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(54) IMPROVEMENTS IN OR RELATING TO THE PRODUCTION OF FLAME-RESISTANT EPOXIDE RESIN-IMPREGNATED SHEET STRUCTURES

(71) We, DYNAMIT NOBEL AKTIENGESELLSCHAFT, a German company, of 521, Troisdorf, Near Cologne, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a process for the production of flame-resistant epoxide resin-impregnated sheet structures.

Laminated structures are known to be built up from resin impregnated glass silk fibres, which may be in the form of woven fabrics or non-woven fabrics such as fleeces or mats and which serve as reinforcements for the resin. Such laminates are produced for example by impregnating glass silk fabric webs with epoxide resins and hardening the resins in a heating press. However, the reinforcements can also be of cellulosic origin, for example in the form of paper, sheets, monofilaments or fibre layers, or formed from synthetic materials such as polyesters and polyamides as synthetic papers, sheets or fibre layers. Laminated structures are generally built up from a plurality of layers placed one above the other, although in some cases only a single such layer may be produced. The structures can be covered on one or both

sides in a laminating procedure or otherwise and by means of a hot adhesive with a metal foil, preferably a copper foil, in order to constitute electrically insulating support materials for conductors, e.g. supports for printed circuits produced from the metal foil by known methods. Printed circuits are used in many instruments used in the electrical and electronic arts, for example in radio and television sets, and computers.

In order that they should be suitable for use in the electrical art, it is of particular importance that support structures produced, for example, as aforesaid should have good electrical insulating capacity, high mechanical strength and ability to be employed in the production of printed circuits. This last requirement is met with epoxide laminates if the material is capable of being cut, drilled and stamped at room temperature or with gentle heating and the support has a good chemical resistivity to solvents such as trichlorethylene and methyl ethyl ketone, and caustic solutions, such as sodium hydroxide solution, which are used in the production of printed circuits.

The following standards generally have to be met if the aforesaid layer structures are to be suitable for use as insulating supports for printed circuits:

- a. insulation resistance after 4 days at 40°C, 92% relative humidity $\geq 10^{10} \Omega$
- b. electrolytic corrosive effect on metals after 4 days at 40°C, 92% relative humidity according to DIN 53 489 AN 1.4
- c. bending strength at 23°C $> 2800 \text{ kp/cm}^2$

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|---|--------------------|
| d. temperature resistivity long term: | 25,000 hours 130°C |
| short term: | 10 min. 260°C |
| e. cutting suitability at 23°C: | free from cracks |
| f. layer-bonding by the ball-pressure test as criterion for stamping capability | reading ≤ 2 |
| g. resistance to trichlorethylene vapour: | at least 5 minutes |
| h. resistance to 3% by weight sodium hydroxide solution at 40°C: | at least 3 minutes |

In addition, the material should have the best possible flame-inhibiting properties for certain uses. This requirement is equally applicable to other uses of single and laminated layer structures as aforesaid produced in the form of decorative sheets for use, for example, in interior decorating.

Flame resistance of epoxide resin-impregnated layer structures can generally be improved by adding flame-inhibiting substances. A flame resistance which conforms to practical requirements is provided if the laminate falls within burning class SE I or SE O with vertical flame effect according to the test UL 492 to be described hereinafter. For example, the burning class SE O or SE I is reached with epoxide resin laminates by adding tetrabromo-bis-phenol A to the epoxide resin impregnating agent used in the production of the layer structure. Usually, in the production of the resin, the bisphenol A normally used is replaced by tetrabromo-bis-phenol A to such an extent that the bromine content of the finished epoxide resins is 18—20%, by weight. However, the resistance to high temperatures of layer structures produced in this way drops considerably.

This resistance to high temperatures is of particular importance with epoxide resin-impregnated layer structures especially when they are used in electrical equipment subject to extreme temperature loadings caused by self-heating of components attached to the laminated material, or by heat emanating from adjacent components.

With epoxide resin layer structures contain tetrabromo-bis-phenol A as flame-protection agent, however, the temperature resistivity of the insulating material in the long-term test at 130° C is considerably reduced. Also particularly noticeable is the lower temperature resistivity of the epoxide resins which contain tetrabromo-bis-phenol A in the short-term heating test of 10 minutes at 260° C which is generally carried out in silicone oil. The stressing which takes place at a relatively early stage in these tests destroys the epoxide resin system and yields a brownish-black layer structure.

The heating test at 260° C in silicone oil

is intended to simulate the conditions when soldering contacts on the layer structure and when removing soldering tin therefrom using hot oil, although it also serves to simulate extreme temperatures during use. The brownish-black discoloration which is due to decomposition of the brominated bis-phenols also results in drastic deteriorations of, for example, the bending strength and the insulation resistance of the layer structure.

According to one aspect of the present invention, there is provided a single, or multi-layer laminated, layer structure which is a heat condensed epoxide resin-impregnated sheet structure of fibrous origin, the heat condensed epoxide resin impregnant comprised by the structure additionally containing pentabromodiphenyl ether as flame inhibiting agent.

According to a second aspect of the invention, there is provided a process for the production of a layer structure suitable for combination with one or more like structures under pressure and heating to produce a laminate, which comprises impregnating a sheet structure of fibrous origin with an impregnating composition comprising an epoxide resin and a flame-protecting agent comprising pentabromophenyl ether, drying and effecting at least partial condensation of the resin of the impregnating composition.

By the use of pentabromodiphenyl ether or a mixture of pentabromodiphenyl ether and other bromination products of diphenyl ether, as for example in the commercially available pentabromodiphenyl ethers, there are obtained single or laminated layer structures which may be shaped so as to be non-planar, the structures having extinguishing properties as well as thermal, electrical, mechanical and processing properties, which are good and may even be better, in some cases than those hitherto obtained.

In particular, it has been found that the addition of pentabromodiphenyl ether (PBD) also improves the bonding of layers of laminated structures and thus the important requirement of suitability for stamping. In addition, there is also obtained an improvement in the bond strength, which, in contrast,

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is lowered when using many other bromine-containing flame-proofing agents in reinforced epoxide layer structures.

However, of particularly great value is the resistance to elevated temperatures which is greatly improved as compared with that of hitherto described flame-proof epoxide resin-impregnated layer structures and which is equally as good as that of epoxide resin impregnated layer structures not provided with flame-proofing agents. For example, layer structures can be produced which have a temperature resistivity of more than 25,000 hours and which are resistant to a temperature of 260° C when immersed for more than 10 minutes in silicone oil.

Of particular advantage is the general characteristic of the layer structures of this invention of being able to withstand being at 200° C for several weeks. The resistance of the layer structures to such thermal stressing has not hitherto been obtained with epoxide resin-impregnated layer structures.

Furthermore the layer structures of this invention fall into classes SE I and SE O of the UL 492 test referred to above. The test for flame resistance is more fully identified as UL Subject 492 paragraph 280. A—K produced by Underwriters' Laboratories, USA, and provides a particularly satisfactory assessment of flame resistance. The test is carried out as follows:

A test specimen 12.7 × 102 mm is clamped with its longitudinal axis vertical and with its front edge disposed 9.5 mm above the tip of a Bunsen burner with a diameter of 9.5 mm. The burner is adjusted until there is a blue flame 19 mm long and is held for a period of 10 seconds centrally beneath the bottom end of the test specimen. After removing the flame, the post-burning period or the smouldering period is measured. After complete extinction of burning the specimen is ignited a second time for 10 seconds. The second burning period or smouldering period is also measured. The assessment of flame-resistant materials subjected to this test is such that they can be considered to fall into one of two combustible classes:

1. SE I (self-extinguishing I)

In this class, the average length of the post-burning period must be equal to or less than 25 seconds. The maximum length of this period must not exceed 30 seconds.

2. SE O (self-extinguishing O)

In this class, the average length of the post-burning period must be equal to or less than 5 seconds and the maximum length of this period must not be greater than 10 seconds.

Materials which fall within either of these two classes and particularly burning class SE O possess good passive protection against burning when used in electrical equipment,

in which there is a good chance of the insulating material being ignited as when a fire is caused by a short circuit.

The burning classes SE I and SE O have hitherto only been obtained with a limited number of resin-impregnated layer structures and, these latter structures do not always meet other requirements thereof owing to their composition and the additives which are used therein. In contrast, the layer structures of this invention do not suffer these disadvantages.

The layer structures of this invention can comprise as reinforcing material, glass, in the form for example of woven fabrics and non-woven structures such as glass fibre fleeces, glass mats or layers of glass fibres. It is also possible to use papers, such as cotton papers, cotton linter papers, layers of sulphated or sulphited cellulose or woven or non-woven structures formed of synthetic fibres, such as polyesters, polyamides and other polymeric organic substances. It is also possible to employ fibrous natural substances, such as asbestos or mineral wool, and such materials as straw, sawdust and fillers, can be used when producing layer structures for use in the building and decorative trades, and for use in vehicles.

The epoxide resin impregnants for the reinforcements are hardenable resins having a viscosity such that they can vary from free flowing liquid to solid. They are the reaction products of aliphatic or aromatic diols with epoxides usually in molar ratios of from 1:1 to 1:2. The diols are exemplified by diphenylol propane (Bisphenol A), diphenylol methane (Bisphenol F), diphenolic acid and phenols which are derived from cashew nut oil, to which may be added aliphatic diols and triols. The epoxides are exemplified by epichlorohydrin. In place of epichlorohydrin, it is also possible to use β -dichlorohydrin which is an open chain compound which reacts with aromatic and aliphatic diols to yield resins containing groupings identical with those obtained when epichlorohydrin is employed. Hence the term epoxide resin as used herein includes such a resin. The epoxide resins are used as such or with addition of non-reactive diluents, preferably in the form of solution of which they constitute from 40 to 80% by weight and more preferably from 50 to 60% by weight. The resins can be hardened with the aid of amines, particularly tertiary amines, for example piperidine, benzyl dimethylamine, *m*-phenylene diamine, diethylaminopropylamine or dimethylaminopropyl amine, tris (dimethylaminoethyl)-phenol, diethylene triamine and dicyandiamide, or anhydrides of polybasic acids, for example phthalic anhydride, pyromellitic anhydride and dodecenyl-succinic anhydride.

It is possible to add PBD to the impregnant, in liquid form by stirring it into the impregnating agent, but preferably a solution of PBD in a common solvent for resin and PBD is used. The concentration of the solution can be chosen within wide limits, solvents with boiling points of from 30 to 120° C. being preferred. Since these solvents will vaporise during the drying of the layer structure, 30 to 80% by weight solutions are preferred. Solvents having some solvent action for PBD which can be used include ethers, alcohols, hydrocarbons and ketones, for example diethylether, methanol, ethanol, the propanols, pentane, hexane, petroleum ethers, benzene and acetone, as well as their mixtures with one another or with water.

Commercially available pentabromodiphenyl ether has a relatively high viscosity at room temperature and even at 50° C., or already contains crystalline fractions and can be contaminated with small quantities of hexabromodiphenyl ether. A crystalline deposit of bromine compounds, more especially hexabromodiphenyl ether and higher products, can be formed when the solutions of PBD are stored at room temperature. The precipitation of crystals can, however, be avoided by using mixtures of the brominated diphenyl ethers, in which are present the bromination products tribromodiphenyl ether to octabromodiphenyl ether. In this way, a problem which might otherwise affect the use as flame-protecting agent of PBD in the production of reinforced shaped laminates can be avoided.

The amounts of the various bromination products of diphenyl ether which are used in the resin impregnant can be varied within

wide limits. For practical purposes, it is generally preferred that the viscosity of the mixture of ethers at 50° C does not exceed 9000 cP and preferably lies in the range of from 300 to 7000 cp, more preferably below 4000 cp. Mixtures of this type which comply with these conditions have the following composition:

Pentabromodiphenyl ether 40—60% by weight
Tetrabromodiphenyl ether 15—45% by weight
Hexabromodiphenyl ether 1—20% by weight
Tribromodiphenyl ether 0—5% by weight
Octabromodiphenyl ether 0—2% by weight

As explained, it is also possible to deviate from this mixture, provided only that a not too high viscosity of the mixture is reached.

In general, the pentabromodiphenyl ether should constitute from 35 to 70% by weight of a mixture of diphenyl ether bromination products used as flame-protective agent. Of the other bromination products of diphenyl ether which can be used with pentabromodiphenyl ether, tri-, tetra- and hexa-bromodiphenyl ethers are preferred.

The bromine content of the mixtures of ethers will in general amount to from 66.0 to 70.5%, more especially from 67.0 to 69.5% by weight.

The results of a solubility test carried out to determine the stability of solutions in acetone for different mixing proportions of brominated diphenyl ethers are set out in Table 1 from which it can be seen that the required good solubility in solvents and the avoidance of crystal formation is obtained when using mixtures of brominated diphenyl ethers.

TABLE 1

Bromine content %	Viscosity at 50°C (cp)	Acetone solution stable for 24 hours	Tri-	Tetra-bromodiphenylether (%)	Penta-	Hexa-	More highly brominated products (%)
67.9	650	yes	1.5	53	42	3.5	0.2
69.0	1900	yes	0.2	43	50	6	0.7
69.0	3960	yes	0.1	35	55	9	0.8
69.2	3650	yes	0.2	30	55	10	1.0

The mixtures of the bromination products can be dissolved without any problems at 60° C in the solvents, as aforesaid, acetone being particularly preferred. Solutions of high concentration, as from 70 to 80% by weight can be achieved. The solutions are stable in storage for more than 24 hours and the production of solutions of relatively high con-

centration simplifies storage problems.

The layer structures of this invention can contain from 35—65% by wt. of the reinforcement and from 65—35% by weight in total of an impregnation of epoxide resin and PBD flame-proofing agent. The layer structures can be used for all known purposes for which such type of structures are

generally employed. They are particularly suitable for use as electrical insulating materials and as support material for printed circuits, formed from metal foils for example, copper foils, applied to one or both sides of laminates comprising the layer structures preferably by pressure with the assistance of a hot adhesive.

Prior to application of the metal foil to one or both sides of the laminates, are as both sides of the laminates can be given a surface layer resistant to leakage current. This layer can consist of a cellulosic paper web, which is impregnated with a melamine resin resistant to moisture or an aliphatic or cycloaliphatic epoxide resin.

Because the layer structures possess high flame resistance and good workability they are also particularly suitable for use in the production of decorative laminates and as facing sheets for use in vehicle construction and interior decorating. In contrast to layer structures of the resin, especially epoxide resin-impregnated support type hitherto used for these purposes, especially impregnated papers containing other flame-resistant additives, the layer structures of this invention are not damaged by water of condensation, possess low moisture absorption and are free from bubble formation. Printed or dyed decorative sheets, films, webs, fabrics, wood veneers or other surface layers can be applied by pressure to one or both sides of the layer structure and these too can be impregnated with the said resins.

The production of the layer structures of this invention is generally effected by impregnating the reinforcing materials with an impregnating solution of the epoxide resin. This can be achieved by spreading, for example with a roller, by spraying or by dipping. In continuous processes, the reinforcement is preferably conveyed through an impregnating bath in a one-step method or also in a multi-step method. Drying and condensation of the synthetic resin is then effected in conventional manner, it is preferable to use a drying tunnel. The final hardening of the thermosetting resins can be effected

in conventional manner in a heating press, using a pressure preferably of from 30 to 150 kp/cm² and a temperature preferably in the range of from 130 to 180° C, more preferably from 160 to 170° C. Usually, several superimposed layers of the impregnated and pre-dried fabric are compressed to form a laminated structure, using a pressing time of 30 to 90 minutes. By suitably designing the heating press it is possible to produce final products which are non-planar.

The following Examples illustrate the invention:

Example 1a.

A glass fibre fabric having a width of 1200 mm, and having a weight per surface area of 200 g. per square metre was continuously supplied from a reel and guided through an impregnating bath having the composition set out in Table 2 and containing 25% by weight of PBD.

The immersion time for the fabric was 30 seconds. The fabric wetted with the impregnating solution was guided over two rollers and freed from excess solution by wringing between two steel rollers. The impregnated fabric was then guided through a drying duct, in which it was heated within 4 minutes from 150° C gradually to 170° C to effect partial condensation of the resin. Rectangular pieces with a length of 1200 mm and a width of 1100 mm were cut from the strip in a cutting apparatus. Eight of these fabric layers were superimposed and heated to 170° C for 60 minutes in a heating press under a pressure of 70 kp/m². In this way, a laminated layer structure with a thickness of 1.6 mm was formed. The proportion by weight of the fabric in the layer structure was 58%.

Example 1b.

The operating procedure of Example 1a was repeated using the same bath to impregnate with the epoxide resin containing PBD instead of glass fibre fabric a cellulose paper having a weight per unit area of 120 g/square metre.

TABLE 2

Impregnant	Parts by wt.
solid resin, glycidyl ether of Bisphenol A epoxide equivalent 475 g/epoxy	68.8
benzyl dimethylamine	0.2
dicyandiamide	4
pentabromodiphenyl ether (commercial grade)	25
antimony trioxide	2
acetone	80

Examples 2a and 2b (comparative).

5 The procedures of Examples 1a and 1b were repeated using an impregnating solution according to Table 3, which did not contain any PBD, but whose epoxide resin was a glycidyl ether of a brominated bisphenol A,

produced from 25% by weight of tetrabromobis-phenol A (TBB) and 75% by weight of bromine-free Bisphenol A. The glass fabric of Example 1a was used in Example 2a and the cellulose paper of Example 1b was used in Example 2b.

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TABLE 3

Impregnant	parts by weight
solid resin, glycidyl ether of brominated Bisphenol A	93.8
benzyl dimethylamine	0.2
dicyandiamide	4.0
antimony trioxide	2.0
acetone	80

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Examples 3 and 4.

The procedure of Example 1a was repeated, but using impregnating baths whose PBD contents were reduced to 20% and 15% by

weight of PBD respectively. The baths used to impregnate the glass fibre fabric thus had the composition set out in Table 4.

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TABLE 4

Impregnant	parts by weight	
	Example 3	Example 4
solid resin, glycidyl ether of Bisphenol A, epoxide equivalent 475 g/epoxy	73.8	78.8
benzyl dimethylamine	0.2	0.2
dicyandiamide	4	4
pentabromodiphenyl ether (commercial grade)	20	15
antimony trioxide	2	2
acetone	80	80

As can be seen from Table 5 which follows, the laminated structures produced in Examples 1a and 1b had much better properties for the uses described herein than the products of Examples 2a and 2b. Furthermore, in Examples 3 and 4 the laminate with 20% of PBD fell within the flame class SE O, while that with the smaller proportion of PBD (15%) only fell within the flame class SE I. The temperature resistivity and also the layer bonding of the products produced using PBD in the impregnant were excellent and generally better than in the cases in which PBD was omitted.

TABLE 5

properties	Example					
	1a (25% PBD)	3 (20% PBD)	4 (15% PBD)	1b (25% PBD)	2a (25% TBB)	2b (25% TBB)
resin support	glass fabric					
temperature resistivity 260°C oil bath, 10 min	not discoloured					
200°C circulating air 3 days 200°C circulating air 10 days flame class UL 492	only superficially discoloured					
1. post-burning time UL 492 (sec.)	SE O	SE O	SE I	SE O	SE O	SE O
2. post-burning time UL 492 (sec.)	1-2	1-5	4-15	1-5	1-3	1-4
Layer-bonding, ball-pressure test*	1-2	1-6	5-20	1-7	1-7	1-8
bonding strength DIN 40802 kp/25mm	note 1	note 1	note 1	—	note 2	—
bending strength DIN 7735 kp/cm ²	5.9	5.8	5.8	4.3	5.4	4.2
„ „ after 3 days at 200°C kp/cm ²	4950	4920	4900	1600	4800	1650
insulating resistance acc. to a. (Ω)	4000	4050	4000	1400	100	90
	>10 ¹³	>10 ¹³	>10 ¹³	>10 ¹²	10 ¹³	>10 ¹²

* A ball of 10 mm diameter is pressed into a laminate with a thickness of 1.5mm. The advance amounts to 3 mm, towards a support with a bore of 20 mm diameter (test specification of Siemens, Siemens-Norm SHM 057032, October 1969).



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WHAT WE CLAIM IS:—

1. A single, or multi-layer laminated, layer structure which is a heat condensed epoxide resin-impregnated sheet structure of fibrous origin, the heat condensed epoxide resin impregnated comprised by the structure additionally containing pentabromodiphenyl ether as flame inhibiting agent.
2. A layer structure as claimed in Claim 1, in which the flame inhibiting agent comprises a mixture of pentabromodiphenyl ether and one or more other bromination products of diphenyl ether.
3. A layer structure as claimed in Claim 2, in which pentabromodiphenyl ether constitutes from 35 to 70% by weight of the mixture of bromination products of diphenyl ether used as flame inhibiting agent.
4. A layer structure as claimed in Claim 2 or 3, in which the other bromination products of diphenyl ether are selected from the tri-, tetra- and hexa- bromodiphenyl ethers.
5. A layer structure as claimed in Claim 3 or 4, in which the mixture of bromination products of diphenyl ether consists of from 40 to 60% by weight pentabromodiphenyl ether, from 15 to 45% by weight of tetrabromodiphenyl ether, from 1 to 20% by weight of hexabromodiphenyl ether, from 0 to 5% by weight of tribromodiphenyl ether and from 0 to 2% by weight of octabromodiphenyl ether.
6. A layer structure as claimed in any one of Claims 2 to 5, in which the bromine content of the mixture of bromination products of diphenyl ether is from 66.0 to 70.5% by weight.
7. A layer structure as claimed in Claim 6, in which the bromine content of the mixture of bromination products of diphenyl ether is from 67.0 to 69.5% by weight.
8. A layer structure as claimed in any one of the preceding claims, which contains from 35 to 65% by weight of the sheet structure of fibrous origin and from 65 to 35% by weight in total of the epoxide resin and pentabromodiphenyl ether based on the total weight of said sheet structure, said resin and the pentabromodiphenyl ether.
9. A layer structure as claimed in any one of the preceding claims, in which the sheet structure is formed from cellulosic or synthetic organic fibres.
10. A layer structure as claimed in Claim 9, in which the sheet structure is a cotton paper, cotton linter paper or sulphated or sulphited cellulose paper.
11. A layer structure as claimed in Claim 9, in which the sheet structure is a woven or non-woven fabric.
12. A layer structure as claimed in Claim 11, in which the sheet structure is formed from polyester or polyamide fibres.
13. A layer structure as claimed in any one of the preceding claims, in which the sheet structure comprises inorganic fibres.
14. A layer structure as claimed in Claim 13, in which the sheet structure comprises glass or asbestos fibres.
15. A layer structure as claimed in Claim 14, in which the sheet structure is a woven or non-woven glass fibre fabric.
16. A single, or multi-layer laminated, layer structure as claimed in Claim 1, substantially as described in any one of the foregoing Examples 1a, 1b, 3 and 4.
17. A process for the production of a layer structure suitable for combination with one or more like structures under pressure and heating to produce a laminate, which comprises impregnating a sheet structure of fibrous origin with an impregnating composition comprising an epoxide resin and a flame-protecting agent comprising pentabromodiphenyl ether, drying and effecting at least partial condensation of the resin of the impregnating composition.
18. A process as claimed in Claim 17, in which the flame inhibiting agent comprises a mixture of pentabromodiphenyl ether and one or more other bromination products of diphenyl ether.
19. A process as claimed in Claim 18, in which the viscosity of the mixture at 50° C does not exceed 9000 cP.
20. A process as claimed in Claim 19, in which the viscosity of the mixture at 50° C lies in the range of from 300 to 7000 cP.
21. A process as claimed in any one of Claims 18 to 20, in which pentabromodiphenyl ether constitutes from 35 to 70% by weight of said mixture.
22. A process as claimed in any one of Claims 18 to 21, in which the other bromination products of diphenyl ether are selected from the tri-, tetra- and hexabromodiphenyl ethers.
23. A process as claimed in Claim 21 or 22, in which the mixture of bromination products of diphenyl ether consists of from 40 to 60% by weight pentabromodiphenyl ether, from 15 to 45% by weight of tetrabromodiphenyl ether, from 1 to 20% by weight of hexabromodiphenyl ether, from 0 to 5% by weight of tribromodiphenyl ether and from 0 to 2% by weight of octabromodiphenyl ether.
24. A process as claimed in any one of Claims 18 to 23, in which the bromine content of the said mixture is from 66.0 to 70.5% by weight.
25. A process as claimed in Claim 24, in which the bromine content of the said mixture is from 67.0 to 69.5% by weight.
26. A process as claimed in any one of Claims 17 to 25, in which the pentabromodiphenyl-ether-containing flame inhibiting agent is incorporated in the impregnating



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composition in solution in a solvent therefor.

27. A process as claimed in Claim 26, in which the solvent has a boiling point of from 30 to 120° C.

28. A process as claimed in Claim 26 or 27, in which a 30 to 80% by weight solution is used.

29. A process as claimed in any one of Claims 26 to 28, in which the solvent is selected from ethers, alcohols, hydrocarbons and ketones.

30. A process as claimed in Claim 29, in which the solvent is selected from diethyl ether, methanol, ethanol, the propanols, pentane, hexane, benzene, petroleum ether fractions, acetone and mixtures of such solvents with one another or with water.

31. A process as claimed in any one of Claims 17 to 30, in which the sheet structure is formed from cellulosic or synthetic organic fibres.

32. A process as claimed in Claim 31, in which the sheet structure is a cotton paper or sulphated or sulphited cellulose paper.

33. A process as claimed in Claim 31, in which the sheet structure is a woven or non-woven fabric.

34. A process as claimed in Claim 31, in which the sheet structure is formed of polyester or polyamide fibres.

35. A process as claimed in any one of Claims 17 to 34 in which the sheet structure comprises inorganic fibres.

36. A process as claimed in Claim 35, in which the sheet structure comprises glass or asbestos fibres.

37. A process as claimed in Claim 36, in which the sheet structure is a woven or non-woven glass fibre fabric.

38. A process as claimed in any one of Claims 17 to 37, in which the epoxide resin constitutes from 40 to 80% by weight of the impregnating composition.

39. A process as claimed in Claim 38, in which the epoxide resin constitutes from 50 to 60% by weight of the impregnating composition.

40. A process as claimed in any one of Claims 17 to 39, in which the impregnating composition contains an amine or polybasic acid anhydride hardener for the epoxide resin.

41. A process as claimed in Claim 40, in which the hardener is benzyldimethylamine, piperidine, *m*-phenylene diamine, diethyl- or dimethyl-aminopropylamine, tris (dimethyl-aminomethyl)-phenol, diethylene triamine, dicyandiamide, phthalic anhydride, pyromellitic anhydride or dodecenyl-succinic anhydride.

42. A process as claimed in any one of Claims 17 to 41, in which condensation of the resin is effected by placing the dried impreg-

nated sheet structure in a heating press operated at a pressure of from 30 to 150 kp/cm² and a temperature of from 130 to 180° C.

43. A process as claimed in Claim 42, in which the heating press is operated at a temperature of from 160 to 170° C.

44. A process as claimed in Claim 17 for the production of a layer structure, substantially as described in any one of the foregoing Examples 1a, 1b, 3 and 4.

45. A layer structure whenever produced by the process claimed in any one of Claims 17 to 44.

46. A laminate comprising a plurality of bonded layer structures as claimed in Claim 45.

47. A method for the production of a laminate as claimed in Claim 46, in which a plurality of the impregnated sheet structures which have been dried are superimposed and heated in a heating press operated at a pressure of from 30 to 150 kp/cm² and a temperature of from 130 to 180° C.

48. A method as claimed in Claim 47, in which the heating press is operated at a temperature of from 160 to 170° C.

49. A method as claimed in Claim 47 or 48, in which the sheet structures are kept in the heating press for from 30 to 90 minutes.

50. A method as claimed in Claim 47 for the production of a laminate substantially as described in any one of the foregoing Examples 1a, 1b, 3 and 4.

51. A laminate whenever produced by the method claimed in any one of Claims 46 to 50.

52. An electrical circuit comprising a laminate as claimed in Claim 46 or 51 as electrical insulation.

53. An electrical circuit as claimed in Claim 52 which comprises a metal foil applied to one or both sides of the laminate and formed into a printed circuit on the laminate as support structure.

54. An electrical insulation material which comprises a laminate as claimed in Claim 46 or 51, to one or both sides of which has been applied a surface layer which is resistant to leakage current.

55. An electrical insulation material as claimed in Claim 54, to one or both sides of which laminate has been applied a cellulose paper sheet which has been impregnated with melamine resin or an aliphatic or cycloaliphatic epoxide resin.

56. A decorative shaped laminate structure which is formed from a laminate as claimed in Claim 46 or 51 by applying a printed or dyed foil by pressure to one or both sides of the laminate.



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