenol compounds from tobacco material.

The methods disclosed herein may be applied to any suitable tobacco material. The tobacco material may be derived from any suitable part of any suitable tobacco plant of the plant genus *Nicotiana*. The tobacco material may then be treated in any suitable way, and may be cured using any suitable method of curing, before being treated according to the method of the invention. In some embodiments, the tobacco material treated by the method of the invention has already been cured and may be cured cut rag and/or cured whole leaf tobacco. Examples of tobaccos which may be used in the method of the invention include, but are not limited to Virginia, Burley, Maryland, Oriental, and Rustica.

The methods disclosed herein lead to a reduction in the polyphenol content of tobacco material compared to the polyphenol content of the untreated tobacco material. This is likely to be advantageous because tobacco material is often incorporated into smoking articles and polyphenol compounds are often found to be, and/or to react during the course of smoking to form substances which are, undesirable for human inhalation.

In some embodiments, the treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin results in a reduction in the content of one or more polyphenols of at least 5%, 10%, 15%, 20%, 25%, 30%, 35%, 40%, 50%, 60%, 70%, 75%, 80%, 85%, 90%, or at least 95%, based upon the polyphenol content of the untreated tobacco extract.

In some embodiments, the treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin results in the extraction of one or more polyphenols in an amount of at least 5%, 10%, 15%, 20%, 25%, 30%, 35%, 40%, 50%, 60%, 70%, 75%, 80%, 85%, 90%, or at least 95%, based upon the polyphenol content of the untreated tobacco material.

Polyphenols which may be removed by the treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin include, but are not limited to: chlorogenic acid, caffeic acid, rutin, scopoletin, and quercetin. Figure 1 illustrates the chemical structure of these polyphenol compounds.

Chlorogenic acid is the most abundant polyphenol in tobacco material and therefore, in some embodiments, it may be desirable to reduce the content of chlorogenic acid in particular. In some embodiments, therefore, the method results in a reduction in the content of chlorogenic acid of at least 5%, 10%, 15%, 20%, 25%, 30%, 35%, 40%, 50%, 60%, 70%, 75%, 80%, 85%, 90%, or at least 95%, based upon the chlorogenic acid content of the untreated tobacco material.

In addition to reducing the polyphenol content of tobacco material, the treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin may also modify the tobacco material in any other suitable way. The tobacco material may be incorporated into a smoking article and, in this case, may be modified by the method in order to provide it with characteristics which would be desirable for a tobacco material used in this way. For example, the tobacco material may be treated for the purpose of removing one or more chemical substances besides polyphenol compounds.

In some embodiments, it may be desirable to remove one or more other chemical substances. It may be considered desirable to remove proteins from tobacco material. In some embodiments, therefore, treatment of a liquid tobacco extract may involve the removal of one or more proteins, such as RuBisCO, from the tobacco material.

In some embodiments, the treatment of the tobacco extract by contacting the tobacco extract with an ion exchange resin results in a reduction in the protein content of at least 5%, 10%, 15%, 20%, 25%, 30%, 35%, 40%, 50%, 60%, 70%, 75%, 80%, 85%, 90%, or at least 95%, based upon the protein content of the untreated tobacco extract.

In some embodiments, the treatment of the tobacco extract by contacting the tobacco extract with an ion exchange resin results in the extraction of protein in an amount of at least 5%, 10%, 15%, 20%, 25%, 30%, 35%, 40%, 50%, 60%, 70%, 75%, 80%, 85%, 90%, or at least 95%, based upon the protein content of the untreated tobacco material.

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In some embodiments, the treatment of a liquid tobacco extract by contacting the tobacco extract with an ion exchange resin reduces or minimises the removal of at least some of the chemical substances whose removal would be undesirable, which could be the case for a variety of different reasons. One reason, for example, could be that the substance makes a positive contribution to the experience of smoking a smoking article which contains the treated tobacco material.

In some embodiments, it may be undesirable for the treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin to remove nicotine. In some embodiments, the treatment removes less than 50%, 40%, 30%, 20%, 10%, or 5% of nicotine from the tobacco material; in further embodiments, the treatment removes less than 5%, 3%, 2%, 1%, 0.5%, or 0.1% of nicotine from the tobacco material; and, in further embodiments still, the treatment removes essentially no nicotine from the tobacco material.

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In some embodiments, it may be undesirable for the treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin to remove sugars and/or any other form of carbohydrate. In some embodiments, the treatment removes less than 50%, 40%, 30%, 20%, 10%, or 5% of sugars from the tobacco material; in further embodiments, the treatment removes less than 5%, 3%, 2%, 1%, 0.5%, or 0.1% of sugars from the tobacco material; and, in further embodiments still, the treatment removes essentially no sugars from the tobacco material.

In some embodiments, the treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin removes a large quantity of polyphenol compounds, a large quantity of protein compounds, a small quantity of nicotine, and a small quantity of sugars from the tobacco material. For example, the treatment may result in a reduction in the content of one or more polyphenols of at least 50% based upon the polyphenol content of the untreated tobacco material, and a reduction in the protein content of at least 50% based upon the protein content of the untreated tobacco

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material, while at the same time removing essentially no nicotine and essentially no sugars from the tobacco material.

5 In embodiments wherein the treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin leads to the removal of one or more chemical substances from the tobacco material, one or more of these may be re-introduced into the material following treatment, and one or more of these may be substances whose removal would be undesirable, such as nicotine.

10 In order to modify the tobacco material in any of the aforementioned ways, the treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin may comprise any suitable steps and any suitable number of steps, one or more of which comprise treating the tobacco material with an ion exchange resin.

15 The treatment of a tobacco extract by contacting the tobacco extract with an ion exchange resin may first comprise an extraction process, in which tobacco material is treated with solvent to generate a tobacco extract (liquid phase) and an extracted tobacco (solid phase).

20 In the extraction process, some chemical substances leave the tobacco material and enter solution. Chemical substances which may leave the tobacco material and enter solution include, but are not limited to: polyphenols, proteins, nicotine, sugars, amino acids, pectins, and salts. In some embodiments, approximately 55% of the original tobacco material, by weight, may be extracted and dissolved in solution during the
25 extraction process.

In the extraction process, the tobacco material treated with solvent may be in the form of ground tobacco. This may be preferred because, in this form, tobacco material has a large surface area to volume ratio, from which it follows that a greater quantity of
30 substances may be dissolved and extracted from the tobacco material per unit volume of the tobacco material.

In the extraction process, the tobacco material may be treated with one or more solvents, and may be treated with one or more solvents any suitable number of times,
35 for any suitable length of time, under any suitable conditions.

In the extraction process, a solvent may be added to the tobacco material in any suitable ratio to form a mixture or slurry. In some embodiments, a solvent may be added to the tobacco material in a ratio of between 10:1 and 50:1, by weight; in further
5 and 30:1, by weight; and, in further embodiments still, a solvent may be added to the tobacco material in a ratio of between 15:1 and 20:1, by weight.

A solvent used in the extraction process may be non-aqueous or aqueous. A non-aqueous solvent used in the extraction process may be liquid or supercritical carbon
10 dioxide, for example.

An aqueous solvent used in the extraction process may be purified water prepared by any suitable purification method, such as distillation and/or de-ionization. Alternatively, an aqueous solvent used in the extraction process may be water mixed
15 with one or more miscible liquids, and/or comprising one or more chemical substances in solution and/or suspension.

In some embodiments, a solvent used in the extraction process is an aqueous solution, and may comprise: one or more alcohols, such as ethanol and methanol; one or more
20 metal salts, such as potassium hydroxide, sodium chloride, and magnesium chloride; and/or one or more surfactants, such as SDS. Suitable concentrations of these additives may range from 0% to 20% (v/v) in some embodiments, 0% to 15% (v/v) in further embodiments, and 0% to 10% (v/v) in further embodiments still. Exemplary concentrations of these additives are 1% or 10% (v/v) potassium hydroxide, 0.5% (v/v)
25 sodium chloride, 0.5% (v/v) magnesium chloride, and 1%, 1.5%, or 2% (v/v) SDS.

In some embodiments, the extraction process may be a two-step process, featuring a first step with the use of an organic solvent, and a second step with the use of one or more of the above aqueous solvents, such as aqueous potassium hydroxide.
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In the extraction process, once the tobacco material has been treated with solvent, the resulting mixture or slurry of tobacco material and solvent may be subjected to one or more intensive mechanical actions, such as blending, homogenising, high shear mixing, and sonication. Such mechanical actions may modify the tobacco material by direct
35 mechanical action and/or cavitation, in which partial vacuums are formed in the solvent as a solid body moves therethrough and further physical agitation results.

It may be advantageous to include one or more intensive mechanical actions in the extraction process. This is because, firstly, their inclusion may increase the quantity of polyphenol and/or protein compounds which are dissolved into the liquid phase from the tobacco material and, secondly, their inclusion may make it easier to separate the tobacco extract and extracted tobacco after extraction due to continuous agitation preventing settlement of the extracted tobacco at the base of the apparatus.

In the extraction process, the mixture or slurry of tobacco material and solvent may be prepared in a first tank (a tobacco “mix tank”) before being pumped into a second tank, such as a plug flow reactor or continuous stirred tank reactor.

The extraction process may be carried out at any suitable temperature, and the temperature may be constant or variable. In embodiments wherein extraction is carried out with the use of an aqueous solvent, any suitable temperature may be used, although, in some embodiments, the temperature lies within the range of about 15°C to 85°C or about 50°C to 70°C. In some especially embodiments, the temperature is about 60°C.

The extraction process may be carried out for any suitable length of time. In some embodiments, the length of time over which the extraction process is carried out is between 10 minutes and 2 hours. In some embodiments, the length of time over which the extraction process is carried is about 40 minutes.

After the extraction process, the tobacco extract (liquid phase) may be separated from the extracted tobacco (solid phase) using any suitable means of separation. For example, separation may involve any suitable filtration method, such as nanofiltration, microfiltration, and/or ultrafiltration; any suitable filtering medium pore size; and any suitable number of filtration steps. Alternatively or in addition, separation may involve centrifugation, any suitable centrifuge system, any suitable angular velocity, and any suitable number of centrifugation steps.

In embodiments wherein filtration is used to separate the tobacco extract from the extracted tobacco, the filter may become blocked with solid particles. In order to overcome this problem, a vibrating sieve may be used, and may be fitted with a mesh whose pore size is suitable for separating tobacco extract from extracted tobacco. In

some embodiments, the pores in the mesh may have a diameter between 10 μm and 50 μm , or 20 μm and 40 μm . In some embodiments, the pores in the mesh may have a diameter of about 15 μm , 20 μm , 25 μm , 30 μm , 35 μm , 40 μm , or 45 μm . In some embodiments, the pores in the mesh may have a diameter of about 40 μm .

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In some embodiments, after separation of the tobacco extract (liquid phase) and extracted tobacco (solid phase), the extracted tobacco may be washed in order to yield more tobacco extract, and may be washed any suitable number of times with any suitable solvent. In some embodiments, the extracted tobacco may be washed with the same solvent as was used in the extraction process.

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After the extracted tobacco has been washed one or more times, the mass of substances from the tobacco material dissolved in the tobacco extract will increase, this being because the tobacco material will have been treated with solvent twice, namely during the extraction process and during the washing process.

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In some embodiments, after the extracted tobacco has been washed one or more times, it may be dewatered in any suitable way. In some embodiments, for example, the extracted tobacco may be squeezed after washing, and the resulting tobacco extract may be in the form of a dewatered mat.

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It may be advantageous to dewater the extracted tobacco because the removal of moisture from the extracted tobacco may increase the extracted tobacco's capacity to reabsorb the tobacco extract at a later stage in the treatment process. In some embodiments, the liquid obtained during the dewatering process may be recycled and used as a solvent in the extraction and/or washing process.

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After extraction of the tobacco material, and after separation of the tobacco extract and extracted tobacco, the tobacco extract (liquid phase) and extracted tobacco (solid phase) may be treated individually.

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The tobacco extract (liquid phase) may be treated in any suitable way. In some embodiments, the tobacco extract is treated by a method which reduces the polyphenol and/or protein content of the tobacco extract. In some embodiments, the tobacco extract is treated by a method which does not significantly reduce the nicotine and/or sugar content of the tobacco extract. In some embodiments, the tobacco extract is

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treated by a method which reduces the polyphenol and/or protein content of the extracted tobacco and, in addition, does not significantly reduce the nicotine and/or sugar content of the tobacco extract.

- 5 The tobacco extract may be treated with an ion exchange resin, and may be treated with an ion exchange resin any suitable number of times. In embodiments wherein the tobacco material is treated with an ion exchange resin more than once, the same or different type of ion exchange resin, and the same or different conditions, may be used on each occasion.

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An ion exchange resin used in the treatment of a tobacco extract may be basic, acidic, and/or chelating; may be strongly or weakly basic/acidic; and may exchange any suitable ions with the tobacco extract. The exchanged ion or ions may be monoatomic or polyatomic; may be cationic, anionic, and/or zwitterionic; and may have any suitable
15 charge sign and magnitude, such as 1+, 1-, 2+, 2-, 3+, or 3-.

In some embodiments, an ion exchange resin used in the treatment of a tobacco extract is basic and exchanges anions with the tobacco extract. In some embodiments, an ion exchange resin used is basic and exchanges anions with one or more carboxylate anions
20 from the tobacco extract. And, in some embodiments, an ion exchange resin used is basic and exchanges anions with one or more polyphenol anions, such as the carboxylate anion of chlorogenic acid, from the tobacco extract.

In some embodiments, the process of ion exchange with the tobacco extract results in a
25 reduction in the content of one or more polyphenols of at least 5%, 10%, 15%, 20%, 25%, 30%, 35%, 40%, 45%, 50%, 60%, 70%, 75%, 80%, 85%, 90%, or at least 95%, based upon the polyphenol content of the untreated tobacco extract.

Advantageously, chlorogenic acid is the most abundant polyphenol in tobacco material
30 and readily forms an anion in aqueous solution by virtue of its acidic carboxy group, thereby enabling the removal a high percentage of polyphenols from tobacco extract by ion exchange. In some embodiments, the process of ion exchange with the tobacco extract results in a reduction in the content of chlorogenic acid of at least 5%, 10%, 15%, 20%, 25%, 30%, 35 %, 40%, 45%, 50%, 60%, 70%, 75%, 80%, 85%, 90%, or at least
35 95%, based upon the chlorogenic acid content of the untreated tobacco extract.

In some embodiments, an ion exchange resin is used which selectively exchanges its counter-ions with polyphenol anions, such as chlorogenic acid, from the tobacco extract. This means that the ion exchange resin adsorbs more polyphenols from the tobacco extract than it does any other type of chemical species and, in some
5 embodiments, may mean that at least 50%, 60%, 70%, 80%, 90%, 95%, 99%, or at least 99.9%, of the anions adsorbed from the tobacco extract are polyphenols.

For example, in some embodiments, the ion exchange resin may selectively exchange its counter-ions with polyphenol anions from the tobacco extract so that essentially no
10 nicotine and/or sugars are exchanged with the tobacco extract. Advantageously, nicotine is unlikely to form an anion in solution because it contains no acidic protons, and most sugars are unlikely to form anions in solution for the same reason, therefore making selective ion exchange more likely.

15 In some embodiments, an ion exchange resin used in the treatment process has a higher affinity for one or more polyphenols in the tobacco extract than it does for its counter-ions. In some embodiments, the ion exchange resin has a lower affinity for sugars and/or nicotine in the tobacco extract than it does for its counter-ions. In some
20 embodiments, the ion exchange resin has a higher affinity for one or more polyphenol species in the tobacco extract than it does for its counter-ions and, in addition, has a lower affinity for sugars and/or nicotine in the tobacco extract than it does for its counter-ions.

In some embodiments, an ion exchange resin used in the treatment process has a low
25 affinity for its counter-ions, thereby enhancing the kinetic and thermodynamic favourability of ion exchange with ions in the tobacco extract. For example, in some embodiments, the counter-ions of the ion exchange resin may be chloride anions and the ion exchange resin may have a low affinity for them.

30 An ion exchange resin used in the treatment process may comprise any suitable chemical structure. An ion exchange resin may comprise one or more natural chemical components, such as an inorganic zeolite, and/or one or more artificial chemical components, such as an acrylic or styrenic polymer.

35 In embodiments wherein an ion exchange resin comprises a polymer, the polymer may or may not be cross-linked. In some embodiments, a resin comprising a polymer which

is not highly cross-linked is preferred over a resin comprising a polymer which is highly cross-linked. This is because a polymer which is not highly cross-linked potentially has more sites available for ion exchange, and a higher total ion exchange capacity, than a polymer which is highly cross-linked.

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An ion exchange resin used in the treatment process may be a gel-type or a macroporous-type resin. In some embodiments, it may be advantageous to use a gel-type resin because gel-type resins tend to have higher total ion exchange capacities, and therefore longer run lengths, compared to macroporous-type resins.

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In some embodiments, an ion exchange resin used in the treatment process may have a cross-linked acrylic gel structure. This may be advantageous because acrylic-based resins are often flexible, and often provide superior physical stability and organic fouling resistance compared to conventional polystyrene-based resins.

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An ion exchange resin used in the treatment process may comprise any suitable functional groups for the adsorption of ions. An ion exchange resin may comprise functional groups which are weakly or strongly acidic, weakly or strongly basic, and/or chelating. In some embodiments, an ion exchange resin comprises weakly basic

20 functional groups; in further embodiments, an ion exchange resin comprises amine functional groups; and, in further embodiments still, an ion exchange resin comprises tertiary amine functional groups.

An ion exchange resin used in the treatment process may comprise ion exchange beads.

25 In embodiments wherein an ion exchange resin comprises ion exchange beads, the beads may be substantially spherical in shape and the diameters of the beads may form any suitable statistical distribution — with any suitable uniformity coefficient, any suitable range, and any suitable harmonic mean. In some embodiments, the diameters of the spherical beads are essentially the same and the uniformity coefficient is

30 approximately 1.0. In further embodiments, the diameters of the spherical beads fall within the range of about 0.25 mm to 1.2 mm. And, in further embodiments still, the harmonic mean diameter of the spherical beads falls within the range of about 0.50 mm to 0.75 mm.

35 In some embodiments, an ion exchange resin used in the treatment process is an AMBERLITE™ FPA53 or an AMBERLITE™ FPA55 resin. In some embodiments, an

AMBERLITE™ FPA55 resin may be preferred over an AMBERLITE™ FPA53 resin because it has a shorter rinse.

5 An ion exchange resin used in the treatment process may have any suitable total ion exchange capacity. In some embodiments, an ion exchange resin used has a high total ion exchange capacity. In some embodiments, an ion exchange resin used has a total ion exchange capacity of at least 1.0 eq l⁻¹, 1.1 eq l⁻¹, 1.2 eq l⁻¹, 1.3 eq l⁻¹, 1.4 eq l⁻¹, or at least 1.5 eq l⁻¹, in its free base form. In some further embodiments, an ion exchange resin used has a total ion exchange capacity of at least 1.6 eq l⁻¹, 1.7 eq l⁻¹, 1.8 eq l⁻¹, 1.9
10 eq l⁻¹, 2.0 eq l⁻¹, 2.5 eq l⁻¹, 3.0 eq l⁻¹, or 3.5 eq l⁻¹, in its free base form.

In some embodiments, the operating ion exchange capacity of an ion exchange resin used is at least 50%, 60%, 70%, or at least 80%, of the total ion exchange capacity. And, in some embodiments, the operating exchange capacity of an ion exchange resin used is
15 at least 90%, 95%, or at least 99%, of the total ion exchange capacity.

An ion exchange resin used in the treatment process may have any suitable moisture holding capacity, wherein the moisture holding capacity is the percentage of wet mass which is moisture in the free base form of the resin. In some embodiments, it may be
20 advantageous for an ion exchange resin to have a high moisture holding capacity because this may lead to a greater rate of ion exchange, a greater resistance to fouling, and a quicker regeneration process. In other embodiments, however, it may be advantageous for an ion exchange resin to have a low moisture holding capacity because this may lead to a greater total ion exchange capacity. In some embodiments,
25 an ion exchange resin may have a moisture holding capacity which lies within the range of about 40% to 80%, 50% to 70%, or 56% to 64%.

Ion exchange may be carried out at any suitable temperature, and the temperature may be constant or variable. In some embodiments, the temperature at which ion exchange
30 is carried out does not adversely affect the chemical structure of the resin and/or the ability of the resin to exchange ions. In some embodiments, this may mean that ion exchange is carried out at a temperature not higher than 50°C. In some embodiments, ion exchange is carried out at ambient temperature.

35 Ion exchange may be a batch process or a continuous process. In some embodiments, ion exchange is a continuous process and an ion exchange resin is contained in an ion

exchange column. This may be preferred because a continuous process of ion exchange can potentially exchange more ions than a batch process of ion exchange. This is because, when ion exchange reaches thermodynamic equilibrium in a batch, the batch must be replaced, but, when ion exchange reaches thermodynamic equilibrium in a column, the ion exchange resin may be regenerated *in situ*, in order to facilitate further ion exchange with relatively little disruption to the process.

An ion exchange resin used in the treatment process may undergo regeneration in an ion exchange column using any suitable regeneration process. In some embodiments, an ion exchange resin may undergo regeneration when it is at, or when it is close to, thermodynamic equilibrium with the permeating solution — that is, when it is no longer, or when it is very slowly, exchanging ions with the permeating solution.

Any suitable method may be used to determine when an ion exchange resin has reached thermodynamic equilibrium with a permeating solution. In some embodiments, the conductivity of the permeate may be measured at regular intervals, or continuously, in order to determine when ion exchange is no longer taking place and the system is at thermodynamic equilibrium. This is possible because the exchanged ions are likely to have different charge-carrying capacities, meaning that the permeate's conductivity is likely to change over time before thermodynamic equilibrium, but to remain constant once thermodynamic equilibrium has been reached.

Regeneration of an ion exchange resin may be carried out using any suitable solution based upon the nature or type of the ion exchange resin. A suitable solution may, for example, include a solution of strong or weak acid and/or strong or weak base. In embodiments wherein the ion exchange resin used in the treatment process is basic, regeneration may be carried out using one or more solutions of NaOH, Na₂CO₃, and/or NH₃.

Regeneration of an ion exchange resin may be carried out using co-flow and/or counter-flow regeneration, and may be carried out using any suitable flow rate and any suitable contact time. In some embodiments, the flow rate during regeneration lies within a range of about 1 BVh⁻¹ to 10 BVh⁻¹, or 2 BVh⁻¹ to 8 BVh⁻¹. In some embodiments, the contact time during regeneration lies within a range of about 20 minutes to 40 minutes, or 25 minutes to 35 minutes.

In some embodiments, a weakly basic resin may be regenerated in an ion exchange column with a solution of NaOH (2% to 4% molar concentration), at a flow rate of about 2 BVh⁻¹ to 8 BVh⁻¹, with a contact time of about 30 minutes.

- 5 In some embodiments, it may be advantageous to use a weakly basic ion exchange resin, this being because a weakly basic ion exchange resin can be regenerated with a weak base and/or small volume of basic solution.

10 Following regeneration, an ion exchange resin may undergo a slow rinse followed by a fast rinse. A slow rinse may be carried out using any suitable volume of liquid, which may be 2 BV, and may be carried out at any suitable flow rate, which may be the same as the regeneration flow rate. A fast rinse may be carried out using any suitable volume of liquid, which may lie within the range of about 4 BV to 8 BV, and may be carried out at any suitable flow rate, which may be 10 BVh⁻¹.

15 Ion exchange may be carried out in an ion exchange column at any suitable temperature, and the temperature may be regulated by the use of a jacketed column, such as a glass column, in which water at the desired temperature is circulated through the jacket.

20 Ion exchange may be carried out in an ion exchange column at any suitable flow rate. In some embodiments, the flow rate during ion exchange lies within the range of about 1BVh⁻¹ to 15BV h⁻¹, 2BV h⁻¹ to 12BV h⁻¹, or 3BV h⁻¹ to 10BV h⁻¹. In some embodiments, which may be preferred, the flow rate during ion exchange lies within the range of
25 about 4BV h⁻¹ to 8BV h⁻¹.

An ion exchange resin used in an ion exchange column may have any suitable Bed Volume (BV), and any suitable bed depth. In some embodiments, the bed depth of an ion exchange resin used in an ion exchange column in the treatment process is between
30 about 1m to 2m, and it may be preferred that the bed depth is no more than about 2m in order to prevent the pressure drop across the resin being especially high.

In some embodiments, an ion exchange resin leads to the removal of polyphenols, such as chlorogenic acid, and the number of Bed Volumes required until the effluent
35 concentration of polyphenols and/or chlorogenic acid from the ion exchange column is equal to the feed concentration of polyphenols and/or chlorogenic acid lies within the

range of about 5 BV to 40 BV, 5 BV to 30 BV, or 10 BV to 20 BV. In some embodiments, a higher value may be preferred over a lower value due a higher value meaning that less regenerant is required in order to regenerate the ion exchange resin in the ion exchange column.

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In some embodiments, in addition to undergoing ion exchange, and before or after undergoing ion exchange, the tobacco extract may undergo membrane filtration. In some embodiments, the tobacco extract undergoes membrane filtration before ion exchange. The tobacco extract may undergo membrane filtration any suitable number
10 of times and, in embodiments wherein the tobacco extract undergoes membrane filtration more than once, the same or different membrane and the same or different conditions may be used on each occasion.

In some embodiments, membrane filtration of the tobacco extract may result in a
15 reduction in the protein content of at least 20%, 25%, 30%, 35%, 40%, 45%, 50%, 55%, 60%, 65%, 70%, 75%, 80%, 85%, 90%, 95%, 99%, or at least 99.9%, based upon the protein content of the untreated tobacco extract. In some embodiments, membrane filtration of the tobacco extract removes essentially no nicotine and/or sugars from the tobacco extract. And, in some embodiments, membrane filtration of the tobacco extract
20 removes at least 95% of the protein from the tobacco extract, while at the same time removing essentially no nicotine and/or sugars from the tobacco extract.

Membrane filtration may be carried out with any suitable Molecular Weight Cut-Off (MWCO) value, wherein the MWCO is equal to the molecular weight of a molecule
25 which is 90% retained by the membrane during membrane filtration. The desired MWCO value may be obtained by modifying the conditions during membrane filtration in any suitable way, for example by adjusting the membrane pore size or adjusting the pressure drop across the membrane.

30 In some embodiments, membrane filtration may be carried out with an MWCO value which lies within the range of about 10 kDa to 500 kDa, 10 kDa to 300 kDa, 10 kDa to 150 kDa, 10 kDa to 100 kDa, or 15 kDa to 50 kDa. In some embodiments, membrane filtration may be carried out with an MWCO equal to about 15 kDa. In some
35 embodiments, an MWCO value of 15 kDa may be preferred because experiments, which are detailed below, have shown that an MWCO of 15 kDa results in the retention of 95

% of proteins from tobacco extract, while at the same time removing essentially no nicotine.

Any suitable type of membrane filtration may be used, such as ultrafiltration, microfiltration, and/or nanofiltration. In some embodiments, however, membrane
5 filtration used in the treatment of a tobacco extract is ultrafiltration. This is because ultrafiltration is often found to provide an MWCO range between about 10 kDa and 100 kDa, therefore being particularly suited to the filtration of proteins from solution.

10 The pressure drop across the membrane may have any suitable value during ultrafiltration, and may be constant or variable. In some embodiments, the pressure drop across the membrane may be approximately 150 kPa, 200 kPa, 250 kPa, 300 kPa, 350 kPa or 400 kPa (1.5 bar, 2 bar, 2.5 bar, 3 bar, 3.5 bar, 4 bar) or any suitable higher value. In some embodiments, the pressure drop across the membrane is 200 kPa (2
15 bar).

Membrane filtration may be carried out at any suitable flow rate and any suitable specific flow rate. In some embodiments, a higher flow rate and a higher specific flow rate are favoured over a lower flow rate and a lower specific flow rate in order to
20 increase the rate of filtration. In some embodiments, the flow rate may be lie within the range of about 10 ml min⁻¹ to 60 ml min⁻¹, or 15 ml min⁻¹ to 50 ml min⁻¹. And, in some further embodiments, the specific flow rate may lie within the range of about 100 l h⁻¹ m⁻² to 450 l h⁻¹ m⁻², or 120 l h⁻¹ m⁻² to 400 l h⁻¹ m⁻².

25 Membrane filtration may be carried out at any suitable temperature. In some embodiments, ultrafiltration may be carried out at ambient temperature or any other suitable temperature, such as 30°C, 40°C, 50°C, 60°C, 70°C, 80°C, or any suitable higher temperature. In some embodiments, ultrafiltration is carried out at 80°C.

30 In some embodiments, diafiltration may be used in membrane filtration. Diafiltration may be continuous or discontinuous and may comprise the use of any suitable solvent. In some embodiments, diafiltration is continuous and comprises the use of the same solvent that was used in the extraction process. Advantageously, diafiltration may minimise the removal of nicotine and/or sugars from the tobacco extract by minimising
35 the amount of substances which do not permeate the membrane despite being membrane-permeable.

In some embodiments, a membrane used may need to be cleaned in order to remove the substances retained on the filter (retentate). This may be required in order to prevent the retentate blocking the pores in the membrane, and to allow the filtration process to continue.

A membrane may be cleaned in any suitable way, under any suitable conditions, using any suitable chemical substance or substances. In some embodiments, a membrane may be rinsed or washed with one or more aqueous solutions.

10

A membrane used may be made of any suitable material or materials, which will be known to a person skilled in the art. In some embodiments, a membrane used in the treatment of a tobacco extract may be a ceramic membrane, such as an alumina membrane, titania membrane, or zirconia oxide membrane; or a polymer membrane, such as a polycarbonate membrane, a cellulosic membrane, or a cellulose acetate membrane. In some embodiments, a membrane used may be a ceramic membrane because ceramic membranes are often stable under extreme conditions, such as extreme temperature and/or pH.

15

A membrane used in the treatment of a tobacco extract may be in any suitable configuration. For example, a membrane used in the treatment process may have a flat, hollow-fibre, or spiral-wound configuration. A membrane used in the treatment process may be a standard membrane or, alternatively, may be a track-etched membrane. Furthermore, the membrane may be configured to facilitate membrane filtration in suspension or deposition mode.

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In an exemplary embodiment of the invention, which is shown in Figure 2, the tobacco extract may undergo ultrafiltration in deposition mode with the use of a ceramic membrane at an MWCO of 15 kDa. The tobacco extract may be heated to 80°C in a holding tank, before being continuously circulated by the action of a peristaltic pump through a membrane at a linear velocity of 5 m s⁻¹. The pressure drop across the membrane may be 200 kPa (2 bar), and the permeate may be recovered from the membrane after filtration. The feed tank may be continuously filled with tobacco extract in order to facilitate the continuous circulation and filtration of tobacco extract.

30
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In this exemplary embodiment of the invention, the membrane filter may be cleaned at regular intervals, and may be cleaned by, firstly, rinsing the membrane with purified water; secondly, treating the membrane with an aqueous solution of NaOH (1% molar concentration) at 80°C, for 15 minutes with the permeate valve (V3 in Figure 2) closed, and for 15 minutes with the permeate valve open; thirdly, rinsing the membrane with purified water until the pH is less than 9; fourthly, treating the membrane with an aqueous solution of HNO₃ (0.5% molar concentration) at 60°C, for 15 minutes with the permeate valve closed, and for 15 minutes with the permeate valve open; and, finally, rinsing the membrane with purified water until the pH is more than 5.

10

In some embodiments, the treatment of a tobacco extract may comprise treating the tobacco extract by both membrane filtration and ion exchange. In some embodiments, the tobacco extract may be treated by ultrafiltration before undergoing ion exchange and, in some further embodiments, ultrafiltration may be carried out with an MWCO value between about 15 kDa to 50 kDa, and ion exchange may be carried out with an ion exchange resin with a total ion exchange capacity of at least 1.0 eq l⁻¹.

15

Alternatively or in addition to undergoing ion exchange and/or membrane filtration, and before or after undergoing ion exchange and/or membrane filtration, the tobacco extract may be treated with one or more adsorbents, and may be treated with one or more adsorbents any suitable number of times. In embodiments wherein the tobacco extract is treated with one or more adsorbents more than once, the same or different adsorbents and the same or different conditions may be used on each occasion.

20

In some embodiments, one or more adsorbents used in the treatment of a tobacco extract have a high affinity for polyphenols and/or proteins. In some embodiments, one or more adsorbents used in the treatment process have a low affinity for nicotine and/or sugars. In some embodiments, one or more adsorbents used in the treatment process have a high affinity for polyphenols and/or proteins and, in addition, have a low affinity for nicotine and/or sugars.

25

30

In some embodiments, one or more of the adsorbents used in the treatment of a tobacco extract are hydrophobic. The use of one or more hydrophobic adsorbents may be preferred because they are likely to have a high affinity for polyphenol compounds due to the hydrophobic nature of the benzene ring(s) in polyphenol compounds.

35

Any suitable adsorbents may be used. Examples of suitable adsorbents include, but are not limited to: adsorbent resins; activated carbon; polyvinyl polypyrrolidone (PVPP), hydroxylapatite, bentonite; and attapulgite.

5 An adsorbent used in the treatment of a tobacco extract may have any suitable specific surface area. In some embodiments, however, an adsorbent used in the treatment process has a high specific surface area because this is likely to result in the adsorption of more substances, such as polyphenols and/or proteins, from the tobacco extract. In order to increase the specific surface area of an adsorbent used in the process, the
10 adsorbent may be particulate and/or may comprise pores, for example.

In embodiments wherein an adsorbent is an adsorbent resin, the adsorbent resin may comprise any suitable chemical functionality. In some embodiments, the adsorbent resin may comprise a natural chemical component, which may be an inorganic zeolite
15 for example, and/or an artificial chemical component, which may be any suitable polymer, such as an acrylic, styrenic, or phenolic polymer compound.

In embodiments wherein an adsorbent is an adsorbent resin, the adsorbent resin may comprise any suitable specific surface area. In some embodiments, the specific surface
20 area of an adsorbent resin used in the treatment of a tobacco extract is at least $100 \text{ m}^2 \text{ g}^{-1}$, $150 \text{ m}^2 \text{ g}^{-1}$, $200 \text{ m}^2 \text{ g}^{-1}$, $250 \text{ m}^2 \text{ g}^{-1}$, $300 \text{ m}^2 \text{ g}^{-1}$, $350 \text{ m}^2 \text{ g}^{-1}$, $400 \text{ m}^2 \text{ g}^{-1}$, or at least $450 \text{ m}^2 \text{ g}^{-1}$. In some embodiments, the specific surface area of an adsorbent resin used is at least $500 \text{ m}^2 \text{ g}^{-1}$, $550 \text{ m}^2 \text{ g}^{-1}$, $600 \text{ m}^2 \text{ g}^{-1}$, $650 \text{ m}^2 \text{ g}^{-1}$, $700 \text{ m}^2 \text{ g}^{-1}$, $750 \text{ m}^2 \text{ g}^{-1}$, or at least $800 \text{ m}^2 \text{ g}^{-1}$.

25 In embodiments wherein an adsorbent is activated carbon, the activated carbon may comprise any suitable fraction of micropores, mesopores, and/or macropores, and may have any suitable specific surface area. In some embodiments, the specific surface area of an activated carbon used in the treatment process is at least $2500 \text{ m}^2 \text{ g}^{-1}$.

30 In embodiments wherein an adsorbent is PVPP, any suitable weight of PVPP may be added to the tobacco extract. In some embodiments, for example, the weight of PVPP added to the tobacco extract may lie within the range 25% to 75%, or 45% to 55% of the weight of the untreated tobacco material. In some embodiments, this quantity of PVPP
35 may be capable of removing between 50% and 90% of the polyphenol compounds from the tobacco extract.

- In embodiments wherein the tobacco extract is aqueous, the pH of the tobacco extract may influence the capacity of one or more of the adsorbents to adsorb substances from the tobacco extract. The pH of the tobacco extract may therefore be modified to any
5 suitable pH value in order to enhance adsorption, and this could be achieved by the addition of one or more buffering agents. For example, the optimum pH at which PVPP adsorbs polyphenol compounds is often less than 5 and, therefore, in some embodiments the pH of the tobacco extract is less than 5 when treated with PVPP.
- 10 Treatment of the tobacco extract with one or more adsorbents may be a batch process of a continuous process. In some embodiments, treatment with one or more adsorbents is a continuous process and takes place in a column. This may be preferred because continuous adsorption in a column can potentially lead to the adsorption of more chemical substances than adsorption in a batch process. This is because, when
15 adsorption reaches thermodynamic equilibrium in a batch process, the adsorbent must be replaced, but, when adsorption reaches thermodynamic equilibrium in a column, the adsorbent may be chemically treated in the column and regenerated to facilitate further adsorption.
- 20 Alternatively or in addition to undergoing ion exchange, membrane filtration, and/or adsorbent treatment, and before or after undergoing ion exchange, membrane filtration, and/or adsorbent treatment, the tobacco extract may be treated in any other suitable way.
- 25 For example, in some embodiments, the tobacco extract may be treated with one or more enzymes and, in further embodiments, these enzymes may be capable of catalysing the modification of polyphenols or proteins. For example, one or more of the enzymes may be a phenol-oxidising enzyme, such as a laccase, or a proteolytic enzyme.
- 30 For example, in some embodiments, the tobacco extract may undergo electrophoresis, and may undergo electrophoresis any suitable number of times under any suitable conditions.

The extracted tobacco (solid phase) may be treated in any suitable way.

Some suitable ways in which the extracted tobacco may be treated are disclosed, for example, in European Patent Nos. 1094724, 0619708, and 0862865.

5 In some embodiments, the extracted tobacco is treated by a method which reduces the polyphenol and/or protein content of the extracted tobacco. In some embodiments, the extracted tobacco is treated by a method which does not significantly reduce the nicotine and/or sugar content of the extracted tobacco. In some embodiments, the extracted tobacco is treated by a method which reduces the polyphenol and/or protein content of the extracted tobacco and, in addition, does not significantly reduce the
10 nicotine and/or sugar content of the extracted tobacco.

In some embodiments, the extracted tobacco may be treated with one or more solvents, and may be treated with one or more solvents any suitable number of times under any suitable conditions. The extracted tobacco may be washed with one or more solvents in
15 order to remove one or more substances which were added during the extraction process, for example.

In some embodiments, the extracted tobacco may be treated with one or more enzymes and, in some embodiments, one or more of these enzymes may be capable of catalysing
20 the modification of polyphenols or proteins. For example, one or more of the enzymes may be a phenol-oxidising enzyme, such as a laccase, or a proteolytic enzyme.

After the tobacco extract (liquid phase) and the extracted tobacco (solid phase) have been treated according to any of the methods described above, the tobacco extract and
25 extracted tobacco may be recombined.

In some embodiments, the tobacco extract may be treated in order to increase its solids concentration before recombination. The solids concentration of the tobacco extract may be increased to any suitable value and, in some embodiments, the solids
30 concentration of the tobacco extract may be increased to between 20% and 50%, by weight.

The solids concentration of the tobacco extract may be increased using any suitable method. Solids concentrations of up to 10% by weight may be most efficiently achieved
35 by reverse osmosis, and solids concentrations of from 40% to 60% by weight may be achieved by means of a falling or rising film evaporator, for example.

In some embodiments, the solids concentration of the tobacco extract may be increased by evaporating liquid from the tobacco extract under vacuum. This method may be preferred because it is likely to remove the most volatile compounds from the tobacco
5 extract, and this may be beneficial because volatile compounds are most likely to affect the flavour and/or character of tobacco smoke.

After the solids concentration of the tobacco extract has been increased, the tobacco extract may be recombined with the extracted tobacco. Recombining the tobacco
10 extract with the extracted tobacco ensures that many of the soluble components of the untreated tobacco material, such as nicotine and sugars, are contained in the final tobacco product.

Any suitable method of recombination may be used. In some embodiments, the most
15 appropriate method to use may depend on whether, and how, the recombined tobacco is to be stored before further processing, for example to generate a reconstituted tobacco sheet.

In some embodiments, tobacco extract may be recombined with extracted tobacco by
20 the spraying of tobacco extract onto extracted tobacco.

Alternatively or in addition, the extracted tobacco and tobacco extract may be fed into a mixing tank and mixed with additional ingredients, such as fibre, humectants, binders, diluents, catalysts, and/or filtration substances. Mixing the extracted tobacco and
25 tobacco extract in this way may be used for the purpose of modifying the properties of the tobacco in any suitable way.

After the recombination process, in some embodiments, the tobacco material formed by the recombination process may be added to an untreated tobacco material, and may
30 be added to an untreated tobacco material in any suitable ratio, by weight.

In some embodiments, the tobacco material formed by the recombination process may be dried according to any suitable drying process. In some embodiments, the initial moisture content of the tobacco material may be 70% to 85%, while the final moisture
35 content of the tobacco material after drying may be approximately 14%.

A heated dryer, such as an apron dryer or a flash dryer, may be used to reduce the moisture content of the tobacco material. In some embodiments, the tobacco material may be treated by two or more dryers in order to reach the desired moisture content. For example, an apron dryer may be used to reduce the moisture content to 20% to
5 30%, before a flash dryer may be used to reduce the moisture content to approximately 14%.

In some embodiments, after the tobacco material has been dried, it may undergo agglomeration to form larger particles than the original grind or mill size. In some
10 embodiments, therefore, the dried tobacco material may be milled, and may be milled to generate a population of particles with any suitable mean diameter. In some embodiments, the mean diameter is less than about 2 mm; in some further embodiments, the mean diameter is less than about 1 mm, 500 μm , 400 μm , 300 μm , 250 μm , 200 μm , 150 μm or 100 μm ; and, in some further embodiments still, the mean
15 diameter is less than about 75 μm , or 50 μm . These mean diameter values may be preferred because they may make the population of particles more suitable for subsequent treatment processes.

Having undergone any of the previously-described treatment steps in accordance with
20 the treatment process, the tobacco material may be further modified in any suitable way before being incorporated into a smoking article. For example, certain chemical substances may be added to the tobacco material, such as flavourants where local regulations permit, and the tobacco material may be cut and/or shredded before being incorporated into a smoking article using any suitable method of incorporation.

25 As used herein, the terms "flavour" and "flavourant" refer to materials which, where local regulations permit, may be used to create a desired taste or aroma in a product for adult consumers. They may include extracts (e.g., licorice, hydrangea, Japanese white bark magnolia leaf, chamomile, fenugreek, clove, menthol, Japanese mint, aniseed,
30 cinnamon, herb, wintergreen, cherry, berry, peach, apple, Drambuie, bourbon, scotch, whiskey, spearmint, peppermint, lavender, cardamon, celery, cascarrilla, nutmeg, sandalwood, bergamot, geranium, honey essence, rose oil, vanilla, lemon oil, orange oil, cassia, caraway, cognac, jasmine, ylang-ylang, sage, fennel, piment, ginger, anise, coriander, coffee, or a mint oil from any species of the genus *Mentha*), flavour
35 enhancers, bitterness receptor site blockers, sensorial receptor site activators or stimulators, sugars and/or sugar substitutes (e.g., sucralose, acesulfame potassium,

aspartame, saccharine, cyclamates, lactose, sucrose, glucose, fructose, sorbitol, or mannitol), and other additives such as charcoal, chlorophyll, minerals, botanicals, or breath freshening agents. They may be imitation, synthetic or natural ingredients or blends thereof. They may be in any suitable form, for example, oil, liquid, or powder.

5

In some embodiments, the methods described herein may comprise one or more further steps to modify the tobacco material in any suitable way. For example, the tobacco material may be modified to provide it with one or more characteristics desirable for a tobacco material. For example, where the treated tobacco material is to be incorporated into a smoking article such as a cigarette, the tobacco material may be treated in order to modify the flavour it generates upon combustion, and/or may be treated in order to remove one or more of its chemical substances.

10

In an exemplary embodiment of the invention, a tobacco material is extracted with water at 60°C to form a tobacco extract (liquid phase) and an extracted tobacco (solid phase), before these two phases are separated by filtration.

15

The tobacco extract (liquid phase), firstly, undergoes ultrafiltration with a MWCO of 15 kDa to result in the removal of more than 90% of proteins from the tobacco extract, and secondly, undergoes continuous ion exchange with an exchange resin capable of selectively adsorbing polyphenols, to result in the removal of more than 90% of polyphenols from the tobacco extract.

20

The extracted tobacco (solid phase), meanwhile, undergoes solvent and enzyme treatment to reduce the content of polyphenols and/or proteins from the extracted tobacco.

25

The treated tobacco extract and treated extracted tobacco are then recombined by the spraying of the tobacco extract onto the extracted tobacco, before the recombined material is dried to a moisture content of approximately 14%. The tobacco product is then further modified in any suitable way before being incorporated into a smoking article.

30

As used herein, the term “smoking article” includes smokeable products such as cigarettes, cigars and cigarillos whether based on tobacco, tobacco derivatives, expanded tobacco, reconstituted tobacco or tobacco substitutes and also heat-not-burn

35

products. The smoking article may be provided with a filter for the gaseous flow drawn by the smoker.

Referring to Figure 8, for purpose of illustration and not limitation, a smoking article **1** according to an exemplary embodiment of the invention comprises a filter **2** and a cylindrical rod of smokeable material **3**, such as tobacco treated in accordance with the invention described herein, aligned with the filter **2** such that one end of the smokeable material rod **3** abuts the end of the filter **2**. The filter **2** is wrapped in a plug wrap (not shown) and the smokeable material rod **3** is joined to the filter **2** by tipping paper (not shown) in a conventional manner.

Experimental Work

A series of experiments were carried out in order to investigate the use of ion exchange, adsorbent treatment, and ultrafiltration for removing proteins and polyphenols from tobacco extract. The disclosed experimental work is not intended to limit the scope of the invention.

Tobacco Extract

The experiments were carried out with tobacco extract (liquid phase). The tobacco extract had been prepared by treating tobacco leaves with water above ambient temperature, before separating the liquid phase from the solid phase by filtration.

Two feeds of tobacco extract were tested in the experiments, and the compositions of the two feeds are detailed in Table 1. The compositions of the two feeds were similar, but one key difference was that the second feed was contained in a bucket with about 5% mud (i.e. solid material).

Table 1

Analyte (mg/l)	First Feed		Second Feed
Sample	A	B	
Nicotine	398	490	990
Reducing sugars	2006	3900	2900
Protein (as RuBisCO)	n.d.	1100	2240
Dry solid % (RI, sucrose scale)	n.d.	2.6	3.4
Colour (absorbency 420 nm)	n.d.	6.48	8.98
pH	n.d.	5.59	5.19

Total polyphenol	n.d.	2080	1880
Caffeic acid	5.6	n.d.	n.d.
Chlorogenic acid	149.0	326	380
Rutin	10.0	n.d.	n.d.
Scopoletin	1.8	n.d.	n.d.

Ultrafiltration

Ultrafiltration was carried out on both feeds, and the permeates collected in each of the experiments were analysed in order to identify which chemical substances, and to what extent certain chemical substances, were filtered. The purpose of the experiments was to identify which membrane may facilitate the removal of a large quantity of proteins and a small quantity of nicotine from tobacco extract.

Results were obtained for the experiments conducted on the first feed tobacco extract but were not obtained for the second feed tobacco extract due to the high solids content resulting in the blocking of the membrane and the prevention of continuous filtration.

The apparatus used in the ultrafiltration experiments are shown in Figure 2.

The experiments were conducted using these apparatus:

Tobacco extract was fed into a feed tank and heated to 80°C. The extract was circulated by the action of a peristaltic pump, and was pumped so that the liquid had a velocity of 5 m s⁻¹ when it reached the ceramic membrane in the membrane housing. A pressure of between 200 and 400 kPa (between 2 bar and 4 bar) was applied on one side of the membrane, and the permeate was collected on the other side. The retentate, consisting of membrane-impermeable molecules, was transported back to the feeding tank and recycled for further filtration. The process was continuous, with tobacco extract continuously fed into the feed tank and permeate continuously collected for analysis.

In order to prevent the membrane being blocked over time, it was cleaned at regular intervals by: firstly, rinsing the membrane with purified water; secondly, treating the membrane with an aqueous solution of NaOH (1% molar concentration) at 80°C, for 15 minutes with the permeate valve (V3 in Table 1) closed, and for 15 minutes with the permeate valve open; thirdly, rinsing the membrane with purified water until the pH was less than 9; fourthly, treating the membrane with an aqueous solution of HNO₃ (0.5% molar concentration) at 60°, for 15 minutes with the permeate valve closed, and

for 15 minutes with the permeate valve open; and, finally, rinsing the membrane with purified water until the pH was more than 5.

Membrane Selection

- 5 Experiments were first carried out to identify how protein filtration depends on the MWCO of the membrane, and to identify which MWCO value to investigate further.

The results of these experiments are shown in Table 2.

10 Table 2

Membrane cut-off	Protein retention	Average flow rate	Specific flowrate
15 KDa	>95%	15 ml/min	120 l/h.m ²
50 KDa	80%	17 ml/min	136 l/h.m ²
150KDa	50%	42 ml/min	336 l/h.m ²
500 KDa (0.1µm)	40%	50 ml/min	400 l/h.m ²

- The results indicate that protein retention in the membrane increases as the MWCO value of the membrane decreases. In particular, the results indicate that more than 95% of the proteins in the tobacco extract are retained in the membrane during ultrafiltration when the MWCO value of the membrane is 15 kDa.

- The results also indicate that the average flow rate and specific flow rate decrease as the MWCO value of the membrane decreases and the resistance to permeation increases.

- 20 The relationship between the level of protein retention and the MWCO value of the membrane, and the relationship between the average flow rate and the MWCO value of the membrane, are shown in Figure 3.

Further Investigation of Selected Membranes

- 25 The membranes with an MWCO value of 15 kDa and 50 kDa resulted in the retention of the greatest percentage of proteins from the tobacco extract, and for this reason were the membranes chosen for further investigation.

- 30 Further experiments were conducted with the first feed tobacco extract and the second feed tobacco extract.

The results obtained for the retention of different substances from the first feed tobacco extract are shown in Tables 3a and 3b. Table 3a provides the results for a membrane with an MWCO of 50 kDa; Table 3b provides the results for a membrane with an MWCO of 15 kDa.

5

Table 3a— 50 kDa Membrane Filtration

	BRIX	PH	Abs @420 nm	Polyphenol (g/l)	Proteins (g/l)	Sugars (g/l)	Nicotine (g/l)	Chlorogenic acid (mg/l)	Volume (ml)
Feed	2.50	5.48	5.82	2.06	0.86		0.49	360	3000
Aver. Permeate	2.30	5.48	4.70	2.01	0.46		0.49	326	2800
Yield in Permeate%	86%		75%	91%	50%		93%	85%	93%

Table 3b — 15 kDa Membrane Filtration

	BRIX	PH	Abs @420 nm	Polyphenol (g/l)	Proteins (g/l)	Sugars (g/l)	Nicotine (g/l)	Chlorogenic acid (g/l)	Volume (ml)
Feed	2.3	5.54	5.87	2.10	0.37		0.49	400	5000
Aver. Permeate	2.1	5.45	4.431	1.88	0		0.49	380	4500
Yield in Permeate%	85%		55%	81%	0%		90%	85%	90%

10

The 15 kDa membrane was found to remove 100% of the protein from the tobacco extract since no protein was detected in the permeate, while the 50 kDa membrane was found to remove only 50% of the protein from the tobacco extract since 50% of the protein was measured in the permeate.

15

Importantly, the BRIX value for both membranes was approximately 85%, and a BRIX value of this magnitude indicates that some liquid evaporated during the filtration process, and that a measured yield of 90% of a particular substance in the permeate suggests that no filtration of that substance has taken place. Consequently, the results in Tables 3a and 3b indicate that both of the membranes filtered essentially no nicotine from the tobacco extract, and that both of the membranes were selective for protein removal over nicotine removal.

20

From these results, it may therefore be concluded a membrane with an MWCO of 15 kDa is most effective for the removal of a large quantity of proteins and a low quantity of nicotine.

25

In addition to the filtration characteristics of the 50 kDa and the 15 kDa membranes being investigated, how their specific flow rates changed over time was also investigated. The results are shown in Figure 4.

5

As expected, the specific flow rate was found to decrease over time for both membranes, this being due to the collection of material on the membrane and the resulting increase in resistance. Furthermore, the specific flow rate was found to be higher for the 15 kDa membrane than for the 50 kDa membrane at all times, therefore providing another advantage of using a 15 kDa membrane rather than a 50 kDa for the selective filtration of protein from tobacco extract.

10

Adsorbent and Ion Exchange Treatment

Adsorbent and ion exchange treatment were carried out on both feeds after the feeds had undergone ultrafiltration with the use of a 15 kDa membrane. In each of the experiments, the concentrations of polyphenols and other substances were measured in the tobacco extract after adsorption had taken place. The collected results were then used to identify the most effective form of adsorption for the removal of a large quantity of polyphenol compounds and a small quantity of nicotine.

20

Two types of adsorbent — activated carbon and adsorbent resins — were tested, and ion exchange was tested with an ion exchange resin comprising chloride counter-ions. All of the adsorbents and ion exchange resins investigated in the experiments are detailed in Table 4.

25

Table 4

	label	Exchange Capacity or specific surface	Structure	comments
Adsorbent resins	ADS 1	800 m ² /g	Styrenic	High specific surface, small pores
	ADS 2	750 m ² /g	Styrenic	High specific surface, large pores
	ADS 3	380 m ² /g	Acrylic	Acrylic low affinity for small organic molecules
	ADS 4	150 m ² /g	phenolic	Phenolic, small pore size
Ion exchange resins	SB 1	1.2	Acrylic	Strong base acrylic chloride form
	SB 2	1.1	Styrenic	Strong base styrenic chloride form
	WB 1	1.6	Acrylic	Weak base acrylic high capacity chloride form
	WB 2	1.3	Acrylic	Weak base acrylic low capacity chloride form
Activated carbons	AC 1	>2500 m ² /g	Carbon	Phenol adsorption
	AC 2	>2500 m ² /g	Carbon	Phenol adsorption
	AC 3	>2500 m ² /g	Carbon	Sugar decolorization

The adsorbents and resins were first tested as part of a batch process.

Batch Tests

The batch tests were carried out at room temperature. A mixture of adsorbent and tobacco extract was stirred, and regularly analyzed until the concentration of substances in the tobacco extract remained steady and adsorption had essentially reached thermodynamic equilibrium.

For activated carbon, 100 ml of tobacco extract was mixed with 1 g of activated carbon. For the adsorbent resins and the ion exchange resins, 100 ml of tobacco extract was mixed with 20 g of resin. Less activated carbon was required because of the high specific surface area of the activated carbon tested in the experiments.

The results of the experiments for each of the adsorbent materials are provided in Tables 5a, 5b, and 5c. Table 5a relates to adsorbent resins; Table 5b relates to ion exchange resins; Table 5c relates to activated carbon. Each table indicates the fraction of certain substances which were adsorbed by each of the adsorbent materials in the experiments.

Table 5a

VARIATION Adsorbents Resins				
	ads 01	ads 02	ads 03	ads 04
nicotine%	97%	100%	50%	96%
sugars %	17%	14%	18%	24%
chlorogenic %	98%	38%	95%	86%
polyphenols %	88%	54%	62%	76%
dry solids %	38%	48%	29%	38%

Table 5b

VARIATION ANIONICS Resins				
	SB01	WB01	WB02	SB02
nicotine%	-2%	5%	2%	7%
sugars %	14%	16%	15%	22%
chlorogenic %	91%	84%	78%	82%
polyphenols %	56%	45%	48%	52%
dry solids %	27%	25%	26%	27%

Table 5c

VARIATION Activated Carbon			
	AC01	AC02	AC03
nicotine%	27%	29%	32%
sugars %	-8%	-9%	7%
chlorogenic %	56%	23%	-17%
polyphenols %	26%	17%	12%
dry solids %	9%	9%	9%

- 5 The fraction of each substance adsorbed in the experiments is expressed in the tables as a percentage calculated by:

$$X\% = 1 - (\text{final concentration of substance in tobacco extract}) / (\text{initial concentration of substance in tobacco extract})$$

10

Table 5a indicates that adsorbent resins can potentially remove a large quantity of polyphenols, and a large quantity of chlorogenic acid in particular, but that they also remove a large quantity of nicotine.

- 15 Table 5b indicates that ion exchange resins can potentially remove a large quantity of polyphenols, and a large quantity of chlorogenic acid, and that they can potentially remove essentially no nicotine.

20 Table 5c indicates that activated carbons do not remove a large quantity of polyphenols, and that they can remove a significant quantity of nicotine.

From these results, it may be concluded that the use of an ion exchange resin may be most appropriate for the purpose of removing a large quantity of polyphenols and a small quantity of nicotine. An ion exchange resin was therefore chosen for further
25 investigation. The weakly basic ion exchange resin (WBO1) was chosen in particular because it had the highest total ion exchange capacity of the tested ion exchange resins (see Table 4).

Continuous Tests

Continuous tests were carried out in an ion exchange column with the use of the ion exchange resin, WBO1, with the first and second feeds after they had undergone ultrafiltration with a 15 kDa membrane.

5 The apparatus used in the continuous tests are shown in Figure 5.

The experiments were carried out in the same way using these apparatus:

10 A feed of tobacco extract, after having undergone ultrafiltration, was loaded into the feed tank, before being pumped towards the ion exchange column. The effluent liquid was continually analysed after ion exchange. The total ion exchange capacity could be determined by calculating the number of Bed Volumes (BV) which needed to undergo ion exchange before the effluent concentration of chlorogenic acid was equal to the feed concentration of chlorogenic acid. The ion exchange resin was regularly regenerated by
15 chemical treatment.

Chlorogenic acid was measured in particular because it is the most abundant polyphenol in tobacco material and forms a carboxylate anion in aqueous solution which is able to undergo ion exchange with the WBO1 resin.

20

For the first feed tobacco extract, the concentration of chlorogenic acid in the effluent was found to equal the concentration of chlorogenic acid in the feed after 20 Bed Volumes (BV) of liquid had passed through the column. Figure 6 shows how the effluent concentration of chlorogenic acid increased with the number of Bed Volumes.
25 Figure 6 also shows how the effluent concentration of polyphenols increased with the number of Bed Volumes.

30

The results therefore indicate that the WBO1 anion exchange resin was able to remove chlorogenic acid from the first feed tobacco extract.

Table 6 shows the quantities of certain other substances in the effluent after certain numbers of Bed Volumes had passed through the column, and therefore indicates the amounts of these substances which were removed by the ion exchange resin. Most importantly, the results in Table 6 suggest that essentially no nicotine and essentially
35 no sugars were removed by the ion exchange column.

Table 6

	volume in (Bed Volume)	Conductivity (mS/cm)	polyphenols (g/l)	pH	brix	Sugars (g/l)	chlorogenic acid (g/l)	nicotine (g/l)
FEED	20	5.21	1.74	5.66	2.0	3.21	0.301	0.49
Outlet 0- 10 BV	10	8.30	0.99	5.74	1.74	2.71	0.02	0.45
Outlet 10-20 BV	10	8.06	1.33	4.65	1.79	2.71	0.18	0.50

For the second feed extract, the results are very similar and are shown in Figure 7 and

5 Table 7.

Table 7

	volume in (Bed Volume)	Proteins (g/l)	polyphenols (g/l)	pH	brix	Sugars (g/l)	chlorogenic acid (g/l)	nicotine (g/l)
FEED	20	0.07	1.88	5.08	1.7	2.93	0.350	0.95
Outlet 0- 10 BV	10	0.00	1.29	4.85	1.42	3	0.004	0.92
Outlet 10-20 BV	10	0.00	1.57	4.09	1.60	3.05	0.15	0.96

10 The results for both feeds show that ion exchange with the use of a weakly basic ion exchange resin can lead to the removal of a large quantity of chlorogenic acid and other polyphenols from tobacco extract, while at the same time removing essentially no nicotine from the tobacco extract. Furthermore, Figures 6 and 7 indicate that continuous ion exchange in a column can be more effective in the removal of

15 polyphenols than the equivalent batch process: the first 10 Bed Volumes in both cases show essentially complete removal of chlorogenic acid while the batch process with WBO1 removed only 84 % chlorogenic acid.

Conclusions

20 The results collected from the experiments suggest that ultrafiltration with a membrane MWCO of 15 kDa is most effective for the removal of proteins from tobacco extract, and that ion exchange in an ion exchange column is the most effective adsorbent method for the removal of chlorogenic acid and polyphenols from tobacco extract. Furthermore, the results suggest that these extraction processes may be combined, and that they may

25 lead to the removal of essentially no nicotine from the tobacco extract.

In order to address various issues and advance the art, the entirety of this disclosure shows by way of illustration various embodiments in which the claimed invention(s) may be practiced and provide for superior tobacco treatment, tobacco material, and products incorporating tobacco material. The advantages and features of the disclosure
5 are of a representative sample of embodiments only, and are not exhaustive and/or exclusive. They are presented only to assist in understanding and teach the claimed features. It is to be understood that advantages, embodiments, examples, functions, features, structures, and/or other aspects of the disclosure are not to be considered limitations on the disclosure as defined by the claims or limitations on equivalents to
10 the claims, and that other embodiments may be utilised and modifications may be made without departing from the scope and/or spirit of the disclosure. Various embodiments may suitably comprise, consist of, or consist essentially of, various combinations of the disclosed elements, components, features, parts, steps, means, etc. In addition, the disclosure includes other inventions not presently claimed, but which
15 may be claimed in future.

Claims

1. A method for treating a liquid tobacco extract, wherein the method comprises contacting the tobacco extract with an ion exchange resin to reduce the polyphenol
5 content.
2. A method according to claim 1, wherein the method comprises contacting the tobacco extract with an ion exchange resin to reduce the chlorogenic acid content.
- 10 3. A method according to claim any one of the preceding claims, wherein the method removes less than 50% of nicotine from the tobacco extract.
4. A method according to any one of the preceding claims, wherein the method removes less than 50% of sugars from the tobacco extract.
- 15 5. A method according to any one of the preceding claims, wherein the method further comprises membrane filtration.
6. A method according to claim 5, wherein membrane filtration is carried out with
20 a Molecular Weight Cut-Off of not more than 500 kDa.
7. A method according to claims 5 or 6, wherein membrane filtration is ultrafiltration.
- 25 8. A method according to any one of the preceding claims, wherein the method of the invention further comprises: treating the tobacco extract with one or more enzymes; treating the tobacco extract with one or more surfactants; and/or treating the tobacco extract with one or more adsorbents.
- 30 9. A tobacco extract which has been treated by a method according to any one of the preceding claims.
10. A method for reducing the polyphenol content of tobacco material, the method comprising:
35 i) extracting components from tobacco material with a solvent to form a liquid extract and a tobacco residue;

- ii) contacting the liquid extract with an ion exchange resin to form a treated extract; and,
- iii) combining the treated extract with the tobacco residue.

5 11. A method for reducing the polyphenol content of tobacco material, the method comprising:

- i) extracting components from tobacco material with a solvent to form a liquid extract and a tobacco residue;
- ii) treating the liquid extract by a method as claimed in any one of claims 1 to 8;
- 10 and,
- iii) combining the treated extract with the tobacco residue.

12. A tobacco material which has been treated by a method according to claims 10 or 11, or a derivative thereof.

15

13. A smoking article which comprises a tobacco material according to claim 12, or a derivative thereof.

14. Use of an ion exchange resin for removing one or more polyphenols from a
20 liquid tobacco extract.

Figure 1

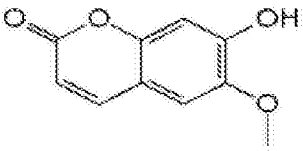
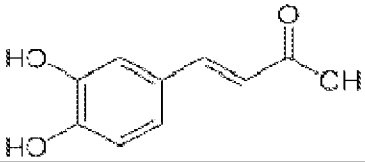
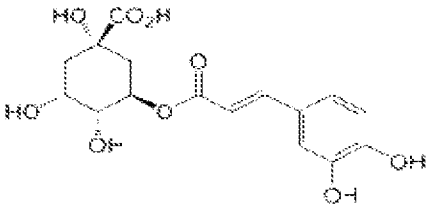
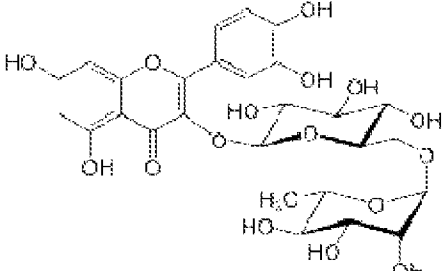
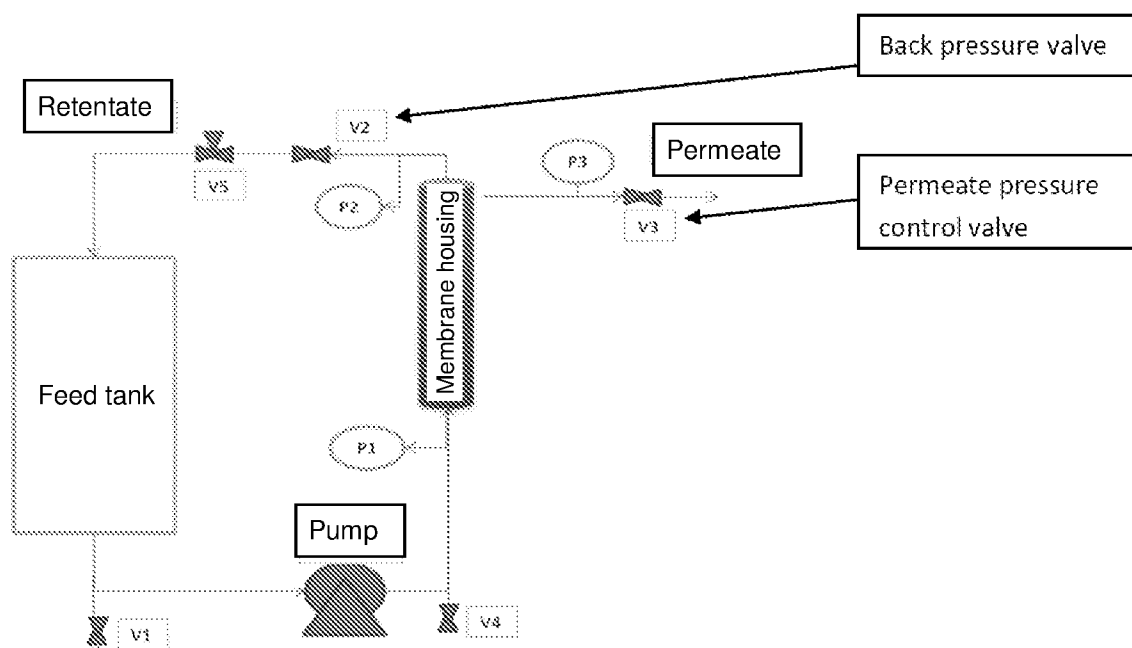
Polyphenol	Chemical Structure
Scopoletin	
Caffeic acid	
Chlorogenic acid	
Rutin	

Figure 2

2/4

Figure 3

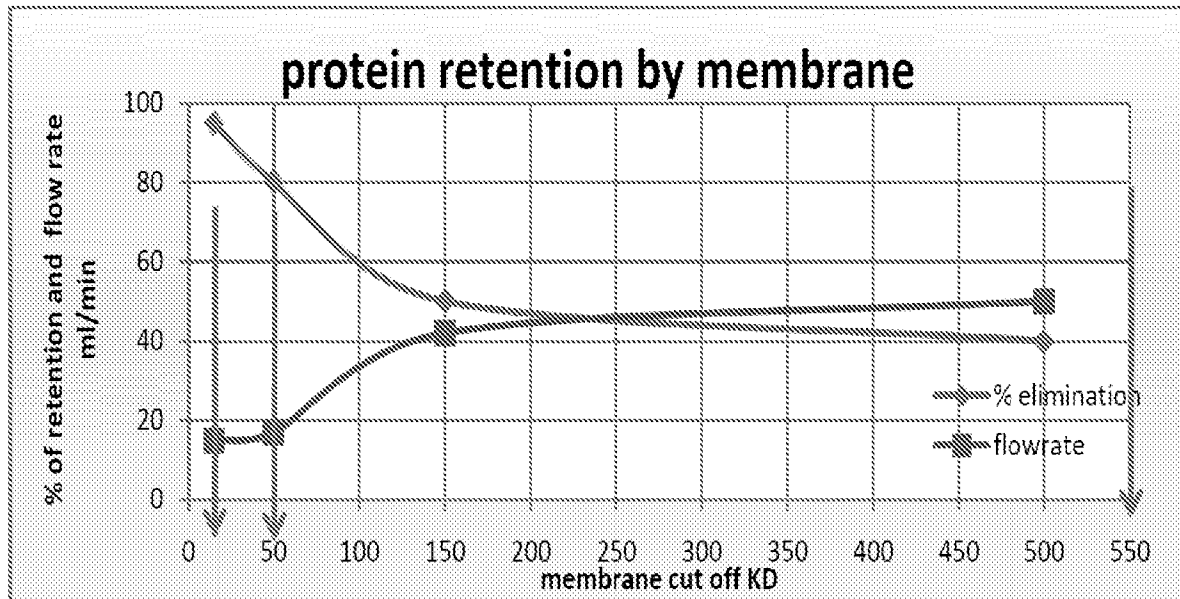


Figure 4

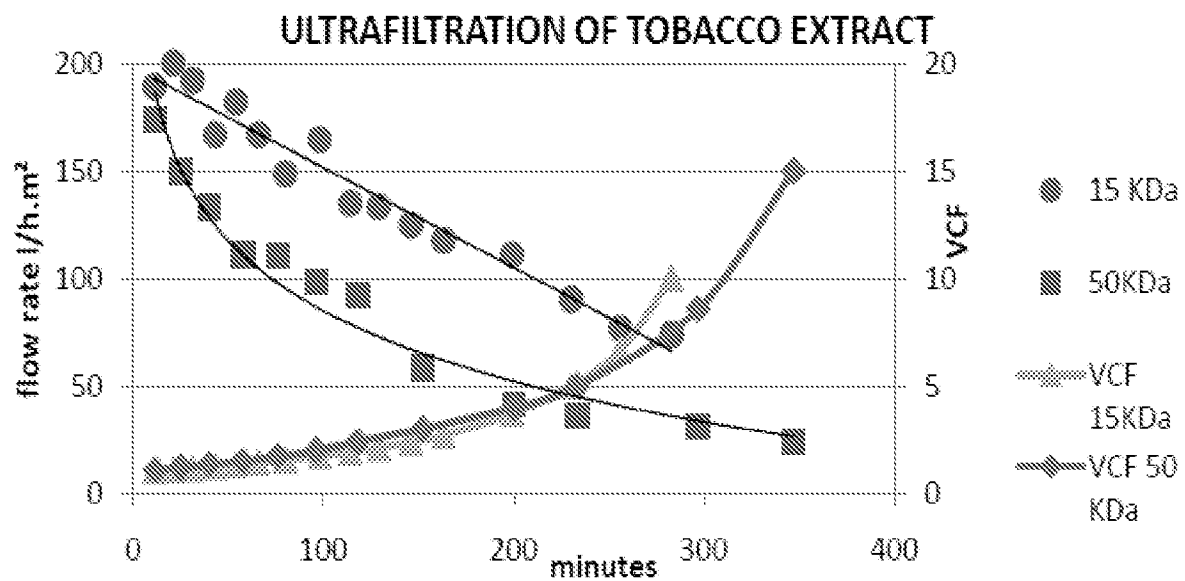


Figure 5

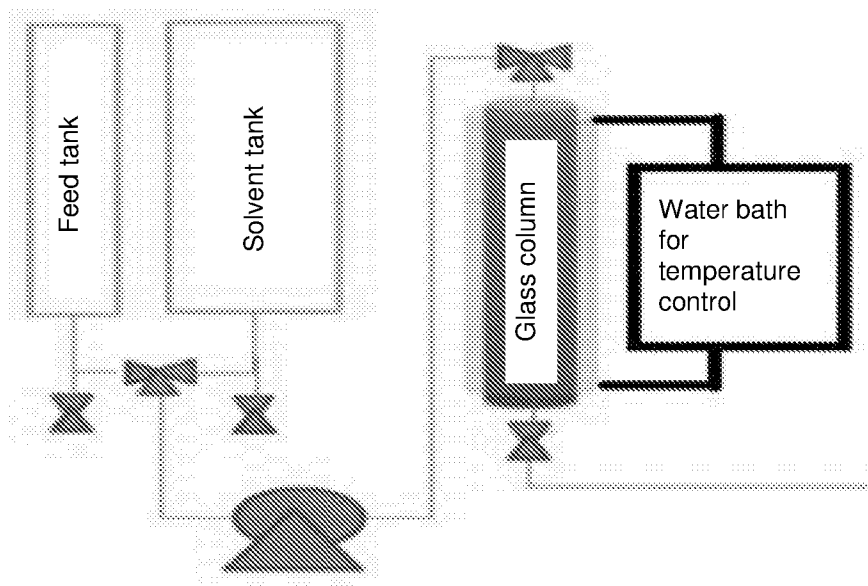


Figure 6

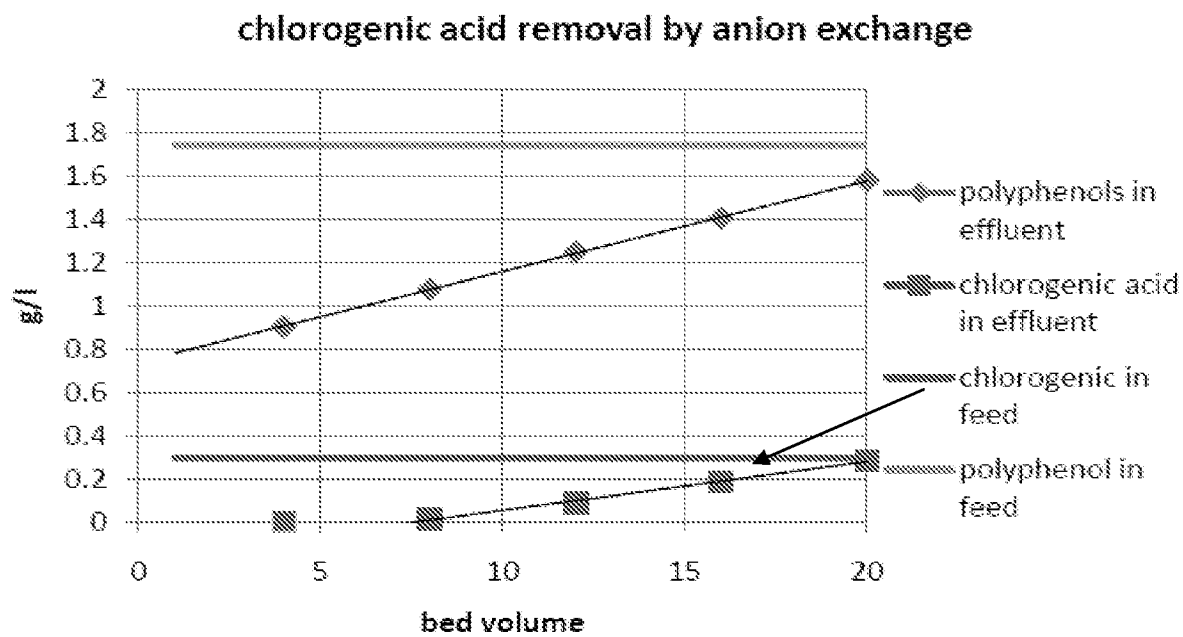


Figure 7

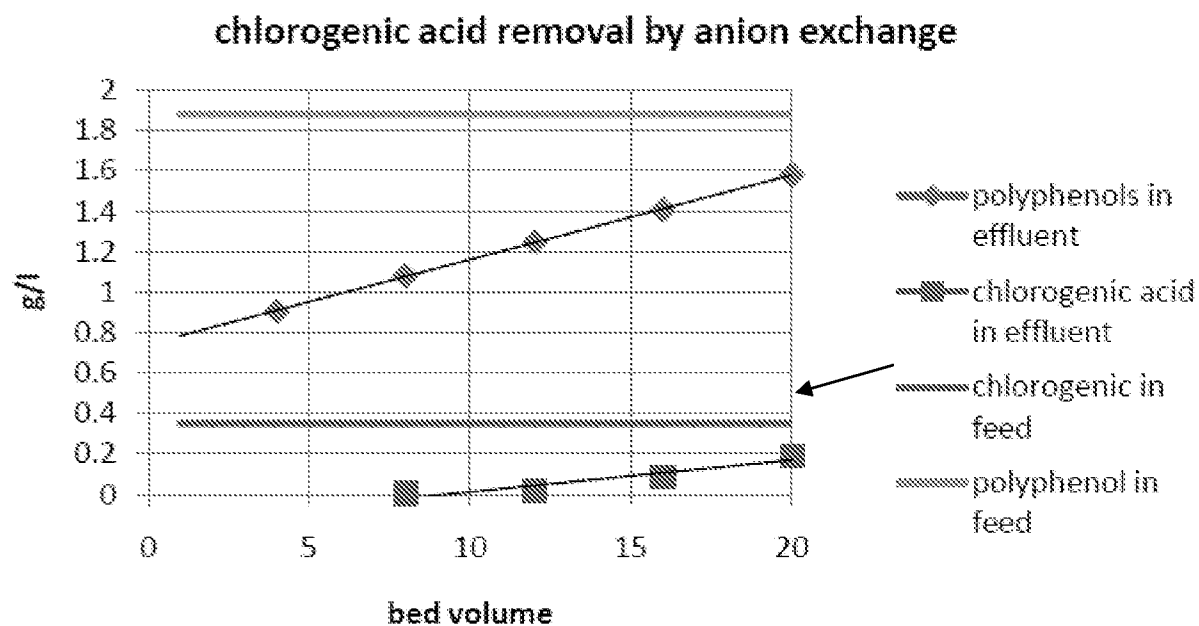
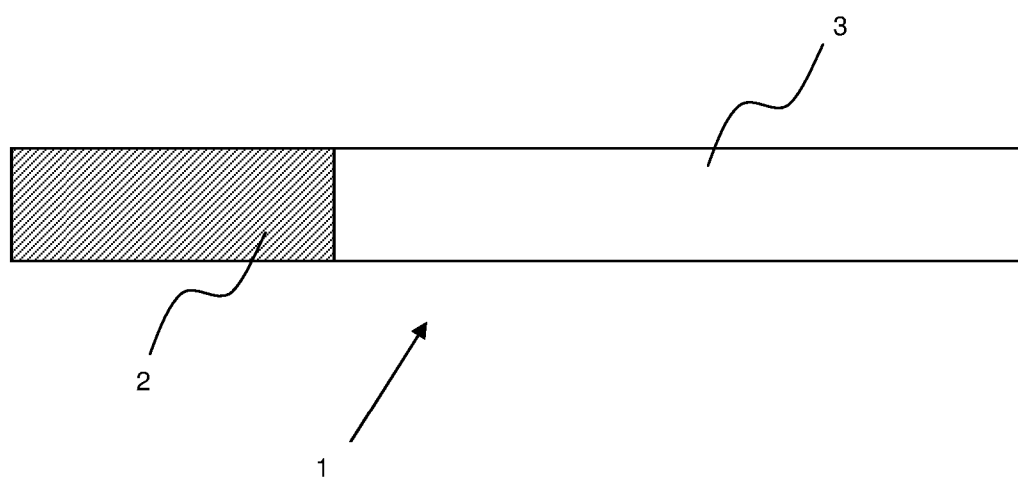


Figure 8



INTERNATIONAL SEARCH REPORT

International application No

PCT/GB2013/053103

A. CLASSIFICATION OF SUBJECT MATTER

INV. A24B15/24

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)

A24B

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

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X	WO 00/02464 A1 (NOVO NORDISK AS [DK]; IMP TOBACCO LTD [CA]) 20 January 2000 (2000-01-20) cited in the application page 31, line 21 - line 30 page 32, line 4 - line 16 page 37, line 24 - line 29 -----	1-14
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A	JP S54 117479 A (JAPAN TOBACCO & SALT PUBLIC) 12 September 1979 (1979-09-12) abstract ----- -/--	1-14

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

☒ See patent family annex.

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Date of the actual completion of the international search

14 February 2014

Date of mailing of the international search report

21/02/2014

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INTERNATIONAL SEARCH REPORT

International application No

PCT/GB2013/053103

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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