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D E C I S I O N
of 5 October 1995

Case Number: T 0526/93 - 3.3.3

Application Number: 87311457.3

Publication Number: 0274273

IPC: C08L 67/06

Language of the proceedings: EN

Title of invention:
Carboxyl modified olefinic copolymer composition

Applicant:
UNIROYAL CHEMICAL COMPANY, INC.

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Novelty (yes - after amendments)
"Inventive step"

Decisions cited:
-

Catchword:
-

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Case Number: T 0526/93 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 5 October 1995

Appellant: UNIROYAL CHEMICAL COMPANY, INC.
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Connecticut 06749 (US)

Representative: Spott, Gottfried, Dr.
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Decision under appeal: **Decision of the Examining Division of the European Patent Office dated 25 January 1993 refusing European patent application No. 87 311 457.3 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: P. Kitzmantel
Members: B. ter Laan
W. M. Schar

Summary of Facts and Submissions

I. The appeal, which was filed on 25 March 1993, lies against the decision of the Examining Division dated 25 January 1993 refusing European patent application No. 87 311 457.3 filed on 24 December 1987, claiming a US priority of 5 January 1987, and published under No. 274 273. The appeal fee was paid together with the Notice of Appeal and a Statement of Grounds of Appeal was filed on 26 May 1993.

II. The decision under appeal was based on a set of 26 claims filed on 23 March 1992 (with letter dated 20 March 1992), independent Claims 1 and 17 reading as follows:

"1. A curable moulding composition with reduced shrinkage upon curing characterised in that it comprises:

(a) an unsaturated polyester resin, and

(b) a non-thermoplastic carboxyl modified polyolefin, wherein the amount of polyolefin used ranges from 5 to 100 parts per 100 parts of the polyester resin."

"17. A non-thermoplastic carboxyl modified polyolefin copolymer characterised in that it is obtainable by free radical polymerisation of an admixture of

(a) an ethylene/alphaolefin copolymer wherein the alphaolefin has the formula $H_2C=CHR$, wherein R is an alkyl radical having 1 to 10 carbon atoms and the ethyl/alphaolefin copolymer having a molecular weight ranging from 250 to 15,000, and

(b) an unsaturated carboxylic acid or anhydride containing three or more carbon atoms and at least one carboxyl group."

Claims 2 to 16 and 18 to 26 were dependent, respectively, on Claims 1 and 17.

III. The decision under appeal held that the subject-matter of Claim 1 lacked novelty over the disclosure in document

D1: US-A-4 258 143,

which embraced a moulding composition exhibiting reduced shrinkage upon curing, which composition comprised 90% unsaturated polyester and 10% maleated polyethylene.

IV. Oral proceedings were held on 5 October 1995, in the course of which the Appellant filed an amended set of 20 claims, comprising the following independent Claim 1:

"A curable moulding composition with net expansion upon curing characterised in that it comprises:

(a) an unsaturated polyester resin, and

(b) a non-thermoplastic carboxyl modified polyolefin copolymer of ethylene and an alphaolefin having the formula $H_2C=CHR$, wherein R is an alkyl radical containing from 1 to 10 carbon atoms, which is liquid at ambient conditions, the polyolefin copolymer having a number average molecular weight ranging from 250 to 15,000, wherein the amount of polyolefin used ranges from 5 to 100 parts per 100 parts of the polyester resin."

Independent Claim 12 corresponds to Claim 17 according to the appealed decision but for the insertion of the statement "being liquid at ambient conditions" between the terms "A non-thermoplastic carboxyl modified polyolefin copolymer" and "characterised in that".

Claims 2 to 11 and 13 to 20 of the amended set of claims are dependent, respectively, on Claims 1 and 12.

The Appellant argued that because of their low molecular weight, non-thermoplastic, liquid character the carboxylated polyolefin copolymers according to Claim 12 and their use as "low profile additives" in unsaturated polyester resins according to Claim 1 were novel over the subject-matter disclosed in D1, which used higher molecular weight, crystalline, solid carboxylated polyolefins, preferably in the form of homopolymers. The unexpectable capability of the claimed copolymers to bring about a net expansion of the cured polyesters, not just the reduced shrinkage values evidenced in D1, would demonstrate their non-obviousness.

V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the documents as filed during the oral proceedings (Claims 1 to 20, description pages 1 to 28).

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments (Article 123 (2) EPC)*

Claim 1 combines the features of original Claims 1, 2 (amount of polyolefin copolymer) and 6 (structure and molecular weight of polyolefin copolymer); the newly introduced terms "net expansion upon curing", "unsaturated polyester" and "liquid at ambient conditions" are respectively based on the following statements in the original application: page 5, lines 11 to 13; page 13, lines 16 to 19, page 14, lines 25 to 27, page 15, lines 13 to 14, page 16, lines 7 to 9; and page 5, lines 14 to 15.

Claim 2 is based on page 11, lines 15 to 22 of the original application.

Claim 12 corresponds to original Claim 18 with the further characterisation of the polyolefin copolymer as being "liquid at ambient conditions" (see Claim 1 above).

Claims 3 to 11 correspond to original Claims 9 to 17; Claims 13 to 20 correspond to original Claims 19 to 22, 24 to 26 and 28.

3. *State of the art*

3.1 Document D1 relates to unsaturated polyester compositions comprising 15 to 50% by weight of a carboxylated polyolefin prepared by reaction of a (backbone) polyolefin with an α,ß-ethylenically unsaturated dicarboxylic acid anhydride (see Abstract; Claim 1; column 2, lines 11 to 29). According to column 3, line 67 to column 4, line 6 the backbone polyolefins are homo- or copolymers of C_{3-12} α-olefins, including polyethylenes and highly crystalline polypropylenes. From the melt viscosity range of from

100 to 40,000 cp at 175°C indicated at column 4, lines 62 to 68 one must conclude that the carboxylated homo- and copolymers are solid at ambient conditions. This conclusion is in agreement with the melt viscosities at 175°C of 4180, 2600, 12000 and <1000 cp of the maleated polyethylenes Epolene(R) C-16, Epolene(R) N-14, Epolene(R) C-15 and the maleated polypropylene Epolene(R) E-43 exemplified at column 8, lines 5 to 16 . According to column 2, lines 43 to 49 the polyester compositions have an improved balance of physical properties, including enhanced resistance to water absorption and chemical attack; it is also set out there that "because of low shrinkage during polymerization, greater dimensional stability and a smooth, crack-free surface of molded parts is realized."

In the table at column 9, lines 5 to 14 shrinkage values are indicated: by incorporation of 10% Epolene(R) C-15, E-43 or C-16 according to Examples 9 to 11 the shrinkage of the control polyester composition of Example 8 of 3,50% was reduced to, respectively, 3,43%, 3,24% and 2,88%.

3.2 Document D2 (DE-A-2 305 246) relates to unsaturated polyester compositions having low shrinkage comprising 3 to 40% of a powder of a polyolefin which was graft polymerized with an unsaturated monomer having affinity for the polyester resin (e.g. (meth)acrylic acid) (see Claims 1, 4; page 7 (typed), line 6 from the bottom). According to page 8 (typed) it is essential for the compositions of D2 that the polyolefin graft copolymer forms a dispersion of insoluble particles in the polyester resin.

4. *Novelty*

4.1 With respect to document D1

By the definition of the carboxylated polyolefins as liquid, non-thermoplastic copolymers a clear distinction is established vis-à-vis the solid, crystalline homopolymers which are exemplified in D1. The polyolefin copolymers mentioned in D1 (column 4, lines 1 and 63) as possible alternatives, are only characterized by their melt viscosity, according to which they are solid; no suggestion is contained in D1 that these copolymers may be liquid (at ambient conditions) or non-thermoplastic.

The carboxylated polyolefin copolymers according to present Claim 12 and the polyester compositions according to Claim 1 containing such copolymers are therefore novel over D1.

4.2 With respect to document D2

The disclosure in this document does not explicitly comprise carboxylated ethylene/α-olefin copolymers. Moreover, by their **solid** state the graft-polymerized polyolefin resins used according to this document are clearly different from the **liquid** polyolefin copolymers used according to the application in suit.

The subject-matter of both Claim 1 and Claim 12 is therefore novel over D2.

5. *Inventive step*

5.1 Closest prior art

In view of the close chemical similarity of the modified polyolefins disclosed in D1 and according to the application in suit, for the assessment of inventive step D1 is clearly the most relevant piece of the known prior art.

As set out above, the essential differences between the polyolefin low profile additives used pursuant to D1 and those according to the present application reside in their physical state (solid or liquid at ambient conditions) and in the organisation of their polymer molecules: crystalline with thermoplastic properties; or amorphous with non-thermoplastic (rubber-like) properties.

5.2 Problem and solution

The shrinkage occurring during curing of unsaturated polyester compositions, with the possible consequences of cracking, formation of fibre imprints on the surface and dull appearance have long been known and have led to the development of "low profile additives" able to reduce the shrinkage (see page 1 to page 4, line 1 of the application in suit). A particular class of "low profile additives" are modified polyolefins, and especially the carboxylated polyolefins used according to D1. However, as set out in Section 3.1 above, last sentence, D1 discloses only a maximum shrinkage reduction of from 3,50% to 2,88% (table in column 9, lines 5 to 14).

When starting from this document, the problem underlying the application in suit was thus the provision of modified polyolefin low profile additives which are able to further reduce the shrinkage upon curing of unsaturated polyester compositions.

This problem has been effectively solved by the carboxylated polyolefin copolymers according to present Claim 12, which are liquid, of low molecular weight and non-thermoplastic and which are able to provide net expansion upon curing. This is demonstrated by the results reported in the application in suit: Table II, Examples 2 and 3; Table III, Examples 6 and 7; Table IV, Examples 9 and 10; Table V, Examples 11 and 12.

5.3 Assessment of obviousness

In respect of this issue it is to be decided whether one skilled in the art was led by the prior art to solve the existing problem in the way it has been solved by the present invention.

The only information in D1 concerning the effect of the modified polyolefins on the shrinkage of the thermoset polyester resin is the one in the table at column 9, lines 5 to 14 referred to above. According to this table Epolene(R) C-16, a polyethylene having a melt viscosity of 4180 cp, provides the highest shrinkage reduction (2,88%), followed by Epolene(R) E-43, a polypropylene having a melt viscosity of <1000 cp (3,24%) and Epolene(R) C-15, a polyethylene having a melt viscosity of 12000 cp (3,43%). It appears impossible to draw any reliable conclusion with respect to the influence of the molecular weight on the shrinkage reducing effect of the polyolefin from so few examples. But if a conclusion would be drawn it would have to consider that the best shrinkage reducing effect was not achieved with the

lowest melt viscosity/molecular weight and that D1 does not, therefore, suggest that lower molecular weights favour lower shrinkage. There is also no information in D1 concerning the effect on the shrinkage reduction of the degree of crystallinity/thermoplasticity; indeed, D1 does not even mention non-crystalline/non-thermoplastic polyolefin species.

The disclosure of D1 does, therefore, not suggest to a skilled person the possible use as low profile additives for unsaturated polyester compositions of liquid, non-thermoplastic carboxylated ethylene/α-olefin copolymers; let alone that the use of such copolymers would lead to a considerable reduction of the shrinkage upon curing, up to a net expansion of the thermoset article.

Since D2 relates only to the use as low profile additives of a solid powder from graft-modified polyolefins, one skilled in the art could also not get any hint from this document at the effectiveness for shrinkage reduction of modified liquid low molecular polyolefins.

It follows that both the carboxylated ethylene/α-olefin copolymers according to Claim 12 and the moulding compositions according to Claim 1 have been non-obvious over the cited prior art. Thus, these claims comply with the requirement of Article 56 EPC.

By virtue of their dependency, the same applies to the dependent Claims 2 to 11 and 13 to 20.

6. The description is in agreement with the claims and also otherwise complies with the requirements of the EPC.

The application in suit is thus in order for grant.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to grant a patent in the following version:

   Claims 1 to 20 as filed during the oral proceedings.

   Description pages 1 to 28 as filed during the oral proceedings.

The Registrar: The Chairman:

E. Görgmaier P. Kitzmantel