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**D E C I S I O N**  
of 26 September 2001

**Case Number:** T 0768/97 - 3.3.7

**Application Number:** 86300424.8

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**IPC:** C09D 183/02

**Language of the proceedings:** EN

**Title of invention:**  
Silica coating

**Patentee:**  
MINNESOTA MINING AND MANUFACTURING COMPANY

**Opponent:**  
Röhm GmbH

**Headword:**  
-

**Relevant legal provisions:**  
EPC Art. 54

**Keyword:**  
"Claims - interpretation"  
"Novelty - yes (after amendment)"

**Decisions cited:**  
-

**Catchword:**  
-



Case Number: T 0768/97 - 3.3.7

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.7  
of 26 September 2001

**Appellant:** MINNESOTA MINING AND MANUFACTURING COMPANY  
(Proprietor of the patent) 3M Center  
P.O. Box 33427  
St. Paul  
Minnesota 55133-3427 (US)

**Representative:** Voortmans, Gilbert J.L.  
3M Europe S.A./N.V.  
OIPC  
Hermeslaan 7  
B-1831 Diegem (BE)

**Respondent:** Röhm GmbH  
(Opponent) Kirschenallee  
D-64293 Darmstadt (DE)

**Representative:** -

**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office posted 16 May 1997  
revoking European patent No. 0 193 269 pursuant  
to Article 102(1) EPC.

**Composition of the Board:**

**Chairman:** R. E. Teschemacher  
**Members:** G. Santavicca  
B. L. ter Laan

## Summary of Facts and Submissions

I. The mention of the grant of European patent 0 193 269, in respect of European patent application 86 300 424.8, filed on 22 January 1986 and claiming the priority of US 694945 of 25 January 1985, was published on 17 February 1993 (Bulletin 93/07). The independent claims as granted read as follows:

"1. A process for forming a coated article comprising coating a substrate with a solution containing 0.2 to 15 weight percent colloidal silica particles having an average primary particle size of less than 20 nm (200A), and drying said coating at a temperature of less than 200°C to form a substrate having a coating of a continuous gelled network of silica particles which is transparent, provides a substantially smooth surface, is substantially uniform in thickness and is substantially permanently adhered to said substrate providing a 180° peelback value of at least 500 g/cm."

"12. A coated article prepared according to the method of any preceding claim."

Dependent claims 2 to 11 were directed to elaborations of the process according to claim 1.

II. Notice of opposition was filed on 16 November 1993 on the grounds of Article 100(a) EPC that the subject-matter claimed in the patent lacked novelty and inventive step. The opposition was supported, *inter alia*, by the following documents:

D1: DE-C-2 949 168

D2: DE-A-2 446 279

D3: DE-A-2 947 823

D4: EP-A-0 051 405

III. By a decision announced at the end of the oral proceedings held on 29 April 1997 before the Opposition Division, issued in writing on 16 May 1997, the patent was revoked for lack of novelty. The decision was based on a main request as well as on first and second auxiliary requests.

IV. On 14 July 1997 the proprietor (appellant) lodged an appeal against the decision and paid the prescribed fee on the same day. With the statement of grounds of appeal, faxed on 16 September 1997, the appellant filed a further document (D5: US-A-4 478 873) as well as a reformulated main request and four auxiliary requests.

In reply to a communication of the Board, sent with the summons for oral proceedings, the appellant filed a new main request and six auxiliary requests, which, during the oral proceedings held before the Board on 26 September 2001, were replaced by a new, sole request consisting of 10 claims.

Independent method claim 1 reads as follows:

"1. A process for forming a coated article comprising coating a polymeric substrate with a solution containing 0.2 to 15 weight percent colloidal silica particles having an average primary particle size of less than 20 nm (200A), and drying said coating at a temperature of less than 200°C to form a polymeric substrate having a coating of a continuous gelled network of silica particles which is transparent,

provides a substantially smooth surface, is substantially uniform in thickness and is substantially permanently adhered to said polymeric substrate providing a 180° peelback value of at least 500 g/cm."

Dependent claims 2 to 10 are directed to elaborations of the process according to claim 1.

V. In support of the claimed subject-matter, the appellant argued essentially as follows:

(a) As regards the wording of claim 1, it comprised a number of process steps, namely:

(i) providing a solution of colloidal silica particles with specific concentration and particle size ranges;

(ii) coating a polymeric substrate with the solution; and,

(iii) drying said coating at less than 200°C.

Since these process steps alone might lead to various products, which did not necessarily have the desired properties, claim 1 set further limitations on the coating to be obtained, in particular that a continuous gelled network of silica particles had to be formed and that the coating had to be substantially adhered to the substrate to provide a 180° peelback value of at least 500 g/cm.

(b1) According to the description of the patent in suit, the term "continuous gelled network of silica particles" referred to a continuous covering of the substrate with a porous three-dimensional network of silica particles. The

particles of silica were linked together to form the network and the porosity resulted from the presence of voids between them.

(b2) The coating methods disclosed in D1 to D4 did not result in a coating with a continuous gelled network of silica particles according to the definition given in the patent in dispute. In fact, in the coating compositions of those documents large amounts of binder were present which reacted with the silica particles and with itself and so filled up the voids. Thus, a silicone resin matrix with particles of silica dispersed therein was obtained, instead of a porous gelled network of silica particles as required by the patent in suit, which resulted in different adhesion and optical properties.

(c) In addition, the strength of adhesion of the coating to the substrate, as defined by the peelback value in claim 1 in suit, set a limitation on the thickness of the coating, which was typically less than those of the coatings disclosed in D1 to D4, and influenced the adhesion properties of the coating.

Thus, the claimed subject-matter was novel.

(d) The request for reimbursement of the appeal fee pursuant to Rule 67 EPC, which had been initially filed during the written proceedings, was, in view of the new set of claims, not maintained during the oral proceedings before the Board.

VI. The arguments of the opponent (respondent) can be summarised as follows:

(a) The process steps defined in claim 1 were all commonly known. The other features of claim 1 were trivial and nothing more than commonly desired conditions for the utility of the coating.

In particular, D1 to D4 all disclosed coatings which could be applied to polymeric substrates. For instance, D1 disclosed the claimed coating composition, the coating step as well as the drying temperature. The good adhesion of the coating was a consequence of the process steps.

(b1) It was well known from general silica chemistry that a continuous gelled network of silica particles inevitably resulted from gelation and drying of a dispersion of colloidal silica particles.

In this respect, attention was drawn to a number of documents which had not been mentioned before, namely:

- (i) Ullmanns Enzyklopädie der Technischen Chemie, 4. neu bearbeitete und erweiterte Auflage, 1982, Volume 21, pp 456-459;
- (ii) Kirk-Othmer Encyclopaedia of Chemical Technology, third Edition, 1982, Volume 20, page 774 "Silica Gel"; and,
- (iii) Document D6 (US-A-2 432 484).

From these documents, it was apparent that dried layers obtained from colloidal silica particles were porous and the porosity also influenced the optical properties of the coating.

(b2) The addition of polymeric binders to the coating composition was known from any of D1 to D4, and from D6 as well. In D2 also very small quantities of binder were used, hence open pores were present. Therefore, the argument of the appellant that a continuous gelled network of silica particles was not formed in the coatings obtained by the processes according to D1 to D4 was not proven.

(c) The upper limit for the coating's thickness as defined in the patent was disclosed in any of D1 to D4 and thus was not novel. The upper limit, as taught in D2, could even be further reduced by appropriate coating methods. Hence, the good adhesion property as claimed was fulfilled also in the coatings of D1 to D4.

Therefore, the claimed subject-matter was not novel.

VII. The appellant (patentee) requested that the decision under appeal be set aside and that the patent be maintained on the basis of claims 1 to 10 as submitted during the oral proceedings; in addition, he also requested that the case be remitted to the first instance for the examination of inventive step.

The respondent (opponent) requested that the patent be revoked; auxiliarily, he also requested that the case be remitted to the first instance for the examination of inventive step.



## Reasons for the Decision

1. The appeal is admissible.

2. *Amendments*

2.1 Claim 1 results from the combination of claim 1 as granted with the additional features of claim 11 as granted, ie the substrate to be coated is now required to be polymeric. Independent product claim 12 as granted has been deleted. Therefore, the amendments fulfil the requirements of Article 123(2) EPC.

2.2 Since the introduction of the term "polymeric" results in a restriction over claim 1 as granted, and since independent product claim 12 as granted has been deleted, the requirements of Article 123(3) EPC are also fulfilled.

2.3 The introduction of the further limitation "polymeric" into claim 1 as granted does not introduce any unclarity. Hence, the requirement of clarity (Article 84 EPC) is also met.

3. *The wording of the claims*

The meaning of the expression "gelled network" contained in claim 1 was, in view of the prior art, the subject of discussion. This term will therefore have to be interpreted, based on the contents of the patent specification (Case Law of the Boards of Appeal of the EPO, 3rd edition, 1998, II.B.4)

In the first paragraph of the "Detailed Description of the Invention", in the patent in suit, some terms which are used throughout the specification are explained, ie construed in their intended meaning. The term "gelled

network" refers to "an aggregation of colloidal particles linked together to form a porous three-dimensional network". The term "porous" refers to "the presence of voids between the silica particles" (page 3, lines 31 to 33).

The presence of voids between the silica particles, which are linked together, influences the porosity, the adhesion and the optical properties of the coating (see page 3, lines 6 to 7 and 50 to 56).

If the open porosity is too small, the properties of the coating such as adhesion and antireflectance may be reduced. If the open porosity is too large, the coating is weakened and may have reduced adhesion to the substrate (page 4, lines 1 to 5).

Hence, the porosity is related to adhesion and desired optical properties by the kind of solution of colloidal silica used, ie by the concentration of the silica particles in the starting solution (page 4, lines 57 to 58; page 5, lines 1 to 5) as well as by the size of the silica particles used therein (page 4, lines 25 to 29).

Claim 1 specifically defines the concentration of the silica particles in the starting coating composition, the relevant particle size and the amount of adhesion required by the obtained coating, as measured by the 180° peel back value.

Given that the description indicates that the term "gelled network of silica particles" is to be construed as a porous network of silica particles with voids between them, that all embodiments consistently concern gelled networks of silica which are porous and that the claims define the steps which result in a porous gelled network of silica particles, it can be accepted that the "gelled network of silica particles" defined in

claim 1 is in fact porous, ie the silica particles are linked together and there are voids between them. The presence of these voids is essential in conferring the desired adhesion and optical properties.

4. Novelty

4.1 D2, on which the novelty discussion focused, concerns a pigment-free, aqueous coating composition characterized by a dispersion of colloidal silica in a lower aliphatic alcohol-water solution of the partial condensate of a silanol of the formula  $\text{RSi}(\text{OH})_3$ , whereby R is selected from the group consisting of alkyl radicals of 1 to 3 inclusive carbon atoms, the vinyl radical, the 3,3,3-trifluoropropyl radical, the gamma-glycidoxypropyl radical and the gamma-methacryloxypropyl radical, at least 70% of the silanol being  $\text{CH}_3\text{Si}(\text{OH})_3$ , said composition containing 10 to 50 weight percent solids consisting essentially of 10 to 70 weight percent colloidal silica and 30 to 90 weight percent of the partial condensate, said composition containing sufficient acid to provide a pH in the range of 3.0 to 6.0 (claim 1).

4.1.1 The silica component of the composition is present as colloidal silica. The colloidal silica particles in the aqueous dispersions generally have a particle size in the range of 5 to 150 nm in diameter and are prepared by methods well known in the art and are commercially available. It is preferred to use colloidal silica of 10 to 30 nm particle size in order to obtain dispersions having a greater stability and to provide coatings having superior optical properties (paragraph bridging pages 3 and 4).

The coating compositions are prepared by adding trialkoxysilanes such as  $\text{RSi(OH)}_3$  to colloidal silica hydrosols and adjusting the pH to the desired level by addition of an organic acid (paragraph bridging pages 5 and 6).

The coating composition can be applied to solid substrates by conventional methods, such as flowing, spraying, dipping, to form a continuous surface film. Many kinds of substrates can be coated, in particular polymeric substrates (paragraph bridging pages 7 and 8).

A hard, solvent-resistant surface coating is obtained by removal of the solvent and volatile material. The composition will air dry to a tack-free condition, but heating in the range of 50 to 150°C is necessary to obtain condensation of residual silanol in the partial condensate. This final cure results in an improvement of the abrasion resistance of the coating (page 9, paragraph 1).

Coating thicknesses range from 0.5 to 20 micrometres, preferably from 2 to 10 micrometres (page 9, first paragraph, last sentence). Especially thin coatings are said to be obtainable by spin coating (page 9, first paragraph) but have not been exemplified. Hence, the coatings in D2 are typically thicker than those exemplified in the patent in suit (20 to 500 nm; page 5, lines 7 to 8).

4.1.2 In D2, however, there is no disclosure of a gelled network of silica particles in the coating nor a quantitative indication of the coating properties such as adhesion (page 10, lines 18 to 22), in particular there is no disclosure of the peel back value required by present claim 1.

4.1.3 Regarding Example 4 of D2, the following is taken into consideration:

(i) The coatings of samples 1 and 2 contain 10 and 20 weight percent condensate and 90 and 80 weight percent silica, respectively, which amounts to 18 and 16 weight percent of silica particles in the coating composition, respectively, since the coating composition contains 20 weight percent of solids. Thus these samples are outside the scope of claim 1 in suit. Moreover, since D2 states that the coatings flake off, they apparently do not have the desired adhesion, so that these samples do not fulfil the peel back value required by claim 1.

(ii) Samples 4 and 5 contain more binder than the maximum value given in the patent specification in suit and do not indicate the presence of a gelled network of silica particles with voids, nor any peel back value as defined in present claim 1.

(iii) The coating of sample 3, which was extensively discussed, contains 30 weight percent condensate and 70 weight percent silica particles. Since the starting composition contains 20 weight percent of solids, this amounts to 14 weight percent of silica particles in the starting composition. Hence, the sample contains an amount of silica particle falling within the range given in present claim 1. As regards the binder, the sample contains  $30/70 = 43$  weight percent of binder based on the weight of the silica particles, ie within the range given in the patent specification (see page 5, line 17). For

this sample, however, D2 does not reveal any peel back value, nor has any proof been presented that the coating fulfils the value required by present claim 1.

According to the patent in suit (examples) the adhesion as defined in present claim 1 is such that the tape does not remove the coating from the coated substrate. Instead the tape exhibits adhesive splits of the adhesive layer leaving adhesive residue on the coating. However, in the cross-hatched adhesion tests in D2, a "good to excellent" adhesion implies that some cross-hatched item of the coating is still removed (page 10, lines 17 to 22).

Nor does D2 disclose that the coating of sample 3 is porous by having a gelled network of silica particles with voids.

The respondent, in support of the argument concerning the presence of voids between the silica particles of sample 3, referred to Ullman's and Kirk-Othmer encyclopaedias as well as to D6, to show that a porous gelled network inevitably results from gelation and drying of a dispersion of colloidal silica particles.

However, since the cited passages in the encyclopaedias all refer to the production of porous silica gels from dispersions of colloidal silica particles without any binder present, nothing can be derived about the presence of voids in the coatings of D2 where at least 33 and 43 weight percent of binder are used, based on the weight of the silica particles. D6 does not disclose the use of silicone resin as binder and, therefore, cannot give any information regarding the structure of the coatings of D2.

In view of the disclosure in the patent specification, that the maximum amount of binder should be limited to about up to 50 weight percent based on the weight of the silica particles, but useful amounts are in the range of 0.5 to 10 weight percent (page 5, lines 14 to 18), it appears plausible that in the coatings of D2, for which the ratio condensate/silica should be 25/75 to 30/70 at least, ie respectively 33 and 43 weight percent based on the weight of silica particles, the binder will fill up all the voids between the silica particles, so that no such porosity will be present.

The binder in Example 4 of D2, sample 3, amounts to 43 weight percent based on the weight of silica particles (30 weight percent condensate and 70 weight percent silica), which is close to the upper limit mentioned in the patent in dispute.

The formation of voids may also depend on other conditions such as the kind of binder. In this connection, the patent in suit does not mention that silicone binders are suitable (page 5, lines 14 to 16).

For these reasons, the argument of a lack of voids in the coating obtained in sample 3 of Example 4 in D2 can be accepted.

4.1.4 In summary, no substantive evidence has been produced before the Board to show that a coating in D2 has a continuous **gelled network** of silica particles, with a peel back value as defined in claim 1 in suit.

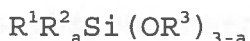
Therefore, the subject-matter of claim 1 in dispute is novel over the coatings disclosed in D2.

- 4.2 D1 discloses the use, for coating solid supports, of a pigment-free composition of a dispersion of colloidal silica and colloidal titania, the colloidal particles having a size in the range of 5 to 150 nm, in a water-alcohol solution of the partial condensate of a silanol of formula  $\text{RSi}(\text{OH})_3$ , in which R is an alkyl of 1 to 3 inclusive carbon atoms, gammachloropropyl, vinyl, 3,3,3-trifluoropropyl, gammaglycidoxypropyl, gammamethacryloxypropyl, gammamercaptopropyl or phenyl, at least 70% of the silanol being  $\text{CH}_3\text{Si}(\text{OH})_3$ , said composition containing 5 to 50 weight percent solids, said solids consisting essentially of 1 to 10 weight percent of colloidal titania, 20 to 56 weight percent of colloidal silica and 34 to 79 weight percent of the partial condensate, said composition containing sufficient acid to provide a pH in the range of 2 to 6 (claim 1).
- 4.3 D3 discloses an aqueous coating composition characterized by a dispersion of colloidal silica in a solution of a lower aliphatic alcohol-water of the partial condensate of a silanol of the formula  $\text{RSi}(\text{OH})_3$ , whereby R is selected from the group consisting of alkyl radicals of 1 to 3 carbon atoms and aryl radicals, at least 70% of the silanol being  $\text{CH}_3\text{Si}(\text{OH})_3$ , said composition containing 10 to 50 weight percent solids consisting essentially of 10 to 70 weight percent colloidal silica and 30 to 90 weight percent of the partial condensate, said composition containing sufficient acid to provide a pH in the range of 7.1 to 7.8 (claim 1).
- 4.4 D4 concerns an antifogging coating film comprising a cured film derived from the following components A, B and C and coated on a substrate, the silicon content thereof being 0.7-2.0/1.0 in terms of carbon/silicon weight ratio, and the carbon/silicon weight ratio of



the surface layer of the film being larger than that of the entire coating film and being not less than 1.7/1.0, (components A, B and C being present as follows):

- A: 100 parts by weight of PVA;
- B: 60-300 parts by weight of finely divided silica having an average particle size of about 5-200 nm;
- C: 0.5-30 parts by weight of at least one compound selected from the group consisting of di- or trifunctional silicon compounds having a general formula:



wherein R<sup>1</sup> is an organic group having 1 to 10 carbon atoms, R<sup>2</sup> is a hydrocarbon group having 1 to 6 carbon atoms, R<sup>3</sup> is an alkyl, alkoxyalkyl or acyl group and a=0 or 1, and the hydrolysates thereof.

The thickness of the film in the surface layer lies within 1 to 500 nm, but the thickness of the entire coating is desirably 0.1 μm or more and up to 30 μm (page 8, lines 19 to 20 and 31; page 9, lines 1 to 5).

The thicknesses of the entire films described in Examples 1 to 4 are between 3.9 and 4.5 micrometres.

The carbon/silicon weight ratio in the entire films described in the examples is always above 1 (Tables 1 and 2), therefore there is a substantial presence of binders, eg PVA and silicone.

4.5 D1, D3 and D4 all disclose a matrix of a resin having dispersed therein particles of silica. The amount of binder actually used in the examples exceeds the limits indicated in the patent in suit. Thus, the formation of a porous gelled network of silica particles with voids is not plausible on the basis of the written information contained in these documents. Since no material evidence concerning this point has been provided by the opponent, the Board accepts that in none of D1, D3 and D4 a porous gelled network of silica particles with voids is formed.

Also the argument that the adhesion in D1, D3 and D4 was excellent and thus corresponded to a peel back value as claimed was not convincingly proven.

In view of these considerations, it cannot be concluded that all the features in the combination as defined by present claim 1 are directly and unambiguously derivable from any of D1, D3 and D4.

4.6 The other documents, cited during the appeal proceedings in relation to claims which are no longer on file, were not used to argue lack of novelty as regards the subject-matter now being claimed. In the Board's view they do not prejudice the novelty of the claimed-subject matter.

4.7 For the above reasons, the Board reaches the conclusion that the subject-matter of present claim 1 is novel over the cited prior art.

4.8 This conclusion also applies to dependent claims 2 to 10, which contain all of the features of claim 1.

5. *Remittal to the first instance*

Following the decision on novelty, both parties requested that the file be remitted to the first instance for further prosecution. Given that novelty alone was decided by the first instance, the Board wishes to remit the case to the first instance for further prosecution and decides accordingly.

6. *Late filed documents*

During the oral proceedings, late filed documents D5 to D6 were no longer used by the opponent to attack novelty. Moreover, as discussed above (see point 4.6 *supra*), they do not prejudice the novelty of the claimed subject-matter. Hence, their admission into the proceedings pursuant to Article 114 EPC has not formed part of the Board's decision.

**Order**

**For these reasons it is decided that:**

1. The decision under appeal is set aside.
2. The case is remitted to the department of first instance for further prosecution on the basis of the claims as submitted during the oral proceedings.

The Registrar:

The Chairman:

C. Eickhoff



R. E. Teschemacher

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