

ALIEN PROPERTY CUSTODIAN

NON-SPLINTERING LAMINATED GLASS

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The invention of my present application, which is a continuation in part of my former application Serial No. 38,854, filed August 31, 1935, relates to laminated glass, in which the glass cover sheets are supported by a flexible intermediate layer.

Intermediate layers have long been employed for the purpose of holding splinters and of cementing the cover sheets together, their function in the case of fracture of the covering layers, being to prevent the glass splinters resulting from the fracture from being released, the intermediate layers by their adhesive action retaining such splinters in position. Security against fracture of such glass, which is the most important requirement if it is to have the character of safety glass, was not, however, at first ensured, since the thin intermediate layers then used were unable to prevent fracture. A certain degree of security against such fracture was subsequently obtained by employing tough, plastic intermediate layers e.g. of a natural or artificial resin with which has been mixed a small percentage of a vulcanisable rubber in solution, if desired with dissolved softening agents, and covering sheets of high oscillating powers, i.e. by employing glass covering sheets of only slight thickness e.g. 1 to 2 mms. the thickness of such intermediate layers being of at least half the thickness of the glass covering sheets employed.

Nevertheless the defect remained that relatively thin glass covering sheets of the laminated glass were unable to offer adequate resistance to additional stresses or strains arising during the construction or transport of the laminated glass but owing to the high flexibility of the intermediate layers, reached the breaking limit when subjected to bending strains, and were fractured.

This disadvantage resulted from the fact that the highly flexible intermediate layer could not afford sufficient resistance to the flexibility of the oscillatable covering sheets.

If, then, it was difficult with these oscillatable glass covering sheets to obtain an intermediate layer, which took up satisfactorily the normal stresses produced by warping or by transport of the additional stresses due to deformations taking place during the running of road or rail vehicles, it was even more difficult to combine thicker glass covering sheets having lower oscillating powers that is to say higher resistance to warping stresses or additional running stresses, because the high stresses in the intermediate layer resulting from deformation of the thicker covering sheets, were not counteracted by any adequate damping resistance.

With covering sheets of the order of 2,5 to 3 mm. in thickness, which have relatively low oscillating or flexing powers, but which are resistant to warping stresses, or the additional constructional or running stresses, intermediate layers which are thin and very flexible cannot be effectively used. Such covering sheets must be given a support capable by reason of its tenacity and elasticity of absorbing the bending forces, even under the strongest bending stresses, exerting a breaking action before any oscillation reaches its maximum.

This fact is taken into account by the present invention, which enables compound glass, having the maximum security against splintering and fracture, to be produced even when covering sheets which are practically non-oscillatory are used, and in which even the greatest constructional, transport or running stresses are scarcely capable of oscillating even through half a complete oscillation owing to their stability. It is furthermore highly desirable that these properties should persist throughout the ranges of atmosphere temperatures met with in all parts of the world.

The present invention involves two factors, both leading to the desired result, firstly, the choice of substance for the intermediate layer, and secondly the choice of the most suitable proportionate thickness of this layer in reference to the cover plates.

As substance I choose a polymer of vinyl acetate or a polymer of an acrylic acid ester, or a mixture of these polymers, the polymerisation being carried to the stage defined below and the polymer being treated as hereinafter described.

In order to obtain the maximum efficiency, however, this intermediate layer so prepared should be approximately 70 per cent of the thickness of the thickest covering sheet employed. The fact that covering sheets of different thickness are used being immaterial. Below this point the efficiency of the layer for the purposes above-mentioned falls off rapidly, whilst a further increase in the thickness does not lead to any increase of efficiency commensurate with increased cost.

Although it is known that products of middle stage polymerisation of vinyl acetate or acrylic esters (as hereinafter defined) which possess great elasticity and flexibility, are well suited for use as intermediate layers to produce a non-splintering glass with adequate security against breakage, it has only recently been discovered

that, with thick layers, this security is confined to certain temperatures.

According to the present invention therefore the intermediate layer is formed from a polymer of vinyl acetate or from a polymer of an acrylic acid ester or from a mixture of these polymers, the degree of polymerisation being between approximately 720 and 800 (cf. H. Staudinger "Die hochmolekularen organischen Verbindungen" (1932) pp. 41 to 45) i. e. a point between the middle and final stages for the following reasons:

The substances which are polymerised, and more particularly the one most generally used, viz. vinyl acetate, are given great resistance against cold and heat if the polymerisation is carried to the aforesaid degree of polymerisation.

As is well known, polymerisation consists in the union of molecules having unsaturated linkages to form a single molecule. The molecules so formed give a substance of excellent toughness and tensile strength and this elastic toughness and tensile strength is at the maximum when the polymerisation is almost complete. If the polymerisation was allowed to proceed to the final stage, however, difficulties would arise, in that the product being saturated would be very difficultly soluble.

Great difficulty has, however, been experienced in employing the aforesaid polymerisation products of a degree of polymerisation between approximately 720 and 800 for the purpose now sought, since it is necessary that permanent softening of this highly molecular body should be effected.

It has now been found that, contrary to prevailing ideas, these products of high molecular weight can be made to retain their properties permanently by the use of only a small proportion of softening agents. As, according to this invention, the solvents are not used for the production of the intermediate layers, the agents intended to produce permanent softening are added by the following method.

A polymerisation product which has reached the above described stage is a highly viscous mass of excellent tensile strength and elasticity, without being hard or brittle like crystal or glass, which would be the case with a product polymerised to the final stage. This product is disintegrated, and there is then added from seven to twenty percent, of a softening agent, the mixture being then heated and formed into a layer whose thickness is approximately 70% of that of the thicker cover glass. The mixture may be conveniently worked up in an autoclave by the use of suitable agitating means and heating up to approximately 80° C. so as to form a homogeneous mass. This mass cannot be made to flow at this temperature after absolute homogeneity has been obtained, but it is converted by the use of considerably higher temperatures, which may, if necessary, be raised to approximately 140° C., into a fluid or incompressible condition, and then is immediately forced out of the vessel by high pressure, and at once formed into plates or foils from the desired form. The heating from 80° to 140° C. must take place relatively rapidly (for example during 60 to 80 minutes) so that no substantial further polymerisation takes place. It is important in this operation to employ as softening agents those high point esters which, at the temperatures in question, do not vary either in their colour or softening properties, since the intermediate layer must be fast to light and preserve its colour.

Suitable "softening agents" for the purpose of this invention are tributyl phosphate, tricresyl phosphate and dibutyl phthalate.

These intermediate layers are produced by one operation whatever the thickness required, and the former lengthy treatment for the purpose of removing bubbles and evaporating off the solvents is avoided.

Intermediate layers so produced have no powers of adhesion owing to the fact that no volatile solvents have been employed, and it is therefore necessary to provide for permanently connecting the intermediate layer of the sheets of glass in such a manner that splinters or fragments do not become detached even when the sheet is completely destroyed. It is advisable to use as an adhesive a strongly cementing solution produced from a middle stage polymerisation product of vinyl acetate or of an acrylic acid ester or mixture of these polymers dissolved in so-called genuine solvents of high boiling point.

By the term "genuine solvents of high boiling point" as used herein is meant difficultly volatilisable organic substances which are not soluble in, miscible with or capable of absorbing water, which have molecular weights of at least 100, and which boil without decomposition at ordinary pressure or in vacuum, their boiling points being not less than 165° C. at normal atmosphere pressure. Example of such genuine solvents of high boiling point are benzyl alcohol, glycol monobutylether, methyl cyclohexanol, methyl cyclohexanone, the acetate of methyl-1,3-butylene glycol, benzyl acetate and cyclohexyl acetate.

Apart from the strong adhesive powers of such solutions there is an advantage that foils covered with such solutions and used as intermediate layers absorb a part of these high boiling point solvents, and enter into intimate connection with the adhesive, and further the portions of the solvents which diffuse into the foils reinforce the action of the softening agents. I make no claim, however, to the use of any particular class or classes of adhesives.

I will now (by way of example only) describe two instances of these further stages of treatment after polymerisation.

Example No. 1

85 parts by weight of polymerised vinyl acetate having a degree of polymerisation of 747 are disintegrated and placed in an autoclave provided with stirring mechanism, and 14 parts of dibutyl phthalate, with a boiling point of 200-216° C. at 20 mm. pressure are poured in. The autoclave is then shut and the stirrer slowly rotated. The steam pipes surrounding the autoclave are raised to a temperature of 120° C., and the rate of rotation somewhat increased. When the temperature of the jacket of the autoclave is seen to be 120° C., which would mean that the contained mass was at about 80° C., the rate of rotation is raised to 120 revolutions per minute. The homogenisation is continued at this temperature for three hours, when the mass should be completely homogenised. The temperature of the homogenised mass is now raised to 140° C. (jacket temperature 180° C.) during two hours and the mass is then drawn off by a suction pump and applied to the cover sheet (to which an adhesive consisting of a middle stage polymerisation product of vinyl acetate in one of the aforesaid genuine solvents of high boiling point has previously been applied) by pressure through an application device.

The layer is formed with a thickness of 70 per

cent., of that of thicker cover sheet. The mass becomes solid immediately since no solvents have been used, the other cover sheet, to which an adhesive has previously been applied, is laid on, and the whole united in a press, at a temperature of 80° to 100° C.

Example 2

85 parts by weight of polymerised acrylic acid ester having a degree of polymerisation of 760 are disintegrated, 15 parts of diamyl phthalate with a boiling point of 210-220° C. under 20 mm pressure is added and the mixture treated as above described. A glass sheet covered with this intermediate layer is united by means of an adhesive with a second glass sheet according to the directions given in Example 1.

The two characteristic features of the invention are firstly the use of suitable highly viscous polymerisation products of high molecular weight, which can be converted into a fluid state by appropriate addition of softening agents and by the use of appropriate temperatures, and which can be formed under pressure into sheets of the required dimensions, and secondly that these intermediate layers are approximately seventy per cent. of the thickness of the thicker cover sheet of the laminated. Suitable glass for such purposes as windscreen can be formed with cover glasses of say 3 mm. thickness and an intermediate sheet of approximately 2 mm. If the cover

glasses are of different thicknesses the thinner sheet should preferably be disposed on that side from which a blow is probable.

Since this invention enables glass of any desired thickness to be used, shop windows and the like may now be produced composed of laminated glass.

Laminated glass according to this invention is also found to be useful as walls for protection against heat and sound. The laminated glass according to the invention is therefore, also particularly suitable for use in the construction of telephone boxes for public telephone systems, and for microphone cells for wireless transmissions.

Another use of this glass is for the walls of explosion chambers and the like, where it is desirable to be able to inspect the interior, and at the same time protection must be afforded against any explosion. For example a suitable laminated glass for this purpose can be made from a sheet 10 mm. in thickness of ordinary window glass and another similar sheet of 4 mm. thickness, which are united by an intermediate layer of 7 mm. thickness.

By the expression "approximately 70%" as used herein is meant 60%, to 80%.

By the expression "middle stage polymerisation product" as used herein is meant one having a degree of polymerisation between 320 and 380.

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