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Production of surface-enhanced moulded fibre parts

30 DEC 1999

The invention relates to the production of mouldings made from fibre web based on natural and/or synthetic fibres, where at least one of the large-area surfaces, generally the visible side, has been enhanced by a coating process, in particular by a high-quality lamination process.

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- 10 Mouldings of this type are used, inter alia, in automotive construction.

- 15 Moulded fibre parts are sheet-like mouldings made from fibre webs, in particular made from reprocessed fibre material, consolidated with thermoset resin, in particular with phenolic resin or epoxy resin.

- To produce these, fibre is mixed with resin powder, shaped to give a web and if desired precured (prehardened). In the case of sheet-like web materials, 20 the curing takes place directly after formation of the web in the oven, or the prehardened web is cut into panels and cured in heated press moulds under pressure, with shaping. The mouldings are then stamped out (cf. Kunststoff-Handbuch [Plastics Handbook], Vol. 10 25 "Duroplaste" [Thermosets], Hanser Verlag, 2nd edition (1988), pp. 763-775). The surface enhancement can take place during the curing of the fibre web or in a second and separate step.

- 30 During the lamination process, thermoplastic adhesive (hot-melt adhesive) or specific contact adhesives are used for adhesive-bonding of the sheet material which will subsequently be visible, the laminating material, to the moulding made from web material, which by then 35 serves as a supporting base.

The coating process involves applying pigmented thermoplastic compositions in the form of pastes or from solutions.

This Sheet Has Been Replaced



- 2 -

In an improved process, a pulverulent thermoplastic mixture of low melt viscosity is spread onto the surface of the laminating material, melted by reheating, and then cooled. This gives a uniform
5 coating on the surface of the lamination material.

The disadvantage of this process is that the recovery forces in the laminating material when hot are frequently more powerful than the adhesive strength of
10 the hot-melt adhesive. In the case of parts with complicated shapes in particular, this leads to defects where the laminating material parts from the substrate. These separation phenomena are particularly marked in the case of mouldings exposed to frequent temperature
15 changes or intense insolation.

The object of the invention is therefore to provide a process intended for the surface enhancement of moulded fibre parts which has similar technical elegance and
20 similar simplicity to the hot-melt coating process mentioned, but in which disadvantageous separation phenomena do not occur.

The object is achieved by a process according to Claims
25 1 to 10, and also by moulded fibre parts produced according to one or more of these claims.

It has been found that moulded fibre parts can also be given a high-quality enhanced surface if the adhesive
30 used for the lamination process is a thermoset resin or resin-hardener mixture which has not yet been fully cured and is nontacky at room temperature. Laminating material provided with adhesive of this type is applied to the prefabricated fibre web or to mouldings made
35 from fibre web, and the resultant laminated article is heated to a temperature above the initiation temperature for the curing reaction of the resin or of the resin-hardener mixture.

This Sheet Has Been Replaced

- 3 -

During this, the thermoset resins cure in the form of a film which bonds to the fibre-web surface during the curing reaction and gives, over the entire surface, irreversible adhesive bonding of the lamination material so that no separation phenomena result, even on exposure to extreme temperature differences or insolation.

This is surprising, since the melt viscosity of thermosets is generally higher than that of thermoplastics and the curing temperature used is significantly above the melting point of the thermoplastic binders used hitherto, and therefore the recovery forces arising when the pressure is reduced in the press tool are significantly higher than when thermoplastics are used.

Thermoset resins are generally brittle. It is therefore also surprising that the lamination materials coated according to the invention with thermoset resins remain stable even when rolled up or shaped, and do not break apart.

Another advantage of the novel process is that the moulded fibre parts may be surface-enhanced by applying the thermoset resins or resin-hardener mixtures which have not yet fully cured either to prefabricated mouldings made from web material or to the laminating material. This method of processing also allows the thermoset resins used for surface enhancement to crosslink at the interface with the resins used as binders for the textile fibres. This applies particularly if the resins used for binding the fibres and for enhancing the surfaces of the mouldings are identical or react with one another.

Another advantage arising from these crosslinking reactions at the interface is an increase in the stability of the mouldings.

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- 4 -

According to the invention, therefore, thermoset resin or, respectively, resin-hardener mixture and a laminating material are applied to the surface(s) to be enhanced of prefabricated mouldings and the resultant units (laminated articles) are heated to temperatures above the melting point and above the initiation temperature for the curing reaction of the thermoset resins. Once the thermoset resins have cured, the resultant surface-enhanced parts are further processed in a manner known per se.

Another simplification of the overall production process results from applying thermoset resin, resin-hardener mixture and a laminating material to the surface(s) to be enhanced of prefabricated or prehardened fibre webs. The parts produced in this way (laminated articles) are heated, under pressure and with shaping, to temperatures above the melting point and above the initiation temperature of the curing reaction of the resins used in the coating and above the initiation temperature of the resins used as binders in the web. During this there is simultaneous curing of the binders in the web and the resins in the coating and, if the binders in the web and the resins of the coating are identical or can react with one another, these thermosets crosslink with one another at the interface.

Once the coating and the binders in the web have cured, the resultant parts are further processed in a manner known per se.

Possible versions of the process for surface enhancement with the use of a laminating material are given below:

- the thermoset resin or resin-hardener mixture is applied to fibre web which has not yet been heat-treated or prehardened, and is heated to a



- 5 -

temperature above the melting point but below the initiation temperature for the curing reaction of the resin. During this the resin is melted and disperses to give a thin layer. The laminating material is applied to this layer, either directly or after interim cooling and reheating, and the resultant product is then heated, if desired with shaping, to a temperature above the initiation temperature for the curing of the resin, and, once the thermoset resin has cured, is cooled and further processed in a manner known per se.

• The thermoset resin or resin-hardener mixture is applied to laminating material and heated to a temperature above the melting point but below the initiation temperature for the curing reaction of the resin. During this, the resin is melted and disperses to give a thin layer. The laminating material thus provided with adhesive is applied, either directly or after interim cooling and reheating, to fibre web which has not yet been heat-treated or prehardened, and the resultant product (laminated article) is then heated, if desired with shaping, to a temperature above the initiation temperature for the curing of the resin, and, once the thermoset resin has cured, is cooled and further processed in a manner known per se.

• The thermoset resin or resin-hardener mixture is applied to a fully cured and preshaped moulded fibre part and heated to a temperature above the melting point but below the initiation temperature for the curing reaction of the resin. During this, the resin is melted and disperses to give a thin layer. The laminating material is applied to this layer, either directly or after interim cooling and reheating, and the resultant product is then heated to a temperature above the initiation temperature for the curing of the resin, and, once the thermoset resin has cured,



- 6 -

is cooled and further processed in a manner known per se.

- 5 • The thermoset resin or resin-hardener mixture is applied to laminating material and heated to a temperature above the melting point but below the initiation temperature for the curing reaction of the resin. During this, the resin is melted and disperses to give a thin layer. A fully cured and preshaped
10 moulded fibre part is applied to this layer, either directly or after interim cooling and reheating, and the resultant product is then heated to a temperature above the initiation temperature for the curing of the resin, and, once the thermoset resin has cured,
15 is cooled and further processed in a manner known per se.

In the first two versions it is advantageous for the thermoset resin used for the coating process to
20 comprise a resin which can be cured using formaldehyde, in particular a hardener-free novolak or hardener-free modified novolak, if the binder in the fibre web is a mixture of a resin curable using formaldehyde and a substance which releases formaldehyde when heated, for
25 example a novolak-hexamethylenetetramine mixture. The formaldehyde released when this binder is cured also suffices for the curing of the thin layer of molten novolak.

Another advantage of this version of the process is
30 that there is a significant reduction in formaldehyde emissions during the production process.

Preferred methods of carrying out the process are given below:

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- In a continuous production process and in the form of a continuous web, laminating material provided with an adhesive which is a thermoset resin or resin mixture curable using formaldehyde is applied to the

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- 7 -

5 prefabricated or prehardened fibre web comprising a
resin curable using formaldehyde and a hardener
releasing formaldehyde when heated, and the resultant
laminated article is then cured under pressure at a
temperature above the initiation temperature for the
curing reaction of the binder used in the fibre web
and that of the adhesive, and the formaldehyde
released in the curing reaction of the binder serves
as hardener for the adhesive. This gives a fully
10 cured web material from which sheets of the desired
size and shape are stamped out or cut out.

15 • In a continuous production process and in the form of
a continuous web, laminating material provided with
an adhesive which is a thermoset resin or a resin
mixture curable using formaldehyde is applied to
prehardened or merely prefabricated fibre web
comprising a resin curable using formaldehyde and a
hardener releasing formaldehyde when heated, and the
20 resultant laminated article is then annealed at a
temperature above the melting point of the adhesive
and, providing a fibre web is used which has not yet
been prehardened, above the melting point of the
resin used as binder, and is then cooled and then
25 stamped out or cut to shape. The laminated article
(semifinished product) which has been stamped out or
cut to shape is then cured under pressure, with
shaping, at a temperature above the initiation
temperature for the curing reaction of the binder
used in the fibre web and that of the adhesive, and
30 the formaldehyde released in the curing reaction of
the binder serves as hardener for the adhesive.

35 • Alternatively, in a batchwise process, cut-to-shape
laminating material provided with an adhesive which
is a thermoset resin curable using formaldehyde is
applied to the prefabricated or, respectively,
prehardened and cut-to-shape fibre web comprising
resin curable using formaldehyde and a hardener

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- 8 -

releasing formaldehyde when heated, and the resultant laminated article is then cured at a temperature above the initiation temperature for the curing reaction of the binder used in the fibre web and that of the adhesive, and formaldehyde released in the curing reaction of the binder serves as hardener for the adhesive.

The pressure used in these processes is of the same order of magnitude as used hitherto in corresponding processes of the prior art.

A preferred method for cut-to-shape laminating mouldings made from fibre web is to apply laminating material provided with an adhesive which is a resin-hardener mixture to cured and shaped mouldings made from fibre web and to heat the resultant laminated article above the initiation temperature for the curing reaction of resin-hardener mixture.

An advantage of laminating prefabricated, moulded fibre parts is that less heat treatment is needed to cure the thin layer of thermoset resin than to cure a textile web. This means that products with relatively low heat resistance can be used as laminating material.

The fibre used for the novel process may be the same as that used in conventional production of moulded fibre parts. This is essentially reprocessed fibre material from textile recycling and is composed predominantly of cotton fibres or polyester fibres, but generally also comprises other fibres, e.g. wool, jute, flax, sisal, nylon fibres or acrylic fibres. The fibres may comprises additives known per se, e.g. lubricants, spin finishes, fungistats or hydrophobicizers. The fibre material used may also comprise inorganic fibres.

The production of the prehardened fibre webs or of the moulded fibre parts also takes place in a manner known

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- 9 -

per se, as described, for example, in Kunststoff Handbuch [Plastics Handbook] Vol. 10 "Duroplaste" [Thermosets], pp. 766-768.

- 5 The laminating materials used may likewise be any sheet material, e.g. high-quality nonwovens, knits or wovens, as used hitherto for laminating moulded fibre parts, as long as they are sufficiently heat-resistant not to be damaged at the curing temperatures for the thermoset
10 resins.

Suitable resins for the adhesive bonding of the lamination materials are in principle any thermoset resin or resin-hardener mixture which is nontacky at
15 room temperature, or also mixtures comprising these resins and, if desired, hardeners, with modifier resins known per se. Examples of these curable resins are epoxy resins, unsaturated polyesters, urea-formaldehyde resins, melamine-formaldehyde resins, melamine-phenol-
20 formaldehyde resins and phenol-aldehyde resins, and also combinations of these resins with one another or with other modifier resins, in particular with from 5 to 50% of flexibilizing resins, e.g. polyvinyl butyral, polyvinyl alcohols or phenoxy resins. Preference is
25 given to phenol-formaldehyde resins.

The direct coating of the moulded fibre parts or of the uncured or, respectively, the prehardened fibre webs, or of the laminating materials, takes place by
30 processes known per se, using spreading, rolling, trowelling, casting, dipping or spraying methods, using appropriate pastes, aqueous dispersions or solutions in organic solvents.

35 The powder method is the preferred coating process. In this, the thermoset resins or resin-hardener mixtures are applied as finely ground powders to the surfaces to be coated. Particularly suitable materials for this are resins or resin-hardener mixtures whose melt viscosity

This Sheet Has Been Replaced



- 10 -

when subsequently heated is sufficiently low to allow adequate wetting of the substrate. The preferred melt viscosity, measured at 175°C, is less than 1000 mPas. During this short period of heating above the melting point of the thermoset resins, preferably using IR radiant heaters, a thin coherent film of the resin or resin-hardener mixture forms on the textile web or laminating material and rapidly cools, thus becoming solid and nontacky.

10 In the next process step, fibre web and laminating material are combined with one another by one of the process methods described above and bonded to one another using pressure and heat. The resultant moulding is then cooled and further processed in a manner known

15 per se.

Example 1

The thermoset resin used for surface enhancement in the example is a phenol novolak which has been modified with 10% phenoxy resin and has a melting point of 70°C and a melt viscosity, measured at 175°C, of 420 mPas.

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Aerodynamic methods of web formation are used to produce an as yet uncured textile web with 25% content of phenol novolak-hexamethylenetetramine as binder (base web).

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A web (40 g/m²) of high visual quality (outer web) coated with a thin layer (20 g/m²) of the modified phenol novolak is added to one side of this fibre web and fixed to the base web using heat (75°C) in an oven. The web laminated in this way is then subjected to a shaping and curing process in a hot tool at from 180 to 220°C. During this, the formaldehyde released from the hexamethylenetetramine present as a hardening agent in the base web suffices for full concomitant curing of the thin phenol novolak layer present on the inner side of the laminated-on outer web. This gives a moulded textile part with a thermoset adhesive-bonded layer

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- 11 -

which enhances its surface. Another advantage of this method of carrying out the process is a marked reduction in the resultant emission of formaldehyde from the base web.

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Example 2

The thermoset resin used for surface enhancement in Example 2 is a phenol novolok which has been modified
10 with 10% by weight of phenoxy resin and has a melting point of 70°C and 6% by weight of hexamethylenetetramine.

A thin layer of this finely ground resin-hardener mixture is spread onto one side of a cured and shaped
15 moulded textile part and heated above the melting point. Once the moulded textile part has been coated, a needled web of high visual quality is applied to it. As a result of rapid cooling, the high-quality web is fixed on the moulded textile part by means of a
20 remeltable, high-flexibility thin film.

In a second process step, heat and pressure are used to cure the thin coating above the initiation temperature for the curing reaction of the resin (90s at 150°C).

25 This method of carrying out the process permits the laminating material used for surface enhancement to have a heat resistance lower than that in Example 1. Another advantage of this version of the process is that the thermoset binder in the web and the thermoset
30 resin used for surface enhancement can have different chemical structures and different chemical crosslinking reactions.

Example 3

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The thermoset resin used for surface enhancement in Example 3 is a phenol novolok which has been modified with 10% by weight of phenoxy resin and has a melting

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- 12 -

point of 70°C and 6% by weight of hexamethylenetetramine.

A thin layer of this finely ground resin-hardener mixture is spread onto one side of a needled web of high visual quality and heated above the melting point of the resin. A prefabricated, cured moulded textile part is applied to this coated web. As a result of rapid cooling, the high-quality web is fixed to the moulded textile part by means of a remeltable, high-
10 flexibility thin film.

In a second process step, heat and pressure are used to cure the thin coating at above the initiation temperature for the curing reaction of the resin (90s at 150°C).

This Sheet Has Been Replaced



- 13 -



Patent claims

1. Process for producing surface-enhanced moulded fibre parts by using a laminating material or a thermoset coating composition to coat the surface(s) to be enhanced, characterized in that the adhesive used comprises a thermoset resin or resin-hardener mixture which is nontacky at room temperature.
2. Process according to Claim 1, characterized in that, in a continuous production process and in the form of a continuous web, laminating material provided with an adhesive which is a thermoset resin curable using formaldehyde is applied to the prefabricated or prehardened fibre web comprising a resin curable using formaldehyde and a hardener releasing formaldehyde when heated, and the resultant laminated article is then cured under pressure at a temperature above the initiation temperature for the curing reaction of the binder used in the fibre web and that of the adhesive, and the formaldehyde released in the curing reaction of the binder serves as hardener for the adhesive.
3. Process according to Claim 1, characterized in that, in a continuous production process and in the form of a continuous web, laminating material provided with an adhesive which is a thermoset resin curable using formaldehyde is applied to the prefabricated or prehardened or merely prefabricated web comprising a resin curable using formaldehyde and a hardener releasing formaldehyde when heated, and the resultant laminated article is then annealed at a temperature above the melting point of the adhesive and, if desired, of the binder used in the fibre web, and then cooled and then stamped out or cut to shape, and the





- 14 -

laminated article which has been stamped out or cut to shape is then cured under pressure, with shaping, at a temperature above the initiation temperature for the curing reaction of the binder used in the fibre web and that of the adhesive, and the formaldehyde released in the curing reaction of the binder serves as hardener for the adhesive.

4. Process according to Claim 1, characterized in that, in a batchwise process, cut-to-shape laminating material provided with an adhesive which is a thermoset resin is applied to the prefabricated or, respectively, prehardened and cut-to-shape fibre web comprising phenolic resin and hardener, and the resultant laminated article is then cured at a temperature above the initiation temperature for the curing reaction of the binder used in the fibre web and that of the adhesive, and formaldehyde released in the curing reaction of the binder serves as hardener for the adhesive.
5. Process according to Claim 1, characterized in that cut-to-shape laminating material provided with an adhesive which is a resin-hardener mixture is applied to cured and shaped mouldings made from fibre web and the resultant laminated article is then heated above the initiation temperature for the curing reaction of the resin-hardener mixture.
6. Process according to one of Claims 1 to 5, characterized in that the resin used as adhesive is a hexamethylenetetramine-free novolac.
7. Process according to one of Claims 1 to 6, characterized in that the resin used as adhesive is a modified phenolic resin.

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- 15 -

8. Process according to Claim 7, characterized in that the resin used as adhesive is a phenoxy-resin-modified phenolic resin.
- 5 9. Process according to one of Claims 1 to 8, characterized in that the resin or resin-hardener mixture is applied as a powder to the laminating material.
- 10 10. Process according to Claim 9, characterized in that the melt viscosity of the resin or resin-hardener mixture, measured at 175°C, is less than 1000 mPas.
- 15 11. Moulded fibre part produced according to one or more of Claims 1 to 10.

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