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D E C I S I O N
of 11 May 1995

Case Number: T 0687/92 - 3.3.3

Application Number: 85115624.0

Publication Number: 0194350

IPC: C08L 101/00

Language of the proceedings: EN

Title of invention:

Fiber reinforced thermoplastics containing silicone
interpenetrating polymer networks

Applicant:

KAWASAKI LNP INC.

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 54

Keyword:

"Novelty (yes) - no implicit disclosure"

Decisions cited:

-

Catchword:

-



Case Number: T 0687/92 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 11 May 1995

Appellant: KAWASAKI LNP INC.
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Representative: Koepsell, Helmut, Dipl.-Ing.
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Decision under appeal: Decision of the Examining Division of the European Patent Office dated 23 September 1991 refusing European patent application No. 85 115 624.0 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: C. R. J. Gérardin
Members: H. H. R. Fessel
J. A. Stephens-Ofner

Summary of Facts and Submissions

I. The appeal lies against the decision of the Examining Division 2.1.15.014 dated 23 September 1991 to refuse European patent application No. 85 115 624.0 filed on 9 December 1985 in the name of LNP Corporation, Malvern, Pa 19355(US), now assigned to Kawasaki LNP Inc., Exton, Pennsylvania 19341(US) (cf. letter received on 3 May 1993). The decision was based on a set of ten claims filed on 31 August 1991 of which the independent Claims 1, 8 and 10 read as follows:

- "1. A melt processable composition comprising:
- a. a thermoplastic matrix material comprising a thermoplastic which has an unfilled flexural modulus greater than 62.05 N/mm^2 (90,000 psi) as measured by ASTM D790;
 - b. 1 to 20 weight percent of the total composition of a polymerizing silicone phase which is made up of a polymeric component containing vinyl groups and of a hydride-containing polymeric silicone which will be vulcanized by the reaction within said thermoplastic matrix in the presence of a catalyst to form a silicone semi-interpenetrating polymer network in which only the silicone phase is cross-linked, the ratio of said hydride groups to said vinyl groups being 1 : 1 to 6 : 1, said vulcanization of said polymerizing silicone phase by reaction of said hydride containing silicone being initiated during thermoplastic melt processing of said phase with said matrix; and
 - c. 5 to 60 weight percent based on the total weight of the composition of a fibrous reinforcing agent mixed with said matrix material and/or said silicone component.

8. Method of reducing mold shrinkage and warpage in a thermoplastic molding resin having an unfilled flexible modulus greater than 62.05 N/mm^2 (90,300 psi) as measured by ASTM D790 and a fibrous reinforcing agent uniformly dispersed in said thermoplastic molding resin, comprising adding to said thermoplastic resin prior to or during thermoplastic melt processing 1 to 20 weight % of the total composition of polymerising silicone phase which is made up of a polymeric component containing vinyl groups and of a hydride-containing polymeric silicone which will be vulcanized by the reaction within said thermoplastic resin in the presence of a catalyst to form a silicone semi-interpenetrating polymer network, the ratio of said hydride groups to said vinyl groups being 1 : 1 to 6 : 1, said vulcanisation of said polymerising silicone phase by reaction of said hydride-containing silicone being initiated during thermoplastic melt processing of said phase with said matrix, the fibrous reinforcing agent mixed with said thermoplastic resin and/or said silicone component amounting to 5 to 60 weight % based on the total weight of the composition.

10. A melt formed composite in the form of a gear having reduced mold shrinkage and warpage, comprising

- a. a thermoplastic matrix comprising a thermoplastic resin having an unfilled flexural modulus greater than 62.05 N/mm^2 (90,000 psi) as measured by ASTM D790;
- b. a silicone semi-interpenetrating polymer network vulcanized within said thermoplastic matrix; and
- c. a fibrous reinforcing agent uniformly dispersed in said thermoplastic matrix."

II. The reason for the decision was that the subject-matter as defined in Claims 1 to 3, 8 and 9 was not novel, and that the subject-matter of Claims 4, 5 and 10 did not involve any inventive step vis-à-vis the teachings of documents:

- (1) R.Keuerleber, "Kunststoffe", vol.73, n^o9, 1983, 509-10,
- (2) Enc.Polym.Sci.Tech., vol.12, 1970, 531-33,
- (3) DE-A-3 314 355, and
- (4) GB-A-2 138 436 (& DE-A-3 314 355).

More specifically, the Examining Division held that there was lack of novelty on the basis of document (1) on the assumptions (i) that the compositions as disclosed e.g. in the fourth example on Table 1, and on page 509 of (1) did not show any interaction between polyamide 66 (component a) and silicon components (component b); (ii) that components b. had necessarily to be used in amounts of 1 to 20 wt.% in order to achieve cross-linking densities of 1 to 2%, and (iii) that silicone IPN's corresponding to mixtures of components of a. and b. were known under the trade name Rimplast^R.

The features of Claims 4, 5 and 10 not disclosed in (1) could not be regarded as inventive since they were known in the field of pseudo-interpenetrating networks, and a skilled person would have regarded it as a normal design procedure to combine these features when seeking to provide further melt-processable compositions from pseudo-interpenetrating networks with good form stability.

Additionally, the Examining Division held that Claim 7 failed to meet the requirements of Article 84 EPC, since the definition of the polymeric component with vinyl

groups leading to the silicone phase by cross-linking reaction encompassed various addition polymers normally used as thermoplastic matrix.

III. A Notice of Appeal was lodged on 16 November 1991 against that decision together with payment of the prescribed fee. A Statement of Grounds of Appeal was received on 24 January 1992. The Appellants disputed the arguments and findings contained in that decision, and contended that three separate passages of (1) had been improperly combined and that in the compositions disclosed in (1) two different phases of the system were "connected" by cross-linking, whereas in the case of semi-IPN's only the silicone component was vulcanised or cross-linked in the thermoplastic matrix, but was not bonded or tied to the polymer matrix, which as such was not cross-linked either. To remove an inconsistency between the actual disclosure of (1) and the Examining Division's analysis of that disclosure, the Examining Division held that the author wrote something he did not mean, and suggested what the author should have written instead. The interpretation of what had been disclosed by (1) as given by the Examining Division was not correct, since a skilled person seeking to reduce shrink and warp in fiber-reinforced molding would not consider (1) simply because it was sufficient to have a look at Table 1 to see that, whatever the teaching given by (1) might be, it was without effect on warpage and shrinkage of fiber-reinforced moldings.

IV. The Appellants requested:

that the decision under appeal be set aside and a patent be granted on the basis of Claims 1 to 10 filed on 31 August 1991 with the description to be adapted;

alternatively to hold oral proceedings.

Reasons for the Decision

1. The appeal is admissible.
2. For the reasons given below, the Board is satisfied that the present wording of the claims meets the requirements of Article 123(2) EPC.

Claim 1 derives from Claim 1 as originally filed by the incorporation of the amounts of the silicone component and the fibrous reinforcing agent, by the presence of a catalyst, and by the definition of the two reactive prepolymers giving rise to the polymerizing silicone phase as well as the relative amounts thereof; these features are disclosed on page 11, lines 5 to 13; page 9, lines 9 to 11; page 12, lines 15 to 18; page 12, lines 6 to 11; and page 13, lines 4 to 9, respectively, of the application as originally filed. The replacement of "pseudo" by "semi" for the characterisation of the interpenetrating polymer networks is admissible since they are synonyms for IPNs having only one cross-linked phase or network which is within a continuous unlinked polymer matrix (see document (4), page 1, lines 29 and 30).

The subject-matter of Claims 2 to 7 corresponds to the originally filed Claims 2, 3, 5, 6, 10 and 11 respectively.

The subject-matter of Claim 10 is based on a combination Claims 12 and 18 as originally filed but does not take into consideration the amendments made in Claim 1 during examination.

Method Claims 8 and 9 were not discussed in the decision under appeal. Claim 8 which is without counterpart in the original application, basically recites all the compositional and quantitative features of Claim 1 and is thus adequately supported for the same reasons as the latter. The Figure "90,300" in brackets is regarded as an obvious error and should read "90,000".

As regards Claim 9, which is a dependent method claim, this corresponds to the "background of the invention" as described in the introduction of the application in suit (page 2, lines 13 to 21), where reference is made to the difficulties in producing high tolerance parts as well as thin walled, variable thickness and thick cross section parts by molding.

3. Subject-matter of Claim 1 of the present patent application is a melt processable composition comprising:
 - a. a thermoplastic matrix material comprising a thermoplastic matrix, such as e.g. a polyamide, which has an unfilled flexural modulus of greater than 62.05 N/mm²;
 - b. 1 to 20 weight% of the total composition of a polymerizing silicone phase made up of
 - b¹. a polymeric component containing vinyl groups and
 - b². a hydride containing siliconewherein the ratio of b² : b¹ is 1:1 to 6:1, leading to a semi-INP when reacted during melt processing in the presence of a catalyst, and
 - c. 5 to 60 weight% based on the total weight of the composition of a fibrous reinforcing agent such as glass fibers.

- 3.1 Column 7 of Table 1 of document (1) discloses a vulcanized product consisting of
- a. polyamide 66
 - b. silicone-IPN and
 - c. 30% of glass fibers.

There is no indication regarding flexural modulus of the unfilled polyamide and the composition of the silicone component and there is no reason to assume that the requirements expressed in Claim 1 concerning these components could be met implicitly. Moreover, in accordance with the general teaching of the citation (cf. introductory summary) the silicone phase is structurally defined as IPN whereas in present Claim 1 this phase is characterized as semi-IPN.

In view of these differences the Board considers the subject-matter of Claim 1 to be novel.

The same conclusion applies to Claim 8, since the subject-matter of that method claim is based on the same combination of features as composition Claim 1.

- 3.2 The arguments provided by the Examining Division to support the objection of lack of novelty vis-à-vis the above composition are not convincing, since they are not only based on different passages of that document not clearly linked to the cited example, but also on different assumptions wrongly said to be evident to a person skilled in the art when reading the above cited document.

~~This applies e.g. to the quantity of the silicone phase combined with a passage referring to "bisher verfügbare Produkte". No convincing argument was provided that the cited example is to be read in the light of that passage and not in the light of the next passage dealing with~~

the "zukünftige Produktpalette" called Rimplast and being partially cross-linked with the matrix "und neben Polyamid auch andere z.T. stärker vernetzende Trägerpolymere enthalten."

Moreover, in view of the interaction between polyamide and silicone component described in the paragraph "IPN auf Basis Polyamid/Silikon" giving rise to "ein ultrahochmolekulares Silikon-Netzwerk mit Quervernetzungen zu Trägerpolymeren" (first paragraph of the right hand column of page 509 in document (1)) as well as the possibility to use other matrix polymers forming stronger cross-links (loc.cit. third paragraph), the final structure must correspond to a system in which both phases are cross-linked and, in addition, there are cross-linkings across the phases. This conclusion is further supported by the statement in the third paragraph that the reacted material can no longer be remelted and reprocessed. This only means that, where the silicone networks are grafted to the carrier resin, they are converted to thermoset resins. This finding is also in line with document (4) which teaches that there are two kinds of INP's, namely (i) the full or true INP's (loc.cit. page 1, lines 18 to 19), which are characterized by "simultaneous coagulation and cross-linking of two latex polymers" (page 1, lines 24 to 25) or "involve a blend of two different prepolymers cross-linked in independent processes and permanently entangled with one another" (page 1, lines 27 to 28), and (ii) semi- or pseudo-INP's, which have only one cross-linked phase on network which is within a continuous unlinked polymer matrix phase" (page 1, lines 29 to 30).

The Examining did not dispute novelty on the basis of documents (3) and (4) concerning semi-INPs, nor does the Board consider (as it well could: see G 10/93 OJ 1995,

page 172) the teaching of these documents to be sufficient to destroy the novelty of the patent application.

4. Turning to the objection under Article 84 EPC concerning the wording of Claim 7, according to which the polymeric component with vinyl groups may not be necessarily different from the thermoplastic matrix material (point 6 of the Reasons), this objection is rejected by the Board for the following reasons:

Firstly, the thermoplastic matrix material should have a flexural modulus greater than 90,000 psi in the unfilled state, which is a high value; by contrast, the polymeric component with vinyl groups used to prepare the polymerizing silicone phase is a prepolymer, thus a compound with a rather low molecular weight as illustrated in the examples, which excludes a high flexural modulus. Secondly, even if the thermoplastic matrix resin may be an addition polymer, i.e. a polymer containing residual double bonds, the amount thereof cannot be equated with the number of vinyl groups in the reactive prepolymer which must be present in significant amounts in order to ensure optimal cross-linking with the silicone hydride groups of the other prepolymer.

5. Although the second ground of refusal of the present application was lack of inventive step of the subject-matter of Claims 4, 5 and 10, a proper examination of that issue concerning the subject-matter of Claim 1 has not yet taken place, and to ensure that it does the case is remitted to the Examining Division for further prosecution.

Since the subject-matter of Claims 1 to 3 and 8 is considered to be novel and the inventive step of the subject-matter of those claims has not yet been discussed the case is remitted to the Examining Division for further prosecution for this additional reason.

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 for further prosecution.

The Registrar:


E. Görgmaier

The Chairman:


C. Gérardin