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D E C I S I O N
of 23 May 2000

Case Number: T 0692/97 - 3.3.3

Application Number: 90100919.1

Publication Number: 0379172

IPC: C08L 63/04

Language of the proceedings: EN

Title of invention:
Epoxy resin composition and semiconductor sealing material comprising same

Applicant:
MITSUI CHEMICALS, INC.

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step - non-obvious combination of known features"

Decisions cited:

-

Catchword:

-

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0692/97 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 23 May 2000

Appellant: MITSUI CHEMICALS, INC.
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Decision under appeal: **Decision of the Examining Division of the European Patent Office dated 27 November 1996 and issued in writing on 7 January 1997, refusing European patent application No. 90 100 919.1 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: C. Géradin
Members: B. ter Laan
C. Rennie-Smith

Summary of Facts and Submissions

I. European patent application No. 90 100 919.1, filed on 17 January 1990, claiming priority from three earlier applications in Japan (JP 7684/89 of 18 January 1989, JP 7685/89 of 18 January 1989 and JP 8692/89 of 19 January 1989) and published on 25 July 1990 under No. 0 379 172, was refused by a decision of the Examining Division of the European Patent Office dated 7 January 1997. That decision was based on a set of nine claims filed on 27 November 1996, Claim 1 reading:

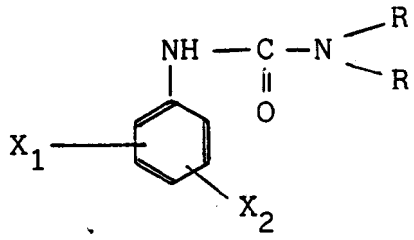
"An epoxy resin composition which comprises, as indispensable components:

(A) an non halogenated epoxy resin

(B) a phenol-novolak resin,

(C) a curing promoter selected from the group consisting of

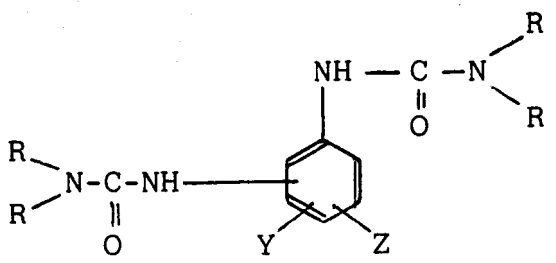
(a) urea derivatives represented by the following formula:



wherein X_1 and X_2, which may be the same or different, represent a hydrogen atom, a halogen atom, a lower alkyl group, a lower alkoxy group or a nitro group, and R's,

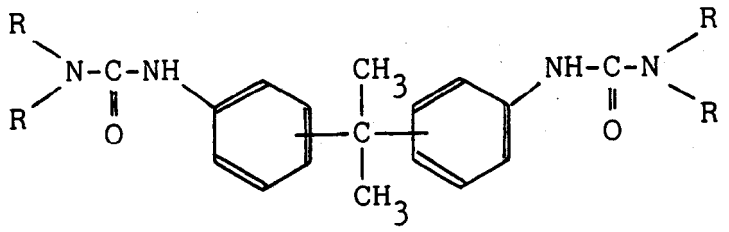
which may be the same or different, represent a lower alkyl group,

(b) urea derivatives represented by the following formula:



wherein Y and Z, which may be the same or different, represent a hydrogen atom, a halogen atom or a lower alkyl group, and R's, which may be the same or different, represent a lower alkyl group,

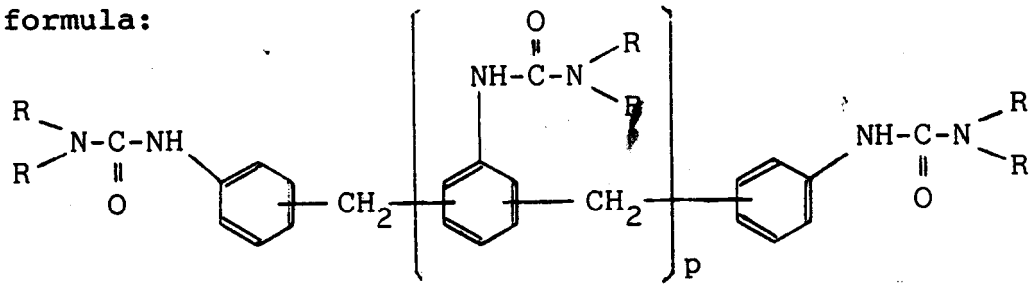
(c) urea derivatives represented by the following formula:



wherein R's, which may be the same or different, represent a lower alkyl group,

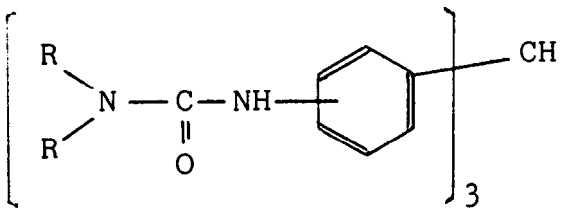
(d) urea derivatives represented by the following

formula:



wherein p is an integer of from 0 to 5, and R's, which may be the same or different, represent a lower alkyl group, and

(e) urea derivatives represented by the following formula:



wherein R's, which may be the same or different, represent a lower alkyl group, and

(D) pulverized silica having a maximum particle size of up to 130 µm and an average particle size of up to 30 µm and/or spherical silica having a maximum particle size of up to 200 µm and an average

particle size of up to 50 µm ;

wherein the phenol-novolak resin (B) is incorporated in an amount of 20 to 120 parts by weight per 100 parts by weight of the epoxy resin (A), the curing promoter (C) is incorporated in an amount of 0.1 to 15 parts per 100 parts by weight of the epoxy resin (A), and the pulverized silica and/or spherical silica (D) is incorporated in an amount of 200 to 620 parts by weight per 100 parts by weight of the sum of the components (A) and (B)."

Dependent Claims 2 to 9 referred to preferred embodiments of the moulding composition according to Claim 1.

II. The Examining Division held that the claimed subject-matter did not satisfy the requirements of Article 56 EPC. It was found that D1 (FR-A-2 061 055) was the closest prior art document. It described similar filled compositions in which the urea promoter comprised most of the urea derivatives of present component (C) and differed only in the amount of silica filler used. Those compositions were suitable for a variety of uses, e.g. moulding or encapsulation. The technical problem to be solved was to provide epoxy resin compositions with improved spiral flow, moulding shrinkage, dimensional stability and surface roughness. From D4 (US-A-4 376 174) it was known that high amounts of filler endowed the compositions with desirable properties, such as shrinkage, and also impaired the flow, thus rendering transfer and injection moulding more difficult. From D2 (US-A-4 701 479) it could be seen that the surface roughness and the flowability could be improved by using a specific mixture of

spherical and pulverized silica filler. Likewise, D3 (Patent Abstracts of Japan, abstract of JP-A-61 283 615) described the use of a silica filler falling within the present specifications, which resulted in excellent crack and moisture resistance. The effects of using silica fillers in encapsulating compositions were also known from D5 (Encyclopedia of Polymer Science and Technology, 1986, vol. 5, pages 800 to 802). Therefore, since the positive influence of silica fillers on epoxy compositions was known from D2 and D3 as well as D5, the subject-matter of Claim 1 was not inventive.

III. On 3 March 1997 a Notice of Appeal was lodged against that decision, together with payment of the prescribed fee. With the Statement of Grounds of Appeal filed on 6 May 1997, the Appellant (Applicant) submitted a set of twelve claims as the sole request, in which the curing promoter had been selected from among the above-mentioned urea derivatives (b).

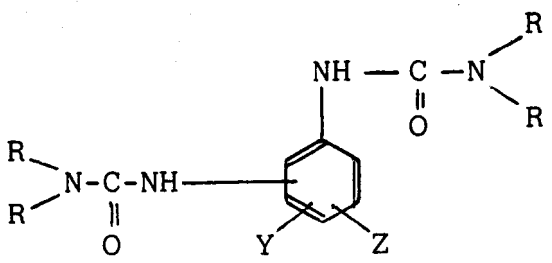
IV. At the oral proceedings before the Board, held on 23 May 2000, in which several objections under Articles 123(2) and 84 EPC were raised, those claims were replaced by a new set of nine claims as the sole request. Claim 1 of that request reads as follows:

"An epoxy resin composition which comprises, as indispensable components:

(A) a novolak-type epoxy resin,

(B) a phenol-novolak resin,

(C) a curing promoter selected from urea derivatives represented by the following formula:



wherein Y and Z, which may be the same or different, represent a hydrogen atom, a halogen atom or a methyl, ethyl, propyl or butyl group, and R's, which may be the same or different, represent a methyl, ethyl, propyl or butyl group, and

(D) spherical silica having a maximum particle size of up to 200 µm and an average particle size of up to 50 µm, or pulverized silica having a maximum particle size of up to 60 µm and an average particle size of up to 8 µm;

wherein the phenol-novolak resin (B) is incorporated in an amount of 20 to 120 parts by weight per 100 parts by weight of the novolak-type epoxy resin (A), the curing promoter (C) is incorporated in an amount of 0.1 to 15 parts per 100 parts by weight of the novolak-type epoxy resin (A), and the spherical silica or pulverized silica (D) is incorporated in an amount of 200 to 620 parts by weight per 100 parts by weight of the sum of the components (A) and (B)."

Dependent Claims 2 to 8 refer to preferred embodiments of the moulding composition according to Claim 1. Claim 9 is an independent claim directed to injection-moulded articles for an electrical or electronic part obtained by injection moulding the epoxy compositions according to Claims 1 to 8.

V. The Appellant's arguments regarding inventive step, submitted in writing and during oral proceedings, can be summarised as follows:

The Examining Division considered D1 to be the closest state of the art. However, D1 neither disclosed the present amounts of filler, nor did it indicate the flowability of the compositions and how it was influenced by the various components of the epoxy composition. Good flowability was essential since the present application was aimed at compositions suitable for injection moulding. Also, in order to be useful in the field of semiconductor applications, the products made from such a composition should fulfil the requirements of good stability, crack resistance, moulding shrinkage, moisture resistance and flame retardancy. Such compositions were known from e.g. D2, which described injection mouldable compositions containing a novolak-type epoxy resin, a phenol-novolak resin and a mixture of two kinds of specific quartz powder particles. Although this composition had good flowability, it could still be improved, in particular at the high filler loads necessary to provide the composition with the desired stability properties. In particular, the gel times at 180°C and 100°C respectively could be ameliorated. The present compositions made it possible to use higher amounts of filler without impairing the injection moulding

capability and also, as illustrated by the examples, due to the choice of a specific urea derivative as a promoter, the gel time had been improved so that the composition was even more suitable for injection moulding than that of D2. Since none of the cited documents disclosed that particular urea derivative or its use as a promoter, the claimed subject-matter was inventive.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 9 as filed during the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

The wording of the claims

2. The amendments to the claims are in conformity with the requirements of Article 123(2) EPC.

2.1 Claim 1 differs from the one as originally filed in

2.1.1 the deletion of "suitable for sealing a semiconductor" in the first line. Since this feature is of a descriptive rather than a limitative nature, it does not change the scope of the claim.

2.1.2 component (A): the support for a novolak-type epoxy resin can be found on original page 7, line 35 and in all the examples.

2.1.3 component (C) is based upon original Claim 10, compound

(b). In the formula of that claim the list of specific alkyl radicals to replace the term "lower alkyl group" in the original definition of X, Y and R is supported by original page 11, lines 16 to 19.

2.1.4 component (D): the limitation of the particle size of the pulverized silica is based on original page 12, lines 12 to 15 and original Claim 9.

2.1.5 The amounts of the various components were originally disclosed in Claim 5 and on page 12, line 30 as filed.

2.1.6 The combination of amendments of the components is supported by the examples; in particular the combination of the compositional features (A) to (D) in the required amounts is to be found in Examples 1, 2, 3, 7-1 and 9-1.

2.2 The wording of Claims 2 to 8 correspond to original Claims 2 and 3, 4, 6 and 7, 9, 11 and 16, with renumbering of the references to other claims.

2.3 Injection moulded articles, now the subject of Claim 9, find their basis on original page 3, lines 3 to 5, page 13, lines 9 to 15 and in the examples.

2.4 The other amendments in the claims are of an editorial nature.

2.5 Therefore, the Board is satisfied that the requirements of Article 123(2) EPC are fulfilled.

3. The present wording of the claims also meets the requirements of Article 84 EPC in that it is clear.

3.1 The replacement of the expression "lower alkyl group" by the specific radicals "methyl, ethyl, propyl and butyl" results in a clear definition of the curing promoter.

3.2 The combination of features (A), (C) and (D) as amended must be regarded as essential for the definition of the invention, because it corresponds to the scope of the claimed subject-matter within which the desired effect has effectively been achieved, as will appear hereinafter.

3.3 The deletion of the unknown term "shirasu balloon" in Claim 8 (original Claim 11) renders the claims compliant with Article 84 EPC.

Novelty

4. The Examining Division did not object to novelty. The Board concurs with that view since none of the cited documents discloses all features in the combination of the present, limited, version of the claims.

The prior art

5. The Examining Division considered D1 as the closest prior art document. However, the Appellant also gave arguments for using D2 as the starting point for assessing the inventive step. Therefore, a preliminary discussion of the documents on file is regarded as appropriate in order to decide upon the closest state of the art.

5.1 D1 discloses curable compositions comprising:

(a) an epoxide resin,

(b) as heat-curing agent, a polyhydric phenol or a compound having at least two primary amino groups attached to a 1,3,5-triazine nucleus and

(c) as accelerator for the curing action, a compound containing, directly attached to a carbon atom in an aromatic nucleus, at least one residue of a specified formula including urea derivatives (Claim 1).

In Claim 4 an accelerator having a structure that encompasses part of the compounds defined by the present component (C) is described. The accelerator may be used in amounts ranging from 0.01 to 10 parts by weight per 100 parts by weight of epoxy resin (a) (Claim 14). The compositions of D1 may also contain fillers (page 10, lines 8 to 16), like quartz flour or colloidal silica having a large specific surface such as that available under the registered trademark "Aerosil" (page 10, lines 11 to 14), but only in very low amounts: in Examples 3 and 5, 5 parts by weight per 100 parts by weight of epoxy resin (a) are added; the compositions described in examples 1, 2, 4 and 6 contain no filler. On page 6, lines 14 to 16, N,N'-bis(dimethylcarbamoyl)-2,4-toluidine and N,N'-bis(dimethylcarbamoyl)-2,6-toluidine, which both fall under the present definition of component (C), are mentioned as preferred accelerators and in the examples 5 and 6 their 80/20 mixture is actually used.

The general teaching of D1 is that the compounds (c), which were known as accelerators for curing epoxy resins with certain cross-linking agents, could also be

used as accelerators for other heat-curing agents (page 1, line 35 to page 2, line 14). The curable compositions of D1 can be used in various applications, such as moulding compositions, sealing compounds and insulating compounds for the electrical industry (page 10, lines 17 to 22). Injection moulding is not mentioned, nor were properties related to injection moulding measured in the examples.

5.2 D2 describes an epoxy resin-based composition for encapsulation of semiconductor devices which comprises:

(a) 100 parts by weight of a phenol-curable epoxy resin; and

(b) from 100 to 500 parts by weight of a combination of silica fillers composed of

(b-1) from 1 to 80% by weight of a quartz powder having a spherical particle form with an average particle diameter in the range from 1 to 25 µm; and

(b-2) from 20 to 99% by weight of a pulverized quartz powder with an average particle diameter in the range from 1 to 25 µm (Claim 1).

As curing agents phenol-novolaks and cresol-novolaks are mentioned (column 3, lines 17 to 22) and a curing accelerator, such as imidazole and derivatives thereof, tertiary amine derivatives, phosphine derivatives and cyclodiamine derivatives, may also be added (column 3, lines 23 to 28).

The general teaching of D2 is that the addition of a

high load of inorganic filler having a specific particle size distribution results in compositions which are not only highly flowable and capable of giving encapsulation of semiconductor devices with little fins as well as small thermal expansion coefficient, but are also highly resistant against crack formation (column 2, lines 37 to 55).

In the Examples 1 to 5 and Comparative Examples 1 to 3 epoxy resin-based compositions were prepared containing 600 parts by weight of a cresol-novolak type epoxy resin, 300 parts by weight of a phenol-novolak resin, 100 parts by weight of a modified epoxy resin, 10 parts of triphenyl phosphine and 2500 parts by weight (2600 parts by weight in Example 5 and Comparative Example 3) of a quartz powder mixture as well as various additives. The compositions of Examples 6 to 9 and Comparative Examples 4 and 5 contained 650 parts by weight of a cresol-novolak type epoxy resin, 300 parts by weight of a phenol novolak resin, 50 parts by weight of a brominated epoxy resin, 10 parts of triphenyl phosphine and 2500 parts by weight of a quartz powder mixture as well as various additives.

The exemplified compositions have a high spiral flow, indicating suitability for injection moulding applications, as well as a favourable thermal expansion coefficient, high resistance against crack formation and good surface properties (Tables), the latter properties being due to the high filler load of specific particles (column 2, lines 51 to 66).

5.3 D3 discloses a composition containing a novolak-type epoxy resin, and a phenolic novolak curing agent as well as a silica filler with reduced particle diameter,

which composition has good resistance to cracking and moisture, and which finds its application in sealing semiconductors. No further details are given.

5.4 D4 discloses a curable epoxy resin composition which comprises:

(a) 100 parts by weight of an epoxy resin,

(b) from 1 to 200 parts by weight of a curing agent,

(c) a specific organosilicon polymer,

(d) a filler and

(e) a curing catalyst (Claim 1).

The amount of filler lies within the range of from 50 to 600 parts by weight of 100 parts by weight of the total amounts of (a) and (b) (column 5, lines 14 to 18). In the five runs of Example 1, 67 parts by weight of novolak-type epoxy resin are mixed with 33 parts by weight of a phenol novolak resin and 220 parts by weight or more of fused quartz powder; as accelerator 2-phenylimidazole is employed (column 7, lines 3 to 13; Table 1).

D4 aims at the development of an epoxy resin composition suitable for use in the resin encapsulation of various electric and electronic parts, which have low stress after curing and with good moisture resistance (column 1, lines 6 to 10 and lines 56 to 64). By using the specific organosilicon polymer (c) this aim is achieved.

5.5 D5 is a general disclosure about the effects of fillers in encapsulating compositions.

6. The application in suit concerns an epoxy resin composition and semiconductor sealing material comprising same.

7. Such epoxy compositions are described in D1, D2, D3 and D4. The Examining Division considered D1 to be the closest prior art document, but that opinion referred to a different set of claims than that now being considered. During the oral proceedings the Appellant argued starting from D2 as the closest state of the art.

7.1 From the above analyses of D1 to D5 it is clear that, from a compositional point of view, D1, D2 and D4 are closer than the other documents: all three documents contain a novolak-type epoxy resin and a phenol novolak resin. Whereas the composition of D1, Example 5, contains a curing accelerator as now claimed, but is lacking in the amount of silica filler, both D2 and D4 describe compositions containing silica filler in the amounts now claimed but they lack the specific curing promoter.

7.1.1 However, in the determination of which document is the closest, the number of common compositional features is not normally decisive. According to the established jurisprudence of the boards of appeal, generally, the claimed invention should be compared with the art concerned with a similar use which requires the minimum of structural and functional modifications. This involves not only comparing the claimed compositions with those of the prior art, but also giving

consideration to the particular properties which render the compositions suitable for the desired use. Therefore, a document serving as the starting point for evaluating the inventive merits of an invention should relate to the same or a similar technical problem or, at least, to the same or a closely related technical field as the application in suit (see decisions T 606/89 of 18 September 1990 and T 795/93 of 29 October 1996; both unpublished in OJ EPO).

7.1.2 According to the description of the application in suit, the selection of the kind and particle size of silica filler, although leading to good crack and moisture resistance, is not sufficient to obtain a composition suitable for injection moulding with very high dimension precision (page 2, lines 17 to 31). Therefore, the problem to be solved as arising from the description is to provide a highly filled epoxy resin composition suitable for precision injection moulding and for semiconductor sealing applications (page 2, line 33 to page 3, line 12). Furthermore, the description refers to highly improved curing stability at 100°C when the 2,4-tolylene diisocyanate/dialkylamine adduct of the formula of page 11 is used as the curing promoter and curing characteristics suitable for injection moulding are attained (page 11, lines 1 to 25).

7.1.3 From points 5.1 and 5.2 above it appears that D2 is the only document which is specifically concerned with injection moulding, although D4, too, describes properties important for that purpose (spiral flow). For that reason, in the Board's opinion, D2 qualifies as a proper starting point for the evaluation of the inventive merits of the claimed subject-matter.

Problem and solution

8. Although, as elucidated above (point 5.2), the compositions of D2 have desirable properties and, in spite of their high load of filler, are even said to be suitable for injection moulding applications, in particular for encapsulation of semiconductor devices, their curing behaviour was still capable of improvement. In particular, the gel time could not be regarded as optimally adapted to the cycle requirements in injection moulding processes.

9. Therefore, the technical problem to be solved by the present application can be defined as to improve the curing behaviour of highly filled injection moulding epoxy resin compositions; more specifically, to arrive at a longer curing time at 100°C and a shorter curing time at 180°C as compared to the known compositions.

10. The examples in the application demonstrate that that problem is effectively solved. In particular, from Examples 1 to 3 it appears that the present compositions, when highly filled, still have a high spiral flow and desirable moulding shrinkage as well as low dimensional change by wet heat treatment, indicating that they are suitable for injection moulding. Examples 7-1 and 9-1, compared with Comparative Examples 7-2 to 7-4 and 9-2 to 9-6, show a significant improvement of the curing behaviour in terms of gel time due to the use of the specific curing promoter (C).

Obviousness

11. It remains to be decided whether the claimed subject-

matter is obvious having regard to the documents on file.

11.1 D2 solves the problem of the suitability for injection moulding by using a specific mixture of spherical and pulverized quartz powder (column 4, lines 4 to 16). However, no mention is made of the curing behaviour, let alone of the importance of selecting an appropriate curing promoter; the compounds mentioned as suitable curing accelerators (column 3, lines 26 to 29) are all conventional catalysts of the reaction between epoxy groups and phenolic groups. Therefore, that document by itself cannot render the present combination of features obvious.

11.2 The same is valid for the other documents on file: none of them refers to curing behaviour and only D1 mentions accelerators within the terms of compound (C) of the application in suit. However, the curing properties achieved with N,N-dialkylmelamine and 5% by weight of silica (Example 5) or with a phenol novolak resin without filler (Example 6) do not demonstrate any advantage resulting from the use of an 80/20 isomer mixture as curing accelerator. From none of the documents could the skilled person derive the positive effect of selecting the present specific curing promoter on the curing behaviour of highly filled epoxy resin compositions.

11.3 For the above reasons, the Board comes to the conclusion that the subject-matter of Claim 1 involves an inventive step.

12. As Claim 1 of the main request is allowable, the same goes for dependent Claims 2 to 8, the patentability of

which is supported by that of Claim 1, as well as for Claim 9 in view of the advantageous properties obtained by using these resin compositions.

13. Although the claims meet the various requirements of the EPC, a patent cannot be granted according to the Appellant's request since the description needs to be adapted to the new wording of the claims. To that end, the case has to be remitted to the Examining Division.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Examining Division with the order to grant a patent on the basis of claims 1 to 9 submitted at the oral proceedings and after any consequential amendment of the description.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin