

Thermoset fibrous mouldings and method for producing the same

The invention relates to mouldings made of fibres which are, for instance, used as conditioning, sound absorbent parts and mouldings with a high surface bonding strength and splinter resistance for the manufacture of automobiles.

Usually, such mouldings are produced from layers of fibres containing phenolic resins as binders (non-wovens) by means of preforming and simultaneous curing.

These products show the disadvantage that at some time, in particular if exposed to high temperatures and humidity, they diffuse a disagreeable smell which mainly results from the curing agent (hexamethylene tetramine) and/or its amine decomposition product.

According to EP-A 0 254 807 this problem is solved in that a powdery mixture of a non-thermoreactive phenolic resin and one or several condensation polymers from the group of phenolic resin, amino resin or epoxy resin are used as a binder. Although the disagreeable smell caused by hexmethylene tetramine is avoided, there remains some smell caused by the phenolic resin.

Thus, it is an objective of the invention to provide thermoset fibrous mouldings which continue to show the good mechanic, sound absorbent and fire resistant properties of fibrous mouldings containing phenolic resin, but avoid, as far as possible, the diffusion of any disagreeable smell caused by phenolic acid. It is a further objective of the invention to produce fibrous mouldings with an increased strength in order to achieve a reduction in the weight of the respective car parts without impairing the quality of the same.

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The problem is solved by the thermoset fibrous mouldings according to claims 1 to 4 as well as by the method for preparing them according to claims 5 to 8.

It was found that the problem can be solved by means of thermoset fibrous mouldings consisting of two or more thermoset layers of fibres, wherein at least two layers of fibres contain different thermosetting binders.

Thermosetting binders are self-curing resins or resins to which curing agents or accelerators are added which harden to form a highly polymeric product if a certain temperature is exceeded. Suitable resins are diallyl phthalate resin, epoxy resin, polyurea, melamine resin, melamine-phenolic resin, phenolic resin and unsaturated polyester resin and their combinations.

Preferred binders are those based on phenolic resin and epoxy resin.

The preferred embodiment of fibrous mouldings thus consists of one or more internal layers of fibres containing binders on the basis of phenolic resin. These are referred to as the inside layer. At the upper and lower side of the inside layer, layers of fibres are provided as covering layer containing binders on the basis of epoxy resin.

In this respect it is surprising that the fibrous mouldings thus produced do not diffuse any smell of phenolic resin or amine decomposition products of hexamethylene tetramine although the individual thermoset layers of fibres and thus the entire fibrous mouldings are permeable to gas and air. It is a further unexpected advantage of the said fibrous mouldings that they show an increased strength which renders it possible to obtain the same strength through the thinner fibrous mouldings according to the invention which can be obtained through the heavier conventional fibrous mouldings containing only phenolic resin as a binder.

In particular, the strength is improved and the smell diffusion decreased if the fibrous mouldings with thermosetting binders are produced according to the carding method.

Binders on the basis of phenolic resin can be all condensates of phenolic compounds and aldehydes, in particular of phenol, cresol or xylenol and formaldehyde, as well as resols and common novolak resin curing agents, in particular mixtures of novolak hexamethylene tetramine. The applied phenolic resins are, as a rule, powdery.

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Binders on the basis of epoxy resin are, as a rule, powdery mixtures of epoxy compounds comprising at last two epoxy groups per molecule and one curing agent. Curing agents are preferably latent curing agents and those which allow sufficient processing time in between mixing and curing. Examples for such curing agents are acid anhydrides, imidazole derivatives, preferably novolaks or metal complex compounds as e.g. known from EP-B 0 518 908.

As fibres anorganic fibres such as e.g. glass fibres or organic fibrous material or random fibrous mixtures can be used.

Preferably, organic fibres such as wool, cotton, viscose staple fibre, jute, flax, hemp, polyester and acrylic fibres are used.

A large part of the used fibrous material is gained from textile waste through a willowing process.

These fibres are mixed with the respective binders according to known methods and are laid as individual layers of fibre (fleece, in particular carded fleece).

According to the invention several layers of fibres (fleece paths) comprising at least two different thermosetting binders are overlaid.

In doing so, preferably one or several layers of fibres containing phenolic resin as a binder are used together to form the inside layer, which layer is provided with one or several covering layers on its upper and lower side containing epoxy resin as a binder.

The individual layers of fibres or the layers (half-stuff) are cut in a known manner and can be precured or be cured at a later time during preforming, or the layers are immediately cured at temperature exceeding the curing temperatures of the thermosetting binders for regular products.

In the easiest case fibrous webs, each of which contain different binders, are combined with each other to form half-stuff and are pressed and cured, and preformed if required, at a temperature exceeding the curing temperature of thermoset binders.

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As an example, a hat rack having a basis weight of 2500 g/m² is manufactured from two half-stuffs weighing 500 g/m² each and containing a mixture of an epoxy resin curing agent as a binder and an intermediate half-stuff weighing 1500 g/m² containing novolak hexamethylene tetramine as a binder.

The preferred method for producing the thermoset layers of fibres according to the invention is the carding method, wherein the fibres are combined to become very fine webs to which binders are then added and which are laid up in longitudinal and cross bands until the desired basis weight is reached, wherein the upper, intermediate and lower layer can contain different binders, as desired. Also in this example the intermediate layers preferably contain phenolic resins as binders, whereas the upper and lower layers on the outside preferably contain epoxy resins as binders.

According to the most preferable process fibrous webs (layers of fibres) are produced in one process step according to the carding method, wherein the continuously produced fibre path in longitudinal direction is sanded with different binders over three sections of similar or different widths such that the inside section contains the binders for the inside layer of the moulding and the two outside sections contain the binder or binders for the upper and lower layer on the outside of the moulding.

The web path thus produced is layered according to the cross-layer method such that the new web path comprises layers of fibres containing different binders.

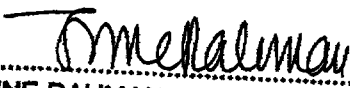
The said web path is heated for a short period of time allowing the binders to melt and fix on the fibre but not to cure. The web path is then cooled and packed. Finally, the half-stuff thus produced is pressed and cured, and preformed if required, according to a known method, wherein the half-stuff can be laid up in several layers and be pressed and cured.

Example

The used phenolic resin binder is a phenolic novolak having a softening point of 98°C mixed with 6 weight-% hexamethylene tetramine.

The used epoxy resin binder is a mixture of an epoxy resin on the basis of bisphenol A (epoxy equivalent: 183; softening point: 75°C) and 5 weight-% of a latent curing agent consisting of a pulverised "solid solution" of 30% 2-methyl imidazole in 70% of high-melting phenolic novolak.

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According to the carding method a fibrous web is continuously produced from a mixture of textile fibres and a powdery resin binder. The web path is sanded with different binders in longitudinal direction over three sections of similar width. The two outside sections are sanded with epoxy resin binders and the inside section with phenolic resin binders.

The web path thus produced is layered according to the cross-layer method to form a new web path comprising an inside layer with phenolic resin as a binder and an upper and a lower layer on the outside with epoxy resin as a binder.

The said web path is led quickly through a forced-air oven heated to 150°C such that the binders melt and fix on the fibrous without being cured.

The path is then cooled, packed and then pressed and cured for 60 s at 180-190°C and 170 bar while being preformed.

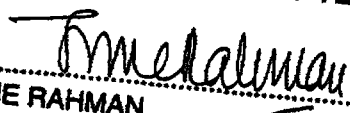
The thermoset fibrous moulding thus produced is self-extinguishing and shows a low flue gas ratio when exposed to external flames. The fibrous moulding remains inodorous also when exposed to higher temperatures (40-70°C) and humidity (90° relative humidity of air). Its strength is increased by 27% compared to a fibrous moulding exclusively containing phenolic resin as a binder and having the same basis weight.

A separation of individual layers, e.g. cracks, is not found.

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Patent claims

1. Thermoset fibrous mouldings, characterised in that they consist of two or more thermoset layers of fibres, wherein at least two layers of fibres contain different thermosetting binders.
2. Thermoset fibrous mouldings according to claim 1, characterised in that they contain binders on the basis of phenolic resin and epoxy resin.
3. Thermoset fibrous mouldings according to claims 1 and 2, characterised in that they consist of one or several internal layers of fibres, the inside layers, which contain binders on the basis of phenolic resin and of one or several upper and lower layers of fibres, the outside layers, which contain binders on the basis of epoxy resin.
4. Thermoset fibrous mouldings according to claims 1 to 3, characterised in that the layers of fibres comprising thermosetting binders are produced according to the carding method.
5. Method for producing thermoset fibrous mouldings according to claims 1 to 4, characterised in that at least two layers of fibres containing different thermosetting binders are combined and are cured at temperatures exceeding the curing temperatures of the thermosetting binders.
6. Method according to claim 5, characterised in that the layers of fibres are combined such that one or several layers of fibres containing binders based on phenolic resin form the inside layer and that the upper and lower surface of the inside layer are each provided with one outside layer formed by one or several layers of fibres containing binders on the basis of epoxy resin.

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7. Method according to claims 5 and 6, characterised in that the layers of fibres to which the thermosetting binders are added are produced according to the carding method.

8. Method according to claims 5 to 7, characterised in that the fibrous web is produced according to the carding method, wherein the fibre path in longitudinal direction is sanded with different binders over three sections of similar or different widths such that the inside section contains binders based on phenolic resin and the two outside sections binders based on epoxy resin, the web path thus produced is layered according to the cross-layer method to form a new web path comprising an inside layer with phenolic resin and an upper and a lower layer on the outside with epoxy resin, the said web path is heated for a short period of time allowing the binders to melt and fix on the fibre but not to cure, and the web path is then cooled, packed and preformed and cured, if required.

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