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Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : A method for improving the adhesion of amine-containing organopolysiloxane composition

Klassifikation / Classification / Classement : C 08 L 83/04

ENTSCHEIDUNG / DECISION

vom / of / du 9 March 1989

Anmelder / Applicant / Demandeur : Wacker Silicones Corporation

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence : Improved adhesion/WACKER SILICONES

EPÜ / EPC / CBE Art. 54 and 56

Schlagwort / Keyword / Mot clé : "Inventive step - improvement in adhesion not foreshadowed by opposed documents"

Leitsatz / Headnote / Sommaire

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Case Number : T 183/87 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 9 March 1989

Appellant : Wacker Silicones Corporation
Adrian, Michigan 49221
USA

Representative : Radke, C. Martin
c/o Wacker-Chemie GmbH
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Decision under appeal : Decision of Examining Division of the European Patent Office dated 13 January 1987 refusing European patent application No. 84 103 456.4 pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : P. Lançon
Members : A.J. Nuss
E. Persson

Summary of Facts and Submissions

I. European patent application No. 84 103 456.4 filed on 29 March 1984 and published with publication No. 0 123 915, claiming priority of a prior US application, i.e. US 481435 of 1 April 1983, was refused by a decision of the Examining Division of the European Patent Office dated 13 January 1987. The decision was based on Claims 1 to 8 filed with a letter dated 22 November 1984.

II. The application was refused on the ground that the invention did not comply with Article 52(1) EPC in lacking inventive step (Article 56 EPC).

In its decision, the Examining Division took the view that it would be obvious for a skilled person seeking an accelerated cure of the room temperature vulcanizable (RTV) organopolysiloxane compositions disclosed in document US-A-3 923 736 (1), to apply temperatures of up to 150°C and carbon dioxide in excess of that present in the atmosphere, in conformity with the teaching of document US-A-4 170 700 (2).

III. On 13 February 1987, a notice of appeal was filed by the Appellant against this decision. The appeal fee was paid on the same day and the Statement of Grounds was filed on 23 April 1987.

IV. In their written submission and at oral proceedings, which took place on 9 March 1989, the Appellant submitted, inter alia, that none of the two citations mentioned in the decision of the Examining Division teaches to heat the coated substrate to at least 50°C in the presence of moisture prior to or while exposing the coated substrate to carbon dioxide in excess of that present in the

atmosphere. Therefore, neither document (1) nor document (2) could provide indications that such measures would yield in an increase in adhesion with regard to the state of the art as disclosed in the table on page 18 of the application as filed.

V. At the end of the hearing, the Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 6 of the main, respectively of the auxiliary request as filed during the oral proceedings, and an amended page 3 of the description as filed during the oral proceedings.

Claim 1 of the main request reads as follows:

1. A method for improving the adhesion of an amine-containing organopolysiloxane composition to a substrate which comprises mixing under anhydrous conditions an organopolysiloxane containing reactive groups with an amine-substituted silicon compound capable of cross-linking with the organopolysiloxane and having an average of at least three Si-bonded NR_2 groups per molecule, where R is selected from the group consisting of hydrogen and a monovalent hydrocarbon radical having from 1 to 18 carbon atoms, coating a substrate with the mixture, then heating the coated substrate to at least about 50°C in the presence of moisture and simultaneously or thereafter exposing the coated substrate to carbon dioxide in excess of that present in the atmosphere.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.

2. Claim 1 of the main request causes no objection on formal grounds, since it is essentially the result of a combination of subject-matter disclosed in original Claims 1, 4, 7 and 8, whereby Claims 1 and 7 were independent claims. Since mixing of the organopolysiloxane with the (amine-substituted) silicon compound always requires anhydrous conditions (see page 10, second last paragraph to page 11, first paragraph of the application as filed), this requirement obviously concerns both alternatives claimed. The same applies to heating the coated substrate to at least about 50°C (see page 12, last paragraph). However, all this is now expressed in one single claim, i.e. Claim 1, and a second independent claim became thus superfluous.

The remaining (dependent) claims, i.e. Claims 2 to 6, correspond to original Claims 2, 3, 5, 6 and 9.

The claims of the main request therefore comply with Article 123(2) EPC.

3.1 Document (2) is to be considered as closest state of the art. It concerns a method for accelerating the surface curing of RTV organopolysiloxane compositions which contain hydroxyl terminated organosiloxanes and/or modified organopolysiloxanes and an amine substituted organosilicon compound and which may be cured at room temperature up to about 150°C in the presence of moisture (see col. 1, lines 9 to 13 and col. 7, lines 15 to 19).

As further stated in document (2), it has been surprisingly found, that surface curing of RTV compositions may be accelerated by exposing them to a gaseous medium containing moisture and carbon dioxide in excess of that present in the atmosphere. This makes it thus possible to use these compositions in assembly plants for preparing formed-in-place gaskets, such plants requiring surface cure times of from 10 to 60 seconds. As

soon as the vulcanizable material has been applied to a part and subjected to carbon dioxide and moisture, the part may be removed from the assembly line and assembled or stacked (see col. 1, lines 35 to 41 and col. 7, lines 20 to 24 and 43 to 49).

In example 1 of this prior document, one mold sample (400 millimeters in thickness) is placed in a chamber where it is cured at room temperature in the presence of atmospheric moisture and carbon dioxide in an amount of about 50% by volume. A skin formed on the surface within about 30 seconds, instead of 8 to 10 minutes after exposure.

In the following examples 2 to 9, curing was carried out at either room temperature or at a temperature of 24 to 25°C.

3.2 However it appears from the introductory part of the present application that when amine-containing organopolysiloxanes are exposed to carbon dioxide and moisture to accelerate surface curing the resultant elastomer does not exhibit good adhesion (see page 2, last line to page 3, line 3).

4. The technical problem vis-à-vis document (2) consisted thus in providing a method for improving the adhesion of RTV compositions to a substrate.

This problem is to be solved by the different measures indicated in Claim 1 of the main request (see point V above). As may be seen from the claim, there are in fact two solutions. The first embraces heating the coated substrate to at least about 50°C in the presence of moisture while simultaneously exposing it to carbon dioxide in excess of that present in the atmosphere, whereas the second solution prescribes that the carbon

dioxide treatment be carried out subsequently as a last step of the treatment.

In view of what is set out in the "Table" on page 18 of the description, the technical problem mentioned above has been credibly solved by both solutions claimed in that manifestly both lead to improved results when compared to those corresponding to document (2), the closest state of the art, i.e. the results indicated in the left column of said table.

5. Document (2) differs from the present application mainly in that it does not explicitly mention any combined treatment, i.e. to always heat the coated substrate in the presence of moisture prior to or while exposing it to carbon dioxide in excess (of that present in the atmosphere). Nor can this specific combination of heat and carbon dioxide treatment be considered as disclosed by implication.

Although document (2) on the one hand mentions heating at temperatures up to about 150°C in the presence of moisture and on the other hand treatment with a gaseous medium containing moisture and carbon dioxide, it is obvious that these items of information were given there without any intention to combine them in the same way as in the present application and, moreover, in a quite different context. Since all nine examples of this prior document mention temperatures not exceeding 25°C, there is no support for the idea of combining working at increased temperatures with carbon dioxide treatment. These two measures concern different aspects of the curing problem: the former relates to the normal curing (or hardening) conditions required for vulcanizable compositions; the latter to what is specifically required for accelerating surface curing, the only improvement actually aimed at in document (2). However, this has hardly anything to do with

the problem of improving the adhesion of RTV compositions to a substrate, the actual problem to be solved in the present case.

In view of this and in the absence of any other document which discloses the spsecial combination characterizing the claimed method, the subject-matter of Claim 1 of the main request is novel.

6. It remains, therefore, to be examined whether the requirement for inventive step is met.

6.1 Apart from the fact that document (2) does not disclose combined heat and carbon dioxide treatment (see point 5 above), the man skilled in the art had no reason to expect improved adhesion from elevated temperatures, since the examples filed together with Applicant's letter dated 23.7.86 clearly show that there exist no relationship between increased adhesion strength and increased curing temperature. This comparison, obtained by moisture curing at 25,50 and 150°C, merely shows that there are cases, where the adhesion is either improved or deteriorated by higher temperatures, but that in most cases no alteration is observed. Moreover, the skilled man had also good reasons to believe that the accelerated surface curing with skin formation practised in document (2) would by no means lead to improved adhesion, since skin formation, which is extremely rapid there (e.g. about 30 seconds in example 1), is known to impede the cure of the material under the skin, which in turn produces poor adhesion of the resultant silicone elastomer to a substrate (see page 2, first paragraph of the description).

Under these circumstances, the teaching of document (2) could hardly be expected to be of any help when trying to improve the adhesion of RTV compositions to a substrate.

6.2 The same also applies to document (1), which differs slightly from document (2) in that curing may well be carried out in the presence of atmospheric moisture at a temperature above room temperature, but without excess of carbon dioxide (see col. 5, line 40 ff.). However, the compositions produced according to the examples of document (1) are all cured at temperatures ranging from -10 to +23°C (see col. 6, line 43; col. 7, lines 3, 9, 19, 29 and 66; col. 8, lines 4, 19 and 26). The man skilled in the art had therefore no reason to envisage curing at temperatures far above room temperature (e.g. 50°C or higher).

Moreover, in this prior document, one of the objects of the invention was to provide RTV compositions having relatively slow skin-over times in view of the drawbacks attached to rapid skin formation. These compositions were said to cure and adhere firmly to a great variety of substrates (see col. 1, lines 51 to 56; col. 1, line 67 to col. 2, line 1; col. 5, line 48 ff). However, from what has been said above only poor substrate adhesion could be expected from an additional carbon dioxide treatment. The man skilled in the art would thus be reluctant to consider carbon dioxide treatment when trying to improve adhesion.

Therefore, neither document (1) nor document (2), alone or in combination, foreshadowed the claimed method.

6.3 Further, the overall result of the claimed method indicates a clear increase in adhesion, which does not seem to have lead to a deterioration of some other relevant properties of the coating. As shown by the results in the "Table" on page 18 of the present application, 24 out of 38 examples listed in the table show an improvement in adhesion when heating (110°C) occurs simultaneously to exposure to carbon dioxide (column in the middle); 38 out of 38 examples show an improvement

when carbon dioxide treatment occurs after heating (110°C) (right column).

6.4 At the oral proceedings the Board expressed some concern about the fact that the comparisons made in the examples were based on experiments carried out at 110°C, a temperature which is far remote from the lowest temperature limit mentioned in the claims (i.e. at least about 50°C). The Appellant explained that this rather high temperature belonged to the most preferred range of temperatures which showed the most unexpected results. However, the advantageous effect of the claimed treatment could already be observed from 50°C and upwards. The Board accepted this explanation in the absence of any evidence to the contrary.

6.5 In these circumstances, the Board takes the view that there are no grounds which prejudice the grant of a patent on the basis of the claims of the main request.

As matters stand, the Appellant's auxiliary request has become purposeless.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The case is referred to the first instance with the order to grant a patent on the basis of Claims 1 to 6 of the main request and the description as amended during the oral proceedings.

The Registrar: The Chairman:

F.Klein P.Lançon

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