TREATMENT OF TOBACCO MATERIAL

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ABSTRACT

A method is provided for treating a tobacco material, comprising treating the tobacco material with an ionic liquid. Also provided is a tobacco material which has been treated by such a method, or a derivative thereof, and a smoking article which comprises a tobacco material treated by such a method.
Figure 1

\[
\begin{align*}
\text{Pyridine} & \quad + \quad \text{Ethyl chloride} \quad \xrightarrow{90^\circ \text{C}, 72\text{h}} \quad \text{BMIMCl} \\
\text{N,N-diethylamine} & \quad + \quad \text{Formic acid} \quad \xrightarrow{90^\circ \text{C}, 24\text{h}} \quad \text{DMAPA FA}
\end{align*}
\]

Figure 2

<table>
<thead>
<tr>
<th>Polyphenol</th>
<th>Chemical Structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scopoletin</td>
<td><img src="image" alt="Scopoletin" /></td>
</tr>
<tr>
<td>Caffeic acid</td>
<td><img src="image" alt="Caffeic acid" /></td>
</tr>
<tr>
<td>Chlorogenic acid</td>
<td><img src="image" alt="Chlorogenic acid" /></td>
</tr>
<tr>
<td>Rutin</td>
<td><img src="image" alt="Rutin" /></td>
</tr>
</tbody>
</table>
TREATMENT OF TOBACCO MATERIAL

FIELD OF THE INVENTION

[0001] The present invention relates to a method for the treatment of tobacco material.

BACKGROUND

[0002] 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.

SUMMARY

[0003] According to a first aspect, there is provided a method of treating a tobacco material, wherein the method comprises treating the tobacco material with an ionic liquid.

[0004] In some embodiments, treating the tobacco material with an ionic liquid results in a reduction in the content of one or more chemical substances in the tobacco material.

[0005] In further embodiments, the one or more of these chemical substances include proteins and/or polyphenols.

[0006] In some embodiments, the method of the invention does not substantially reduce the amount of nicotine in the tobacco material.

[0007] In some embodiments, the ionic liquid used is a Deep Eutectic Solvent. The Deep Eutectic Solvent may, for example, comprise and/or be formed using choline chloride and a range of hydrogen donors.

[0008] In some embodiments, the ratio of the ionic liquid to the tobacco material is at least 1:1 by weight.

[0009] In some embodiments, the tobacco material is treated with an ionic liquid for at least 1 hour.

[0010] In some embodiments, the tobacco material is treated with an ionic liquid at a temperature which is above ambient temperature.

[0011] In some embodiments, the tobacco material treated with an ionic liquid is subsequently separated from the ionic liquid. This separation may involve, for example, filtration and/or centrifugation.

[0012] In some embodiments, the tobacco material is washed with water following treatment with an ionic liquid.

[0013] In addition to treating tobacco material with an ionic liquid, the method of the invention may further comprise: treating the tobacco material with one or more non-ionic liquids, such as water; treating the tobacco material with one or more enzymes; treating the tobacco material with one or more surfactants; and/or treating the tobacco material with one or more adsorbents.

[0014] According to a second aspect, there is provided a tobacco material which has been treated by a method according to the first aspect, or a derivative thereof.

[0015] According to a third aspect, there is provided a smoking article which comprises the tobacco material according to the second aspect, or a derivative thereof.

[0016] According to a fourth aspect, there is provided a use of an ionic liquid for removing proteins and/or polyphenols from a tobacco material.

BRIEF DESCRIPTION OF THE DRAWINGS

[0017] Embodiments of the invention will now be described, by way of example only, with reference to the accompanying drawings, in which:

[0018] FIG. 1 shows how two ionic liquids, 1-butyl-3-methylimidazolium chloride ([BMIM][Cl]) and 3-(dimethylamino)-1-propylaminium formate ([DMAP][FA]), may be synthesised.

[0019] FIG. 2 shows the chemical structure of the four reference polyphenol compounds detected and measured in experiments using High-Performance Liquid Chromatography (HPLC): scopoletin, caffeine acid, chlorogenic acid, and rutin.

[0020] FIG. 3 shows an HPLC spectrum obtained from a sample containing each of the four reference polyphenol compounds at a concentration of 100 ppm.

[0021] FIG. 4 shows an HPLC spectrum obtained from one of the experimental samples, in which the peaks attributable to the four polyphenol compounds are labelled.

[0022] FIG. 5 is a schematic side view of a smoking article including treated tobacco material according to embodiments of the invention.

DETAILED DESCRIPTION

[0023] There is provided a method of treating a tobacco material, wherein the method comprises treating the tobacco material with an ionic liquid.

[0024] In the past, methods attempting to remove proteins or polyphenols have been proposed, although they have tended to be complex and expensive.

[0025] Treating the tobacco material with an ionic liquid may be used for the purpose of modifying the tobacco material in any suitable way. In some embodiments, treating the tobacco material with an ionic liquid leads to the removal of one or more chemical substances. In particular, in some embodiments, treating the tobacco material with an ionic liquid leads to the removal of one or more chemical substances which are undesirable in tobacco material in certain circumstances. In some embodiments, treating the tobacco material with an ionic liquid leads to the removal of one or more proteins and/or polyphenols.

[0026] The treatment of tobacco material with an ionic liquid 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 with an ionic liquid. In some embodiments, however, the tobacco material treated with an ionic liquid has already been cured and may be cured cut rag and/or cured whole leaf tobacco. Examples of tobaccos which may be treated with an ionic liquid include, but are not limited to: Virginia, Burley, Maryland, Oriental and Rustica.

[0027] The treatment of tobacco material with an ionic liquid may remove one or more chemical substances from the tobacco material. In some embodiments, one or more of the chemical substances removed from the tobacco material are proteins and/or polyphenols.

[0028] The treatment of tobacco material with an ionic liquid comprises at least one step in which the tobacco material is treated with an ionic liquid, and may comprise more than one step in which the tobacco material is treated with an ionic liquid. In embodiments wherein there is more than one step in which the tobacco material is treated with an ionic liquid, the same or different ionic liquids and the same or different conditions may be used in each step.

[0029] Polyphenols which may be removed by the treatment include, but are not limited to: chlorogenic acid, caffeic...
acid, rutin, scopoletin, and quercetin. A wide range of polyphenols are expected to be solubilised.

[0030] In some embodiments, the treatment of tobacco material with an ionic liquid, and in particular the step of treating tobacco material with an ionic liquid, 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.

[0031] Nicotine may be an example of such a substance, and for this reason in some embodiments it is undesirable to remove this molecule. In some embodiments, the treatment of tobacco material with an ionic liquid removes less than 50%, 40%, 30%, 20%, 10%, or 5% of the nicotine from the tobacco material; in further embodiments, the treatment removes less than 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.

[0032] In embodiments wherein treating the tobacco material with an ionic liquid 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.

[0033] It may be advantageous to use an ionic liquid to selectively remove one or more chemical substances from tobacco material because an ionic liquid is often a good solvent for some substances yet a poor solvent for others. Consequently, an ionic liquid may be selected which is a good solvent for chemical substances in tobacco material whose removal would be desirable, such as proteins and/or polyphenols, while at the same time being a poor solvent for chemical substances in tobacco material whose removal would be undesirable, such as nicotine.

[0034] An ionic liquid is an ionic chemical substance made up of anions and cations in the liquid phase. The treatment of tobacco material with an ionic liquid may involve the use of any suitable ionic liquid, any suitable mixture of ionic liquids, and any suitable number of ionic liquids.

[0035] An ionic liquid used in the treatment may have any suitable properties. In some embodiments, for example, the ionic liquid may have a melting point below ambient temperature. In further embodiments, the ionic liquid may be adapted and/or include additives to provide it with properties which may, in some circumstances, be beneficial.

[0036] In some embodiments, the ionic liquid may comprise one or more solutes to modify its melting point and/or solvating properties. In some embodiments, the pH of the ionic liquid may be adjusted to modify its solvating properties, for example, to make it a better solvent for protein and/or polyphenol compounds.

[0037] An ionic liquid used in the treatment may comprise organic and/or inorganic ions. Furthermore, in some embodiments, an ionic liquid may comprise ions with chemical functionality that may be modified in order to change one or more properties of the ionic liquid and, in some embodiments, change one or more properties of the ionic liquid in a predictable way. Cations and/or anions of the ionic liquid may, for example, comprise one or more alkyl and/or aryl substituents, and one or more of these may be changed in length and/or functionalised in order to modify the solvating properties of the ionic liquid.

[0038] In some embodiments, an ionic liquid used in the treatment is a Deep Eutectic Solvent (DES): an ionic solvent which is in a eutectic composition—that is, comprises two or more components which have a lower melting point when mixed together than when apart. It may be advantageous for one or more of the ionic liquids used in the treatment to be a DES for a number of reasons. Firstly, for example, DESs tend to have lower melting points than other ionic liquids by virtue of being eutectic, which is likely to be useful since ionic liquids often have high melting points. Secondly, for example, DESs are often simpler to synthesise and cheaper to make compared to other ionic liquids.

[0039] The treatment of tobacco material with an ionic liquid may involve the use of any suitable DES, any suitable mixture of DESs, and any suitable number of DESs. A suitable DES may comprise a salt mixed with a species capable of complexing to the anions and/or cations of the salt in order to lower its melting point, for example. One or more DESs used in the treatment may be Type I (metal salt & organic salt), Type II (metal salt hydrate & organic salt), Type III (organic salt & hydrogen bond donor), and/or Type IV (metal salt & metal salt hydrate) & hydrogen bond donor), for example.

[0040] In some embodiments, a DES used in the treatment comprises choline chloride or any suitable derivative thereof, since in addition to being non-toxic, cheap, and biodegradable, choline chloride can be mixed with many different chemical species to form many different DESs, many of which have been found to effectively dissolve polyphenol compounds. Examples of suitable choline-chloride-containing DESs are acetic acid/choline chloride (in a 2:1 ratio, for example), lactic acid/choline chloride (in a 2:1 ratio, for example), and urea/choline chloride (in a 2:1 ratio, for example).

[0041] In addition to the foregoing exemplary DES ionic liquids, suitable non-DES ionic liquids include, but are not limited to: phosphate salts; hydrogen phosphate salts; dihydrogen phosphate salts; dimethylaminopropylamine ([(DMAP+A)]) salts, such as 3-(dimethylamino)-1-propylaminium formate [(DMAP+A)FA]; and 1-butyl-3-methylimidazolium ([(BMIM)+]) salts, such as 1-butyl-3-methylimidazolium chloride ([(BMIM)+]Cl).

[0042] Similar ionic liquids to those specifically mentioned above which would be expected to give similar results include those with a range of imidazolium cations, such as 1-butyl-3-methylimidazolium (BMIM) and related 1-propyl-3-methylimidazolium (PMIM) and 1-ethyl-3-methylimidazolium (EMIM), etc. Suitable anions include all halides (especially Cl and Br) as well as acetate, carboxylates, tetrafluoroborate (BF4) and hexafluorophosphate (PF6).

[0043] Analogues of [(DMAP+A)FA which would be expected to give similar results include those where the 3-(dimethylamino)-1-propylamine (DMAPA) cation is replaced by similar diamines or triamines. Alternative anions to formic acid (FA) could also be used, for example acetic acid or trifluoroacetic acid.

[0044] An ionic liquid used in the treatment may be synthesised using any suitable method of synthesis. FIG. 1 illustrates how, for example, [(BMIM)+Cl could be made by a simple S2O2 reaction between a nucleophilic amine and an electrophilic chloroalkane, and how, for example, [(DMAP+A] FA could be made by the transfer of a proton from an acidic carboxyl group to a basic amino group. More specifically, FIG. 1 illustrates how [(BMIM)+Cl could be synthesised by reacting 1-methylimidazole with 1-chlorobutane at 90°C, for
72 hours, and how [DMAPA]FA could be synthesised by reacting dimethylaminopropylamine (DMAPA) with Formic Acid (FA) at 0°C for 24 hours.

In some embodiments, the treatment of the tobacco material with an ionic liquid 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 content of the polyphenol(s) in the untreated tobacco material.

In some embodiments, the treatment of the tobacco material with an ionic liquid 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 content of the polyphenol(s) in the untreated tobacco material.

Alternatively or in addition, the treatment of the tobacco material with an ionic liquid 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 material.

In some embodiments, the treatment of the tobacco material with an ionic liquid 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.

When tobacco material is treated with an ionic liquid, the ionic liquid may have any suitable weight and the tobacco material may have any suitable weight. The ratio of ionic liquid to tobacco material, by weight, may have any suitable value. In some embodiments, the ratio of tobacco material to ionic liquid results in the removal of proteins and/or polyphenols from the tobacco material. In some embodiments, the ratio of ionic liquid to tobacco is selected so that removal of desirable substances (whose removal would be undesirable), such as nicotine, is minimised. In some embodiments, a ratio is selected which results in as little of the desirable substances being removed as possible, whilst still resulting in the removal of proteins and/or polyphenols. This ratio may be different for different ionic liquids and tobacco materials.

In many embodiments, a greater quantity of protein and/or polyphenol is likely to be removed when the weight of the ionic liquid is greater than or equal to the weight of the tobacco material. Without wishing to be bound by any particular theory, it is hypothesised that this is because the greater the weight of an ionic liquid, the greater its volume, and having a greater volume of ionic liquid means that more of the tobacco material is contacted with the ionic liquid. Additionally, when the ionic liquid dissolves a chemical substance from the tobacco material, the concentration of that substance is lower, and so the entropy change of the system upon dissolution is more positive. In some embodiments, therefore, the weight of the ionic liquid is greater than or equal to the weight of the tobacco material and, in some embodiments, the ratio of ionic liquid to tobacco, by weight, may be at least or about 1:1, 2:1, 3:1, 4:1, 5:1, 6:1, 7:1, 8:1, 9:1, 10:1, 15:1, 20:1, 25:1, 30:1, or any suitable higher ratio, optionally with a maximum ratio of about 3:1. In some embodiments, the boiling point of the ionic liquid may be modified using any suitable means, such as by adjusting the surrounding pressure and/or adding any suitable chemical substances.

When tobacco material is treated with an ionic liquid in the treatment, the tobacco material may be treated for any suitable length of time. In some embodiments, the adopted length of time results in the removal of proteins and/or polyphenols from the tobacco material. In some embodiments, the length of time for which the tobacco material is treated with an ionic liquid is such that the treatment 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 content of the polyphenol(s) in the untreated tobacco material. Additionally or alternatively, the length of time for which the tobacco material is treated with an ionic liquid is such that the treatment 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 protein content of the untreated tobacco material.
tobacco material. In some embodiments, the duration of the treatment of the tobacco with the ionic liquid is selected so that removal of desirable substances (whose removal would be undesirable), such as nicotine, is minimised. In some embodiments, a duration of the treatment is selected which results in as little of the desirable substances being removed as possible, whilst still resulting in the removal of proteins and/or polyphenols. The duration of the treatment may be different for different ionic liquids and tobacco materials.

In most cases, however, a greater quantity of proteins and/or polyphenols is likely to be removed when the tobacco material is treated with an ionic liquid for at least 1 hour. Without wishing to be bound by any particular theory, it is hypothesised that this is because the substances removed from the tobacco material begin in the tobacco material and so, by extending the reaction time and therefore bringing the reactions closer to thermodynamic equilibrium, the concentration of substances dissolved in the ionic liquid can only increase. In some embodiments, the tobacco material is treated with an ionic liquid for at least or about 1 hour, 2 hours, 3 hours, 4 hours, 5 hours, 6 hours, 7 hours, 8 hours, 9 hours, at least or about 10 hours, or any suitable longer length of time. Optionally, the treatment period may be up to about 3 hours, 4 hours, 5 hours, 6 hours, 7 hours, 8 hours, 9 hours, 10 hours, 11 hours or 12 hours.

When tobacco material is treated with an ionic liquid, the mixture may be agitated in any suitable way using any suitable means. For example, the mixture may be agitated by stirring, shaking, and/or rocking in any suitable way using any suitable apparatus. Working on a small scale, the reaction could be carried out in a standard glass beaker, using a magnetic or overhead stirrer to agitate the mixture, with the temperature optionally being controlled by means of a hot plate.

After the tobacco material has been treated with an ionic liquid, the residual tobacco material may be separated from the ionic liquid. This separation may involve any suitable filtration method, any suitable filtering medium or size, and any suitable number of filtration steps. For example, the tobacco material may be filtered by nanofiltration, microfiltration, and/or ultrafiltration. Alternatively or in addition, the tobacco material may be separated from the ionic liquid by centrifugation using any suitable centrifuge system, any suitable angular velocity, and any suitable number of centrifugation steps.

Once separated from the ionic liquid by filtration and/or centrifugation, the tobacco material may be washed with any suitable number of times using any suitable liquid or liquids, such as water, in order to remove any residual ionic liquid.

In addition to treating tobacco material with an ionic liquid, the method of treating a tobacco material may comprise further treatment steps. Suitable additional treatment steps include, but are not limited to: treating the tobacco material with one or more suitable non-ionic liquids, such as water; treating the tobacco material with one or more enzymes, which may be enzymes which catalyse the modification of polyphenols or proteins, such as phenol-oxidising and proteolytic enzymes; treating the tobacco material with one or more surfactants, such as sodium dodecylsulfate (SDS), in any suitable solvent; and treating the tobacco material with one or more suitable absorbent materials, such as polyvinyl polypyrrolidone (PVPP), hydroxyapatite, bentonite, activated carbon or attapulgite, in any suitable solvent if appropriate.

Additionally or alternatively, the tobacco material treated with an ionic liquid may be subsequently subjected to further extraction processes.

Having undergone any of the previously-described treatment steps in accordance with the method of the invention, the tobacco material may be dried and 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.

As used herein, the term “smokeable products” 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 products. The smoking article may be provided with a filter for the gaseous flow drawn by the smoker.

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, cinnamon, herb, wintergreen, cherry, berry, peach, apple, Drambuie, bourbon, scotch, whiskey, spearmint, peppermint, lavender, cardamon, celery, cascara, nutmeg, sandalwood, bergamot, geranium, honey essence, rose oil, vanilla, lemon oil, orange oil, cassia, caraway, coriander, jasmine, ylang-ylang, saffron, pimento, ginger, anise, coriander, coffee, or a mint oil from any species of the genus Mentha), flavour 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.

In an exemplary embodiment, a sample of cured whole leaf tobacco is added to the ionic liquid [DMPA]/[FA] at a temperature of 60°C, so that the ratio of tobacco material to ionic liquid is 1:10, by weight. The mixture of ionic liquid and tobacco material is then left for 1 hour before the tobacco material is filtered from the ionic liquid, dried, washed, and modified in any suitable way before being incorporated into a smoking article.

Referring to FIG. 5. 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.

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 incorpor-
rated 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.

Experimental Work

[0066] A series of experiments were carried out in order to investigate how the treatment of tobacco material with an ionic liquid can affect the protein, polyphenol, and nicotine content of tobacco material. The disclosed experimental work is not intended to limit the scope of the invention.

[0067] Two types of ionic liquid were investigated: DES ionic liquids and non-DES ionic liquids.

DES Ionic Liquids

[0068] Three Deep Eutectic Solvents (DESs) were tested for how they affect the protein, polyphenol, and nicotine content of tobacco material. The three tested DESs were acetic acid:choline chloride in a 2:1 ratio (AA/CC), lactic acid:choline chloride in a 2:1 ratio (LA/CC), and urea:choline chloride in a 2:1 ratio (U/CC).

Experiments

[0069] A total of eight experiments were carried out. In each of these, a tobacco material was treated with one of the three DESs, before being filtered, and then washed. Following this protocol, there were three samples—residual tobacco material, ionic liquid extract, aqueous washing extract—ready to be analysed for the purpose of determining any changes in the protein, polyphenol, and nicotine content of the tobacco material.

[0070] Five variables were used in different combinations in each of the experiments: DES solvent (AA/CC, LA/CC, or U/CC), tobacco type (whole leaf or cut rag), tobacco material to ionic liquid ratio (3:1 or 1:10), and temperature (60°C or 120°C). The conditions used in each of the experiments are detailed below in Table 1.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>DES</th>
<th>Tobacco Material</th>
<th>Tobacco Material:DES (by weight)</th>
<th>Temp (°C)</th>
<th>Reaction Time (hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>AA/CC</td>
<td>Whole leaf</td>
<td>3:10</td>
<td>60</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>LA/CC</td>
<td>Whole leaf</td>
<td>3:10</td>
<td>60</td>
<td>1</td>
</tr>
<tr>
<td>3</td>
<td>U/CC</td>
<td>Whole leaf</td>
<td>3:10</td>
<td>60</td>
<td>1</td>
</tr>
<tr>
<td>4</td>
<td>AA/CC</td>
<td>Whole leaf</td>
<td>1:10</td>
<td>60</td>
<td>1</td>
</tr>
<tr>
<td>5</td>
<td>LA/CC</td>
<td>Whole leaf</td>
<td>1:10</td>
<td>60</td>
<td>1</td>
</tr>
<tr>
<td>6</td>
<td>U/CC</td>
<td>Whole leaf</td>
<td>1:10</td>
<td>60</td>
<td>1</td>
</tr>
<tr>
<td>7</td>
<td>LA/CC</td>
<td>Whole leaf</td>
<td>1:10</td>
<td>120</td>
<td>2</td>
</tr>
<tr>
<td>8</td>
<td>LA/CC</td>
<td>Cut rag</td>
<td>1:10</td>
<td>60</td>
<td>1</td>
</tr>
</tbody>
</table>

Analysis: Polyphenol Content

[0071] High-Performance Liquid Chromatography (HPLC) was used to provide a measure of the quantity of polyphenol compounds removed from tobacco material in each of the eight experiments.

[0072] In order to obtain this measure, the concentration of four reference polyphenol compounds—scopoletin, caffeine acid, chlorogenic acid, rutin—were measured by carrying out HPLC on two of the three collected samples, the ionic liquid extract and the aqueous washing extract.

[0073] The chemical structures of the four reference polyphenol compounds are pictured in FIG. 2.

[0074] HPLC could only be used to measure the concentration of the four reference polyphenol compounds due to the way in which the analytical method was used to quantify their concentrations. Each polyphenol compound provides a peak at a particular position on an HPLC spectrum, and, importantly, the position of this peak was only known for the four reference polyphenol compounds. Consequently, only the concentrations of the four reference polyphenol compounds could be determined using HPLC by comparing the peaks in the collected spectra with the peaks in a spectrum obtained for a sample containing known concentrations of the four reference polyphenol compounds.

[0075] The HPLC spectrum obtained for a sample containing each of the four reference polyphenols at a concentration of 100 ppm is shown in FIG. 3. The relative positions and integrals of the peaks in the spectrum were used to qualitatively and quantitatively measure the four reference polyphenols in experimental samples.

[0076] The concentrations of the four reference polyphenols measured in the two extracts—the ionic liquid and the aqueous washing—in each of the experiments are detailed below in Table 2.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Cntr. % of Caffeine Acid (ppm)</th>
<th>Cntr. % of Scopoletin (ppm)</th>
<th>Cntr. % of Chlorogenic Acid (ppm)</th>
<th>Cntr. % of Rutin (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0</td>
<td>36</td>
<td>97</td>
<td>92</td>
</tr>
<tr>
<td>2</td>
<td>13</td>
<td>44</td>
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</tr>
<tr>
<td>8</td>
<td>7</td>
<td>26</td>
<td>257</td>
<td>126</td>
</tr>
</tbody>
</table>

[0077] An example of an HPLC spectrum collected from one of the experimental samples, namely, experiment 7, is shown in FIG. 4. The Figure shows that there were several peaks detected in addition to those attributable to the four reference polyphenol compounds, some of which were likely to be other polyphenol compounds removed from the tobacco material.

[0078] Once the concentrations of the four reference polyphenol compounds had been measured using HPLC, they were converted into masses, before the masses were calculated as percentages of the mass of the original tobacco material on a Dry Weight Basis (% DWB).

[0079] The total mass of the four reference polyphenols measured in each of the extracts could be taken as a measure of the total mass of the four reference polyphenol compounds removed from the tobacco material by the ionic liquid. Furthermore, the total mass of the four reference polyphenols removed from the tobacco material could be taken as an indication of the quantity of all types of polyphenol removed from the tobacco material by the ionic liquid, thereby acting as reference compounds.

[0080] The masses of the four polyphenol compounds (% DWB) measured in the ionic liquid extract and aqueous washing extract in each of the experiments are detailed below in Table 3.
The results provided in Table 3 indicate that treating a tobacco material with a DES does indeed lead to the removal of polyphenol compounds.

In addition, the results indicate how the tested variables affect the removal of polyphenol compounds from tobacco material. Conclusions which may be drawn from these results include, but are not limited to, the following. The solvent LA/CC appears to be the most effective DES for reducing the polyphenol content of tobacco material, the method appears to reduce the polyphenol content of cut rag tobacco more than whole leaf tobacco, a higher ratio (1:10) appears to reduce the polyphenol content of tobacco material more than a lower ratio (3:10); a higher temperature (120°C) appears to reduce the polyphenol content of tobacco material more than a lower temperature (60°C), and a longer reaction time (2 hours) appears to reduce the polyphenol content of tobacco material more than a shorter reaction time (1 hour).

Analysis: Protein Content

The total nitrogen content of the collected samples was measured and, using a simple conversion protocol, used to provide a measure of the total quantity of protein removed from whole leaf tobacco material.

Proteins are molecules of amino acids, each of which contains one nitrogen atom in its generic structure and possibly one or more Nitrogen atoms in its R group. By measuring the total Nitrogen content of a sample, therefore, the total protein content could be estimated by simply translating the total Nitrogen content into total protein content using a suitable conversion factor, known as the Jones factor.

A Jones factor of 6.25 was used in the experiments which is the standard value used for a sample of protein when the type of protein is not considered. If this conversion factor were used alone, however, it would significantly overestimate the protein content in each sample, since there were many other nitrogenous compounds besides protein in each sample. Most importantly, every DES ionic liquid which was tested contains Nitrogen atoms. In order to obtain more accurate estimates of protein content, therefore, the Nitrogen content attributable to the DES in each sample was calculated and incorporated into the conversion calculation.

The total mass of protein measured in the extracts could be taken as a measure of the amount of protein removed from the tobacco material by the ionic liquid. The mass of protein (% DWB) measured in the two extracts and the residual tobacco material is detailed below in Table 4.

The results provided in Table 4 indicate that treating a tobacco material with a DES does indeed lead to the removal of protein.

In addition, the results provide an indication of the way in which each of the tested variables affect the removal of protein from tobacco material. Conclusions which may be drawn from these results include, but are not limited to, the following. The solvent AA/CC appears to be the most effective DES for reducing the protein content of tobacco material, a higher ratio (1:10) appears to reduce the protein content of tobacco material more than a lower ratio (3:10); a higher temperature (120°C) appears to reduce the protein content of tobacco material more than a lower temperature (60°C), and a longer reaction time (2 hours) appears to reduce the protein content of tobacco material more than a shorter reaction time (1 hour).

Analysis: Nicotine Content

HPLC was used to provide a measure of the quantity of nicotine removed from whole leaf tobacco material.

As with the four reference polyphenol compounds, the molecule nicotine provides a peak at a known position on an HPLC spectrum following HPLC, and the quantity of nicotine in each of the analysed samples could therefore be measured by comparing the peak size in the collected HPLC spectra with the peak size in a spectrum obtained for a sample containing a known concentration of nicotine.

The total mass of nicotine measured in the extracts could be taken as a measure of the amount of nicotine removed from the tobacco material by the ionic liquid.

The mass of nicotine (% DWB) measured in the two extracts—ionic liquid and aqueous washing—in each of the experiments are detailed below in Table 5.

The results provided in Table 5 indicate that treating a tobacco material with a DES can result in the removal of a significant quantity of nicotine, but that under certain conditions the quantity of nicotine removed from the tobacco material can be small.
From the results, it would appear that a larger quantity of nicotine is removed from tobacco material when treated with a DES ionic liquid at high temperature and/or for a long period of time.

### Non-DES Ionic Liquids

Two non-DES ionic liquids were tested for how they affect the protein, polyphenol, and nicotine content of tobacco material: [DMPAFA] and [BMIM]Cl.

#### Experiments

Two experiments were carried out. In each of these, a tobacco material was treated with one of the two ionic liquids, before being filtered, and then washed. Following this protocol, there were three samples—residual tobacco material, ionic liquid extract, aqueous washing extract—ready to be analysed in order to determine any changes in the protein, polyphenol, and nicotine content of the tobacco material.

Different variable combinations were not trialled in the experiments for non-DES ionic liquids, since the experiments with DES ionic liquids had already indicated how different variables can affect the extraction of protein, polyphenols, and nicotine from tobacco material. Instead, the purpose of these experiments was to investigate how effective the two non-DES ionic liquids are for the removal of proteins, polyphenols, and nicotine.

The conditions used in each experiment are detailed in Table 6 below.

### Analysis: Polyphenol Content

High-Performance Liquid Chromatography (HPLC) was used to provide a measure of the quantity of polyphenol compounds removed from the tobacco material in each of the two experiments.

HPLC was used to measure the concentration of four reference polyphenol compounds—cyclopein, caffeic acid, chlorogenic acid, and rutin—in the ionic liquid extract.

The total mass of the four reference polyphenols measured in the extract could be taken as a measure of the total mass of the four reference polyphenols removed from the tobacco material by the ionic liquid. And, furthermore, the total mass of the four reference polyphenols removed from the tobacco material could be taken as an indication of the quantity of all types of polyphenol removed from the tobacco material by the ionic liquid.

The masses of the four polyphenol compounds (%) DWB and the concentrations of two of the four polyphenols found in the ionic liquid extract in each of the experiments are detailed below in Table 7.

The results provided in Table 7 indicate that treating a tobacco material with either of the two tested non-DES ionic liquids does not reduce the polyphenol content of tobacco material to the same extent as DES ionic liquids.

It is also worthy of note that, of the four polyphenol compounds, chlorogenic acid and rutin were found to have the highest concentration in the ionic liquid extract in both experiments, which may suggest that these polyphenols are most abundant in tobacco material. The results also show that the concentrations of chlorogenic acid and rutin, measured in each of the two experiments were very different, with the measured concentration of chlorogenic acid greater than rutin in [DMPAFA]Cl, yet smaller than rutin in [BMIM]Cl.

**TABLE 7**

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Mass of Four Polyphenols in Ionic Liquid Extract (% DWB)</th>
<th>Concentration of Chlorogenic Acid in Ionic Liquid Extract (ppm)</th>
<th>Concentration of Rutin in Ionic Liquid Extract (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.05</td>
<td>46</td>
<td>3</td>
</tr>
<tr>
<td>2</td>
<td>0.08</td>
<td>22</td>
<td>31</td>
</tr>
</tbody>
</table>

The total mass of protein measured in the extracts could be taken as a measure of the amount of protein removed from the tobacco material by the ionic liquid.

The masses of protein measured in the two extracts and the residual tobacco material are detailed below in Table 8.

**TABLE 8**

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Assumed Mass of Solubilised Tobacco (% DWB)</th>
<th>Mass of Protein in Extracts (% of extracts mass)</th>
<th>Mass of Protein in Residual Tobacco Material (% of residual tobacco mass)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>30</td>
<td>&lt;0</td>
<td>~40</td>
</tr>
<tr>
<td>2</td>
<td>n/a</td>
<td>n/a</td>
<td>n/a</td>
</tr>
</tbody>
</table>

Protein content data is not available for [BMIM]Cl, but is available for [DMPAFA]. The mass of solubilised tobacco material in the ionic liquid needed to be assumed in order to estimate protein content from measured nitrogen content.

Interestingly, the results provided in Table 8 suggest that the mass of protein in the extracts was less than 0%, despite this clearly being impossible. The negative value is simply a consequence of the method used to calculate protein content, and indicates how the quantity of protein measured in this way is only an approximation. Such an approximation is still useful, however, and obtaining such a low percentage does still provide useful information. It suggests that the
amount of protein in the extracts was low and that the amount of protein in the residual tobacco material was high, due to the way in which protein content was calculated from nitrogen content.

0111] Importantly, the results in Table 7 suggest that non-DES ionic liquids are less effective for the removal of protein from tobacco material than DES ionic liquids.

Analysis: Nicotine Content

0112] The reduction in nicotine content of tobacco material was determined in the same way as for the DES ionic liquids: HPLC was used to provide a measure of the quantity of nicotine removed from whole leaf tobacco material.

0113] The masses of nicotine (% DWB) measured in the two extracts—the ionic liquid and the aqueous washing—in each of the experiments are detailed below in Table 9.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Mass of Nicotine in Extracts (% DWB)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.28</td>
</tr>
<tr>
<td>2</td>
<td>n/a</td>
</tr>
</tbody>
</table>

Nicotine content data is not available for [BMIM] Cl, but is available for [DMAPA]FA.

0115 The results provided in Table 9 indicate that treating a tobacco material with a non-DES ionic liquid may lead to the removal of a significant quantity of nicotine under certain conditions. Despite there being no data for [DMAPA]FA, it may be tentatively concluded that the results provide an indication of how non-DES ionic liquids can affect the nicotine content of tobacco material.

Analysis: Recovery of Whole Leaf Tobacco

0116] In each of the two experiments, the residual whole leaf tobacco material which remained after washing was weighed in order to determine the percentage of the material which remained after treatment.

0117] The masses of residual tobacco materials (% DWB) recovered in each of the two experiments are detailed below in Table 10.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Mass of Residual Tobacco Material (% DWB)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>71.5</td>
</tr>
<tr>
<td>2</td>
<td>11.5</td>
</tr>
</tbody>
</table>

The results provided in Table 10 indicate that [DMAPA]FA dissolves a much greater proportion of whole leaf tobacco than [BMIM]Cl. Without wishing to be bound by any particular theory, the difference may be so large because [BMIM]Cl is able to dissolve chemical constituents of the plant cell walls, such as cellulose, more effectively than [DMAPA]FA, thereby opening the plant cells and facilitating the removal of significantly more chemical substances.

0119] 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 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 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 may be claimed in future.

1. A method for removing one or more chemical substances from a tobacco material, the method comprising: receiving tobacco material; and treating the tobacco material with an ionic liquid to remove one or more chemical substances, the one or more chemical substances including proteins and/or polyphenols.

2. The method according to claim 1, wherein the method does not substantially reduce an amount of nicotine in the tobacco material.

3. The method according to claim 1, wherein the ionic liquid is a Deep Eutectic Solvent.

4. The method according to claim 3, wherein the Deep Eutectic Solvent comprises and/or is formed using choline chloride and a range of hydrogen donors.

5. The method according to any claim 1, wherein a ratio of the ionic liquid to the tobacco material is at least 1:1 by weight.

6. The method according to claim 1, further comprising treating the tobacco material with the ionic liquid for at least 1 hour.

7. The method according to claim 1, further comprising treating the tobacco material with the ionic liquid at a temperature above ambient temperature.

8. The method according to claim 1, the method further comprising separating treated tobacco material from the ionic liquid by filtration and/or centrifugation.

9. The method according to claim 1, the method further comprising washing the tobacco material with water following treatment with the ionic liquid.

10. The method according to claim 1, the method further comprising treating the tobacco material with: one or more non-ionic liquids; one or more enzymes; one or more surfactants; and/or one or more adsorbents.

11. A tobacco material that has been treated by a method according to claim 1.

12. A smoking article comprising the tobacco material according to claim 1.

13. (canceled)

14. The method according to claim 10, wherein the method comprises treating the tobacco material with a non-ionic liquid comprising water.