



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>6</sup> :</b> <b>E06B 3/67, 3/677, C03C 17/28</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 95/25870</b> <b>(43) International Publication Date:</b> 28 September 1995 (28.09.95)
<b>(21) International Application Number:</b> PCT/SE95/00283 <b>(22) International Filing Date:</b> 21 March 1995 (21.03.95)  <b>(30) Priority Data:</b> 9400923-0                      21 March 1994 (21.03.94)                      SE  <b>(71)(72) Applicant and Inventor:</b> KARLSSON, Jan [SE/SE]; Hällskriftsgatan 2 B, S-417 26 Göteborg (SE).  <b>(74) Agents:</b> GRAUDUMS, Valdis et al.; Albihn West AB, P.O. Box 142, S-401 22 Göteborg (SE).		<b>(81) Designated States:</b> AM, AT, AU, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, JP, KE, KG, KP, KR, KZ, LK, LR, LT, LU, LV, MD, MG, MN, MW, MX, NL, NO, NZ, PL, PT, RO, RU, SD, SE, SI, SK, TJ, TT, UA, US, UZ, VN, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG), ARIPO patent (KE, MW, SD, SZ, UG).  <b>Published</b> <i>With international search report.</i> <i>In English translation (filed in Swedish).</i>
<b>(54) Title:</b> A METHOD FOR MANUFACTURING INSULATING PANES AND AN INSULATING PANE MANUFACTURED IN ACCORDANCE WITH THE METHOD  <b>(57) Abstract</b> <p>A method for manufacturing insulating panes and an insulating pane manufactured in accordance with the method. The pane consists of at least two glass panes having an intermediate gas filled space formed by means of a distance element between the panes. In the space one or several gases are introduced, chosen among such gases which by means of adsorption and/or chemical sorption on glass form a surface layer having a thickness corresponding to at least half a wave length within the area of heat radiation. The gases shall have a boiling point not exceeding -20 °C and preferably not over -40 °C. The surface layer shall be stable up to a temperature preferably not less than 60 °C, preferably not less than 75 °C. The gases can be introduced by being adsorbed in a porous material, which is applied in connection with the space. After mounting the pane the material will explode so that pores will be opened and the gases will be unbound.</p>		

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**TITLE:**

A method for manufacturing insulated window panes and an insulated window pane manufactured in accordance with said method

**TECHNICAL FIELD:**

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The present invention relates to a method for manufacturing of insulating panes and an insulating pane manufactured in accordance with the method. The term insulating pane refers to an element consisting of two or more glass panes which are hermetically joint to each other and separated in pairs by means of a gas filled intermediate space.

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**BACKGROUND OF THE INVENTION:**

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Glass is used for windows and similar building elements due to its property of being transparent within the visible wavelength spectrum, 0.4-0.8 micrometers. In spectrums of longer wavelengths glass is more or less non-transparent, which means that radiation within these ranges which is incident upon the glass, i.e. heat radiation, is absorbed, after which said radiation is re-transmitted off the glass in the form of radiation of long wavelengths, with half the radiation in each direction.

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Consequently, glass functions well for seeing through, but is however less advantageous for screening off sun radiation which is strong, and as insulation against heat leakage from a building at low exterior temperatures.

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In order to improve windows in these respects, certain sheet glass manufacturers have introduced coating of window glass with different types of surface coatings consisting of metals and/or metal oxides. Windows having such glass provide a decrease both of incident sun heating radiation

and of heat leakage. However, the layers imply certain drawbacks: a decreased input light flux, miscolouring of the transmitted, visible light and also increased air draught currents at a low exterior temperature as a result of a low surface temperature of the inwardly facing glass pane.

#### SUMMARY OF THE INVENTION:

The object of the present invention is to improve the reflection of heat radiation of the glass, in the form of inwardly directed sun heating radiation and heat leakage in the form of outwardly directed radiation, for use in connection with windows. In accordance with the invention, this object is solved by means of adsorption and chemical sorption of gaseous molecules on the surface of the glass. In this manner, a completely transparent surface coating with a refractive index between 1,2 and 1,3 is obtained. This does not involve any reduction or miscolouring of the visible light, at the same time that an increased sun screening and a decreased heat leakage is achieved as a result of a decreased transfer of radiation in the spectrum of wavelengths which corresponds to heat radiation. At low temperatures, a higher surface temperature is obtained on the inwardly facing glass surface as compared with said glass being coated with a transparent layer of metal/metal oxide so that the tendency for air draught currents is decreased, which results in improved heat comfort.

#### PREFERRED EMBODIMENT:

In order to achieve said object in an optimum way, i.e. to limit the heat transfer due to radiation, a surface coating having a refractive index corresponding to the square root of the index for glass, which is 1.5, is required. This gives a desired index for the coating of approximately 1.2. In order to obtain said object, the surface coating should also always be arranged on the rear side of the glass, as

regarded with reference to the direction from which the heat radiation is inputted, i.e. the non-coated surfaces of the glass pane should be turned towards the sun and the room, respectively. Both these requirements can be accomplished in the same window if at least two glass panes are put together to form a so called insulating pane, in which the surface coating is arranged on the two glass surfaces which are facing one another. The coating is thus arranged inside an intermediate space between two glass panes, and for an insulating pane these intermediate spaces can be one, two or several in number depending on the number of glass panes and they are hermetically sealed when used in an insulating pane.

The desired refractive index cannot possibly be obtained by means of solid materials due to their refractive indexes, which generally are high. The only possibility is to use a liquid film if the refractive index is to be decreased to approximately 1.2. In order to prevent such a film from running down along the glass surface, it must adhere tightly to the surface. This can be achieved by using strong molecular forces between the glass and certain liquids, chemical sorption, which results in a liquid film which remains on the glass and which must be boiled off in order to be removed. Coating the glass with a liquid is a laborious process which is difficult to perform with a result so perfect that the transparency is not affected. Moreover, many liquids do not provide the desired refractive index. Water, for example, has a refractive index of approximately 1.4. The method according to the invention, i.e. to start with a gaseous substance, is more simple and provides a perfect layer without any difficulties. In this regard, the gaseous substance is brought into contact with the glass and the substance is chemically sorbed by the glass surface, i.e. a number of molecular layers are fastened to the surface by means of

covalent bonding forces between the gas and the gas/liquid. In this regard, the molecular layer can be up to 3 micrometers thick and can be regarded as a liquid film on the glass surface. The liquid film can thus be considerably thicker than that which is required in order to affect the electromagnetic radiation, the minimum for affecting the light and heat radiation being half a wavelength.

Under the given conditions, the above-mentioned surface film increases the light transmission by a few percent at perpendicular incidence through the glass. The surface film also gives a substantially increased reflection of radiation for angles of incidence over 35°, both for light and heat radiation. As a comparison, such an increased radiation reflection is not obtained until 60° for glass without such a surface film.

The gaseous substance which is chosen for the surface film should in the liquid phase be waterproof and steamproof, i.e. water should not be able to be dissolved in the liquid film. In this manner, an additional diffusion protection against water penetration in a hermetically sealed insulating pane is achieved and the useful life for said pane increases. In practice, it is impossible during manufacture of insulating panes to obtain a seal which is so tight that water molecules cannot pass through it, which in the course of time leads to water condensation on the inner surfaces of the pane, which is something that disturbs the transparency and during practical use limits the lifetime of the insulating pane. By means of the extra "seal" which is provided by the liquid film, the transfer of water molecules is decreased, which in turn leads to an increased useful life of the pane.

For environmental reasons, the substance should neither be poisonous nor inflammable and should neither affect the

solar heating of the atmosphere or the ozone layer if it were to be released. Furthermore, the gas should not be harmful to the substance which has been chosen for sealing the surface.

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The above-mentioned selection criteria leads to the selection of a small number of substance groups such as: carbon-flouric compositions, carbon-hydrogen-flouric compositions and certain simple alcohols. The substances should be gaseous at room temperature and atmospheric pressure. They are preferably added by means of a diluting gas having a low heat conductivity. The gas mixture should have a relatively low boiling point, preferably not exceeding  $-20^{\circ}\text{C}$  and most preferably  $-40^{\circ}\text{C}$ , but should be able to form a liquid layer on the glass surface, which layer is stable up to a relatively high temperature, preferably not lower than  $60^{\circ}\text{C}$  and most preferably not less than  $75^{\circ}\text{C}$ , whereby each one of the included gases acts like a super liquid during generation of the film. It is suitable to choose a gas or gases which form the liquid film with a refractive index ( $n_g$ ) which is lower than the index for glass which is normally used as window-glass, the  $n_w$  of which is equal to 1.5. A refractive index ( $n_g$ ) below 1.4 is preferred and the most preferable is 1.2 or less.

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Examples of gas mixtures.

1. Argon,  $\text{SF}_6$  and  $\text{CHF}_3$ , where  $\text{CHF}_3$  is the active component for obtaining the liquid film.
- 30 2. Argon,  $\text{SF}_6$ ,  $\text{CF}_4$  and  $\text{CH}_2\text{F}_6$ , where  $\text{CF}_4$  and  $\text{CH}_2\text{F}_6$  are the active components for obtaining the liquid film.
3. Argon,  $\text{SF}_6$ ,  $\text{CCl}_2\text{F}_2$  and  $\text{CClF}_3$ , where  $\text{CCl}_2\text{F}_2$  and  $\text{CClF}_3$  are the active components for obtaining the liquid film.

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Other gases such as CO, CO<sub>2</sub>, NO, NO<sub>2</sub> and other hydrocarbons can be added to the mixture for assisting the absorption of heat radiation. They occur in the gas phase in the intermediate gas space, instead of or together with air.

5 However, the relatively simple gas compositions (1, 2, 3) mentioned above have per se been found to provide a very advantageous result both as regards the heat transfer in and out of a window of a building, and also as regards an increase of the surface temperature at the inside during

10 cold weather, which consequently leads to a decrease of air draught currents.

#### INDUSTRIAL APPLICABILITY:

The invention thus implies that by means of one or several

15 gases in a gas mixture, a liquid film is deposited on a glass surface in order to affect the transparency for electromagnetic radiation of the glass, especially within the heat range, the selection of gas being crucial for obtaining the desired effect. The prerequisite is thus that

20 the liquid is deposited on the inner glass surfaces of the intermediate gas space in an insulating pane; when more than two glass panes are used, such an insulating pane can have several intermediate gas spaces in which liquid films can also be deposited. Such an insulating pane of a known

25 type consists of at least two glass panes between which a frame-shaped spacer element is arranged, usually in the form of a metal tube which opens partially towards the intermediate space. This is attached to the glass panes by means of a sealing paste, which often can be butyl due to

30 its low permeability and high adhesion to glass. The metal tube is usually used as a container for a drying agent which is intended for receiving water vapour which may be present in the intermediate gas space, so as to prevent the water vapour from being deposited as condensate on the

35 glass surfaces. For manufacturing reasons, the intermediate gas space will be filled with air from the surrounding



atmosphere after assembly, and such air always contains a certain amount of moisture.

5 Some manufacturers prefer to exchange, completely or partly, the atmospheric air for a gas having a lower heat conductivity than air, the most common gases being the rare gases.

10 In this regard, a known method is to make two holes in the frame element at positions which face one another and to supply the gas which is intended to replace the air through the one hole and discharge the air through the second hole. The pane is finally sealed by means of sealing of the holes.

15 This method can also be utilized during filling of the gas which is intended to form the liquid layer according to the invention. However, this filling method is somewhat lengthy and if the air is to be replaced more or less completely  
20 the filling of gas must be carried out extremely slowly and as an even flow so that mixing due to turbulence can be avoided to a great extent. In spite of this it may be necessary to carry out a considerable overfilling in order to force out enough air, something which preferably should  
25 be avoided especially in connection with the gases according to the invention.

A new, alternative method for introducing gas will be described in the following, which method advantageously can  
30 be used in connection with the introduction of the gases, which gases according to the invention are intended to form said liquid film. The method is based on the fact that gas can be adsorbed in certain porous substances having a very large area as compared with their bulk volume. Among such  
35 substances, perlite should be especially mentioned, which is a type of volcanic glass (obsidian) which has been

exposed to high heating during which bonded water has exploded the solid material, thereby forming cavities. Zeolite is a substance with a similar structure which is constructed in a chemical way. It contains silicon, aluminium and oxygen and counter ions such as sodium, potassium or calcium. The components which are included are exchangeable and may impart suitable properties on the zeolite for example regarding cavity size, repulsion or absorption of water, properties as ion exchanger etc. These materials have in common the property of having a very large area in relation to their size. They are often referred to as molecule sieves. If these substances are exposed to a gas, this gas can be adsorbed and may then be released by means of an explosion of the material by means of ultrasonic sound, so that the pores are exposed. (The gas can also be forced out in some other way, for example by means of heating, but this should not be applied in the present case). As has been mentioned above, the zeolites can be given different properties during manufacture and can have different resonance frequencies, which means that they are exploded at different ultrasonic wavelengths. Real gases may be adsorbed, these gases being in the gas phase at normal pressures and temperatures (atmospheric pressure and in the temperature range at the surface of the ground) and preferably they contain gas molecules of the types oxygen, carbon, hydrogen, fluorine, chlorine, bromine or iodine. In certain zeolites, rare gases can be adsorbed.

These possibilities imply that gas adsorption can be limited to gases which are suitable for forming a surface film according to the invention, or that adsorption may occur for gases which solely or together with air are intended to generate a gas filling in the insulating pane. For example, rare gases can be used. It is also possible by means of a selection of the gas or gases which form the liquid layer to make the layer coloured. Through the use of

for example an iodine composition, the pane can consequently be given a green tone which makes it suitable for greenhouses, since it lets through light in the red section of the spectrum to a great extent, which is what the plants primarily need for their photosynthesis, whereas the heat radiation is limited which is desired for many plant varieties.

It should also be mentioned that the porous body can be composed of several material varieties having different resonance frequencies and also having different adsorbed gases. These gases can thereby be selectively released by means of stepwise exploding at different frequencies. In this manner, layered liquid films can be constructed and gases can be released for filling the intermediate gas space in the gaseous phase.

In the following, a few examples of filling of gas by the use of adsorption in materials of the above-mentioned kind will be explained:

1. The gas or gases which are to be introduced in the intermediate gas space of the pane are adsorbed in zeolite or perlite having pores of 5-10 Å (angstrom units). The gases can be selected among surface layer-forming gases and other gases, for example rare gases.

2. The zeolite is filled in the spacer element together with a drying agent which can be a silicate gel, the spacer element being provided with openings facing inwardly towards the inside of the intermediate space. The spacer element is provided with at least one through hole.

3. The glass panes are assembled around the spacer element and are joint thereto by means of a joining paste.
- 5 4. The assembled insulating pane is clamped during the bonding time of the joining paste.
- 10 5. The zeolite is exploded by exposing the unit to an ultrasonic sound source. The explosion takes place instantaneously.
- 15 6. The adsorbed gas is released and fills the intermediate gas space during which air is expelled through said through hole. The time for filling and expelling the air is approximately 30 seconds, but does however depend on the volume of the gas space.
- 20 7. The surface layer-forming gas or gases condense on the glass surfaces and are fixed as a liquid film by means of chemical sorption.
- 25 8. The through hole intended for the expelling is sealed by means of a final sealing, preferably by means of a sealing paste.

Since the filling of gas is carried out as an automatic process, the production cycle is not delayed more than the time it takes for sealing the expelling hole. If essentially surface layer-forming gas is used and if the air in the intermediate gas space is not exchanged, then no expelling hole is needed. Since the gas is chemically sorbed on the glass - it is thus condensated after the release - no remaining pressure increase in the pane is generated in spite of the fact that no expelling hole exists; the pressure increase will only be instantaneous during the explosion.

## 5 CLAIMS:

1. Method for manufacturing of insulating panes which consist of at least two glass panes having an intermediate gas filled space formed by a spacer element between the panes, c h a r a c t e r i z e d i n that in said space one or several gases are introduced, which are chosen among such gases which by means of adsorption and/or chemical sorption on glass form a surface layer having a thickness corresponding at least to half a wavelength within the range of heat radiation, the gas or gases having a boiling point preferably not exceeding  $-20^{\circ}\text{C}$  and most preferably not exceeding  $-40^{\circ}\text{C}$  and having the quality of forming said surface layer in the form of a liquid film on the glass, which is stable up to a temperature preferably not less than  $60^{\circ}\text{C}$ , and most preferably not less than  $75^{\circ}\text{C}$ .

2. Method according to claim 1, c h a r a c t e r i z e d i n that the gas or the gases are chosen so that the surface layer obtains a refractive index which does not exceed 1,35.

3. Method according to claim 1 or 2, c h a r a c t e r i z e d i n that the gas or gases are chosen so the surface layer which is formed is not soluble in water and/or steam.

4. Method according to claim 1, 2 or 3, c h a r a c t e r i z e d i n that one or several compositions of carbon-fluorine, carbon-hydrogen-fluorine and/or gaseous alcohols are chosen as an active gas component such as methanol, ethanol.

5. Method according to claim 4, characterized in that for said gas and gas mixture, respectively, one or several of the gases  $\text{CHF}_3$ ,  $\text{CF}_4$ ,  $\text{CH}_2\text{F}_6$ ,  $\text{CCl}_2\text{F}_2$  and/or  $\text{CClF}_3$  are chosen as an active component for obtaining said surface layer.

6. Method according to any one of the preceding claims, characterized in that except for the gas or gases forming said surface layer, one or several gases, chosen among gases which in the gaseous form have the quality that they limit the heat transfer by means of a low heat conductivity or absorption, are introduced.

7. Method according to any one of the preceding claims, characterized in that the gas is adsorbed in a porous material such as perlite or zeolite, that during assembly of the glass panes together with the spacer element the material is introduced into a space which is connected to the intermediate gas space, and that after the joining of the glass panes the material is exploded so that its pores are opened and the gas is released in the intermediate gas space.

8. Method according to claim 7, characterized in that said gas which is intended to be trapped in the intermediate gas space is adsorbed in the material, whereby during the exploding an exhaust hole is kept open, through which air trapped in the intermediate gas space can be expelled during release of said gas.

9. Method according to claim 7 or 8, characterized in that the material is exploded by being subjected to ultrasonic sound.

10. Method according to claim 9, characterized in that the material, which preferably is zeolite, is

composed of parts having different resonant frequencies through its structure, that different gases are adsorbed in the different parts and that the different parts are exploded in a sequence by being subjected to ultrasonic sound of different frequencies, each one corresponding to one of the resonant frequencies of the components, so that the different gases can be released in turn so as to form a surface layer consisting of several liquid film layers.

11. Insulating pane manufactured by means of any of the methods according to claim 1-10, consisting of at least two glass panes and joined by means of a frame element to form an intermediate gas space, characterized in that the surfaces of the glass panes which are facing towards the gas space carry layers consisting of a liquid film which is condensed from substances which are in gaseous form at atmospheric pressure and at temperatures down to  $-20^{\circ}$  and which, completely or partly, are taken from the groups carbon-fluorine compositions, carbon-hydrogen-fluorine compositions and/or single alcohols and having a layer thickness of at least half a light wavelength.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/SE 95/00283

## A. CLASSIFICATION OF SUBJECT MATTER

IPC6: E06B 3/67, E06B 3/677, C03C 17/28

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)

IPC6: C03C, E06B

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

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

DIALOG

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE, A1, 2424651 (N.V. PHILIPS' GLOEILAMPENFABRIEKEN), 4 December 1975 (04.12.75) ---	1,6,11
A	EP, A2, 0492417 (MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD), 1 July 1992 (01.07.92) ---	1,7,9
A	US, A, 4276350 (H. FRANZ), 30 June 1981 (30.06.81) --- -----	1

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Date of the actual completion of the international search

21 June 1995

Date of mailing of the international search report

27 -06- 1995

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## INTERNATIONAL SEARCH REPORT

Information on patent family members

03/05/95

International application No.

PCT/SE 95/00283

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