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**Datasheet for the decision  
of 10 May 2006**

**Case Number:** T 0393/04 - 3.3.05

**Application Number:** 91909506.7

**Publication Number:** 0534997

**IPC:** C01B 33/193

**Language of the proceedings:** EN

**Title of invention:**  
Reinforced precipitated silica

**Patentee:**  
PPG Industries Ohio, Inc.

**Opponents:**  
Degussa AG  
RHODIA CHIMIE

**Headword:**  
Silica/PPG

**Relevant legal provisions:**  
EPC Art. 123(2), 54, 56

**Keyword:**  
"Novelty: yes - after amendment (3rd auxiliary request)"  
"Inventive step (yes)"

**Decisions cited:**

-

**Catchword:**

-



Case Number: T 0393/04 - 3.3.05

**D E C I S I O N**  
**of the Technical Board of Appeal 3.3.05**  
**of 10 May 2006**

**Respondents:**

(Opponent 01)

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-

(Opponent 02)

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**Appellant:**

(Patent Proprietor)

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**Representative:**

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**Decision under appeal:**

**Interlocutory decision of the Opposition  
Division of the European Patent Office posted  
12 January 2004 concerning maintenance of the  
European patent No. 0534997 in amended form.**

**Composition of the Board:**

**Chairman:** M. Eberhard

**Members:** B. Czech

S. Hoffmann

## Summary of Facts and Submissions

I. The appeal is from the decision of the opposition division concerning the maintenance of European patent No. 0 534 997 in amended form according to the 2nd auxiliary request filed during the oral proceedings on 27 November 2003 and containing only process claims.

II. Process claims 1 to 20 according to the said request are identical with claims 1 to 20 as granted. Independent claim 1 as granted reads as follows:

*"1. A process for producing reinforced amorphous precipitated silica having, on a coating-free and impregnant-free basis, a surface area of from 220 to 340 square meters per gram, a pore diameter at the maximum of the volume pore size distribution function of from 9 to 20 nanometers, and a total intruded volume of from 2.6 to 4.4 cubic centimeters per gram, said process comprising:*

*(a) establishing an initial aqueous alkali metal silicate solution comprising from 0.5 to 4 weight percent  $\text{SiO}_2$  and having an  $\text{SiO}_2:\text{M}_2\text{O}$  molar ratio of from 1.6 to 3.9;*

*(b) over a period of at least 20 minutes and with agitation, adding acid to said initial aqueous alkali metal silicate solution at a temperature below  $50^\circ\text{C}$  to neutralize from 60 to 100 percent of the  $\text{M}_2\text{O}$  present in said initial aqueous alkali metal solution and to form a first reaction mixture;*

*(c) over a period of from 115 to 240 minutes, with agitation, and at a temperature of from  $80^\circ\text{C}$  to  $95^\circ\text{C}$ , substantially simultaneously adding to said first reaction mixture:*

- (1) additive aqueous alkali metal silicate solution, and
- (2) acid,

thereby to form a second reaction mixture wherein the amount of said additive aqueous alkali metal silicate solution added is such that the amount of  $\text{SiO}_2$  added is from 0.5 to 2 times the amount of  $\text{SiO}_2$  present in said initial aqueous alkali metal silicate solution established in step (a) and wherein the amount of said acid added is such that from 60 to 100 percent of the  $\text{M}_2\text{O}$  contained in said additive aqueous alkali metal silicate solution added during the simultaneous addition is neutralized;

(d) adding acid to said second reaction mixture with agitation at a temperature of from  $80^\circ\text{C}$  to  $95^\circ\text{C}$  to form a third reaction mixture having a pH below 7;

(e) aging said third reaction mixture with agitation at a pH below 7 and at a temperature of from  $80^\circ\text{C}$  to  $95^\circ\text{C}$  for a period of from 1 to 120 minutes;

(f) With agitation and at a temperature of from  $80^\circ\text{C}$  to  $95^\circ\text{C}$ , adding to said aged third reaction mixture additive aqueous alkali metal silicate solution to form a fourth reaction mixture having a pH of from 7.5 to 9;

(g) forming a fifth reaction mixture by adding to said fourth reaction mixture with agitation and at a temperature of from  $80^\circ\text{C}$  to  $95^\circ\text{C}$ , a further quantity of additive aqueous alkali metal silicate solution and adding acid as necessary to maintain the pH at from 7.5 to 9 during the addition of said further quantity of said additive aqueous alkali metal silicate solution, wherein:

- (1) the amount of said additive aqueous alkali metal silicate solution added in steps (f) and (g) is such that the amount of  $\text{SiO}_2$  added in steps (f)

and (g) is from 0.05 to 0.75 times the amount of  $\text{SiO}_2$  present in said third reaction mixture, and

(2) said additive aqueous alkali metal silicate solution is added in steps (f) and (g) over a collective period of at least 40 minutes;

(h) aging said fifth reaction mixture with agitation at a temperature of from  $80^\circ\text{C}$  to  $95^\circ\text{C}$  for a period of from 5 to 60 minutes;

(i) adding acid to said aged fifth reaction mixture with agitation at a temperature of from  $80^\circ\text{C}$  to  $95^\circ\text{C}$  to form a sixth reaction mixture having a pH below 7;

(j) aging said sixth reaction mixture with agitation at a pH below 7 and at a temperature of from  $80^\circ\text{C}$  to  $95^\circ\text{C}$  for a period of at least 1 minute;

(k) separating reinforced precipitated silica from most of the liquid of said aged sixth reaction mixture;

(l) washing the separated reinforced precipitated silica with water; and

(m) drying the washed reinforced precipitated silica, wherein:

(n) said alkali metal silicate is lithium silicate, sodium silicate, potassium silicate, or a mixture thereof; and

(o) M is lithium, sodium, potassium, or a mixture thereof."

Independent claim 2 as granted comprises all the features of claim 1 but with most of the numerical ranges being narrower.

The patent in suit also contains product claims 21 to 42. Independent claims 21 and 33 as granted read as follows:

"21. Reinforced amorphous precipitated silica having, on a coating-free and impregnant-free basis, a surface area of from 220 to 340 square meters per gram, a pore diameter at the maximum of the volume pore size distribution function of from 9 to 20 nanometers, and a total intruded volume of from 2.6 to 4.4 cubic centimeters per gram."

"33. An elastomeric composition comprising:

- (a) crosslinked poly(diorganosiloxane) [sic], and
  - (b) reinforced amorphous precipitated silica having, on a coating-free and impregnant-free basis, a surface area of from 220 to 340 square meters per gram, a pore diameter at the maximum of the volume pore size distribution function of from 9 to 20 nanometers, and a total intruded volume of from 2.6 to 4.4 cubic centimeters per gram;
- said silica being distributed substantially uniformly throughout said crosslinked poly(diorganosiloxane)."

III. Opponent 1 had invoked sales of products falling under claim 21 of the patent in suit before the priority date thereof. In support of this objection, it relied on the following evidence:

P1: An internal letter ("Hausbrief") of Dr.Meon (Degussa) dated 15.08.1996;

P2: an offer letter from Degussa to Kalle AG, dated 25.3.1965;

P3: an internal Degussa report ("Besuchsbericht") dated 7.11.1984 of a visit to Kalle AG; and

P4: a printed document ("Schriftenreihe Anwendungs-technik Pigmente Nummer 2"), allegedly published on 15.5.1969.

IV. Opponent 2 had cited the following prior art documents:

D1: EP-B-0 157 703 and

D2: EP-A-0 031 288

and had also filed

R1: an experimental report included in a declaration of Frédéric Amiche dated 18 August 1998.

V. Concerning the objections of opponent 1, the opposition division concluded that the former had "failed to demonstrate that the product identified as being silica FK 3009 [*sic*] analysed in 1996 was identical with the products prepared and sold before the priority date of the patent in suit". The opposition division considered that the amendment consisting in the introduction of the phrase "*comprised of precipitated aggregates of ultimate particles of colloidal amorphous silica that have not at any point existed as macroscopic gel during preparation*" into claim 21 as granted according to the main request then on file did not satisfy the requirements of Article 123(2) EPC. Concerning the amended claim 21 of the first auxiliary request then on file, it came to the conclusion that in view of the results reported in R1, examples 18 and 19 of D1 were novelty-destroying although claim 21 as granted had been amended to include the phrase "*obtainable according to any of claims 1 to 20*". The opposition

division considered that the patent amended to contain only process claims 1 to 20 met the requirements of the EPC.

VI. In its statement of grounds of appeal, the appellant (proprietor of the patent) requested the maintenance of the patent as granted as a main request. It also filed five pairs of amended claims 21 and 33 which, according to the first to fifth auxiliary requests, were to be substituted for claim 21 and 33 as granted, respectively.

In claims 21 and 33 according to the first auxiliary request, the expression "*precipitated silica having*" as present in claims 21 and 33 as granted is replaced by the phrase "*precipitated silica **comprised of precipitated aggregates of ultimate particles of colloidal amorphous silica that have at no point existed as macroscopic gel during preparation wherein the silica has***" (emphasis added).

According to the second auxiliary request, the phrase "*, **said silica being obtainable by the process of any of claims 1 to 20***" is appended to the wording of claim 21 as granted and inserted at the end of part (b) of claim 33 as granted.

According to the third auxiliary request, the phrase "*, **and an average ultimate particle size of less than 0.1  $\mu\text{m}$  as determined per transmission electron microscopy***" is appended to the wording of claim 21 as granted and inserted at the end of part (b) of claim 33 as granted.



Moreover, according to all of the said auxiliary requests, the term "*poly(diorganosiloxane)*" is replaced by the corrected term "*poly(diorganosiloxane)*" (emphasis added).

The appellant argued that the subject-matter of the respective claims 21 and 33 was novel over D1. In this connection it referred to some earlier prior art documents acknowledged on page 2 of D1, i.e. to US-A-2 731 326, US-A-2 765 242, US-A-3 954 944, US-A-3 969 266, EP-A-0 031 271 and FR-A-2 353 486, as well as to US-A-5 911 963 and G. W. Sears Jr., Analytical Chemistry, Vol.28, no.12 (December 1956), pages 1981-1983.

It also relied on the following documents:

T1: Ullmanns Encyklopädie der technischen Chemie, 4<sup>th</sup> edition, 1982, pages 465 and 466, filed during the opposition proceedings

T2: Ullmann's Encyclopedia of Industrial Chemistry, 6<sup>th</sup> edition, 1998 Electronic Release, print-out of section Silica - Precipitated Silica - 7.2 Production, 2 pages filed during the opposition proceedings

T3a: R. K. Iler, "The chemistry of silica", 1979, John Wiley & Sons, page 174, filed with the grounds of appeal

T4: Hackh's Chemical Dictionary 4th edition, page 400, filed with the grounds of appeal

- VII. In its reply, respondent 2 (opponent 2) requested that the appeal be dismissed. Without going into any details, it alleged that the appellant's main and auxiliary requests did not fulfil the requirements of Articles 123(2)(3), 54 and 56 EPC.
- VIII. The parties were summoned to oral proceedings.
- IX. In its reply to the summons, respondent 1 (opponent 1) indicated that it would not attend. It requested the revocation of claims 21 to 42 of all the appellant's requests for lack of novelty and lack of inventive step. Concerning reasons, it merely referred to its notice of opposition, its written submission of 12 September 2003, and the documents cited therein.
- X. Respondent 2 announced in writing that it would not attend the oral proceedings and maintained its request that the appeal be dismissed.
- XI. Oral proceedings were held on 10 May 2006 in the absence of both respondents. At the oral proceedings, the appellant filed three complete sets of claims as new first, second and third auxiliary request, respectively. The independent claims of these sets are identical to the ones of the previous first, second and third auxiliary requests. During the oral proceedings reference was in particular made to page 554 of the textbook by R. K. Iler referred to above (hereinafter T3b).
- XII. The appellant requested that the decision under appeal be set aside and the patent be maintained as granted or in the alternative on the basis of one of the sets of

claims according to the auxiliary requests 1 to 3 filed during the oral proceedings.

XIII. Respondent 2 has not substantiated any of its objections at the appeal stage. Respondent 1 presented no argumentation in support of its objections but merely referred to its written submissions made during the opposition procedure. The arguments of the appellant as presented in writing and at the oral proceedings can be summarised as follows:

The term "*precipitated*" qualifying the silica of claim 21 was consistently used in the field to distinguish this particular type of silica from silica gel. This product-by-process feature limited the scope of the claims to silicas obtained in a particular manner, and in particular to those which have not at any point existed as macroscopic gel during their preparation. This "definition" was also given on page 2, lines 28 to 30 of the patent in suit. In case the claim was considered unclear in this respect, this definition was therefore to be used, even if considered as a non-standard definition. However, the said definition was not arbitrary but corresponded to the common definition of precipitated silica as found in standard textbooks such as T1, T2 and T3b. The process of D1 clearly required the formation and maturing of a gel, and it was specifically mentioned in D1 that the preparation of precipitated silica was a complex process during which a gel occurred and was ruptured. It was clear from D1, and in particular from the prior art references cited therein, that the expression "precipitated silica" as used in D1 encompassed different types of wet process silicas as opposed to

fumed silica. The use of this expression in D1 was thus imprecise or wrong since not in line with the "restricted" definition found in standard textbooks and in the patent in suit. Although the values for the surface area, the pore diameter and the total intruded volume of the silicas of examples 18 and 19 of D1 were within the claimed ranges, these silicas were not novelty-destroying for the subject-matter of claim 21, since they were not "true" precipitated silicas in the sense of the present patent. In the appellant's understanding, since the process according to D1 involved a gelification, the products obtained were not "true" precipitated silicas, but could be described as a kind of hybrid between silica gel and precipitated silica in the narrower sense, or "as silica gel coated with precipitated silica". Due to their different preparation process, the silicas of examples 18 and 19 of D1 were necessarily different, in terms of structure or of some other properties, from the ones according to the respective claims 21 of all requests. In particular, the claimed silicas had lower oil absorption values than the silicas of D1. Moreover, due to their more gel-type structure, the silicas of D1 would have a higher proportion of smaller pores and thus a higher Sears surface than the silicas of the present claims. The appellant was of the opinion that since it had pointed out specific properties (oil absorption and Sears surface) that would necessarily differ depending on the preparation process used, the burden of proof concerning lack of novelty over D1 rested with the respondents.

Concerning the allowability of the amendments in claims 21 and 33 according to the first auxiliary request, it

submitted that the paragraph bridging pages 10 and 11 of the application as filed provided an additional basis.

Concerning the third auxiliary requests, the appellant argued that the subject-matter of claims 21 and 33 was novel over examples 18 and 19 of D1 since D1 did not disclose the average ultimate particle size. Furthermore, the appellant pointed out at the oral proceedings that no evidence had been presented concerning the porosity of the FK 300 DS silicas products referred to in P2 to P3 at the time of the alleged sales and/or publication date. The fact that the BET surface of the FK 300 DS product mentioned in P4 fell within the range of claim 21 of the patent in suit did not necessarily mean that the porosity related requirements of claim 21 of the patent in suit were also met. The measurements described in P1 were undertaken in 1996 on a more recent FK 300 DS product. The mere fact that it bore the same trade name did not permit the conclusion that the earlier silica products referred to in P2 to P4 had the same porosities as the tested silica, in particular since porosity was not a property addressed in these documents. The appellant also submitted that the prior art cited did not suggest the preparation of silicas having all of the claimed properties, which silicas were suitable for being used in various applications.

## Reasons for the Decision

### *Main request*

#### 1. Novelty

1.1 Document D1 relates to the preparation of precipitated silica having specific properties such as a high oil absorption (DBP) in conjunction with a high CTAB surface area. The silica is to be used as filler ("charge") in silicone polymers, rubbers and thermoplastic polymers, in particular as reinforcing filler in elastomers like rubber, more specifically for improving the abrasion resistance thereof or as thickening agent ("épaississant"). Reference is made to page 2, lines 3 to 5; page 2, line 61 to page 3, line 1; page 3, lines 14 to 16; page 4, lines 1 to 3 and lines 24 to 25; and claims 1, 2, 12 and 13.

1.1.1 D1 is silent about the pore diameter at the maximum of the volume pore size distribution function and about the total intruded volume of the silicas disclosed. However, as also acknowledged by the appellant in its statement of grounds of appeal (page 3, 2<sup>nd</sup> paragraph), R1 shows that for the silicas obtained by reproducing examples 18 and 19 of D1, the numerical values of these properties lie within the ranges specified in claim 21 of the patent in suit. The BET surface areas of these two silicas also fall within the claimed ranges.

1.1.2 The silicas of D1, and in particular the ones of examples 18 and 19, are obtained by reacting aqueous solutions of a silicate, in particular sodium silicate, and an acid, in particular sulphuric acid, in a multi-

step reaction. In a first reaction step the acid is added until apparent gel formation. Acid addition is stopped and the gel is then aged ("mûrissement") for a certain time. In subsequent steps, further acid is added to bring the pH to a value of 9 or less, this step being followed by either the simultaneous addition of acid and silicate at a pH between 7 and 9 (claim 3, example 18) or by the addition of further acid (claim 4 and example 19). An electrolyte is added in at least one of these steps. The final suspension is filtered, and the filtered product is washed and dried. The preparation process of D1 permits to use elevated reaction temperatures of from 70 to 95°C, see claims 1, 3 and 4 for the states BE to SE and page 4, lines 11 to 14. Examples 18 and 19 mention a reaction temperature of 90°C and agitation of the reaction mixture. As indicated in examples 18 and 19, the reaction leads to a suspension ("bouillie") of silica, i.e. to small discrete silica particles, see page 10, lines 31, 33 and 50. Considering the particular preparation methods disclosed, it is understood that the silicas obtained are amorphous and reinforced, and the appellant has not argued the contrary. The final products obtained are referred to *expressis verbis* as silica obtained by precipitation or as precipitated silica (see the expressions "silice de précipitation" and "silice précipitée" used e.g. in claims 1 to 4 of D1) despite the fact that a gelification occurs during a first stage of their preparation.

- 1.1.3 Thus D1 discloses precipitated silicas having all the properties required by present claim 21.

1.2 The appellant considered that the silicas disclosed in D1 were not "true" precipitated silicas within the meaning of the patent in suit since they involved the formation and aging of a gel and thus necessarily had properties making them different from the precipitated silicas according to present claim 21. However, the supporting evidence and arguments submitted by the appellant do not, for the following reasons, convincingly establish that a skilled person would necessarily understand that the expression "*precipitated silica*" as used in claim 21 of the patent in suit excluded the precipitated silicas prepared as described in examples 18 and 19 of D1.

1.2.1 The appellant argued that "a clear definition of precipitated silica" was given in the patent in suit. The passage invoked (page 2, lines 28 to 30) reads as follows: "*Precipitated silica, **then, may** be described as precipitated aggregates of ultimate particles of colloidal amorphous silica that have not at any point existed as macroscopic gel during the preparation*" (emphasis added by the board). The board acknowledges that a patent may be regarded as a specific glossary for the claimed features. However, the board notes, first of all, that the statement on page 2, lines 28 to 30 of the patent in suit is not worded as a "definition" of the expression "precipitated silica", let alone a definition generally applicable to all occurrences of the said expression in the patent in suit. By virtue of the term "*then*", this passage can be considered to refer back to the commercial prior art methods for preparing precipitated silica discussed in the previous paragraph of the description. Moreover, it has to be noted that as far as the claimed silica



products are concerned, the patent contains no other passage generally imposing the absence of macroscopic gel formation during their preparation. The sentence bridging pages 5 and 6 which mentions "*avoiding gellation on the macro scale*" merely concerns one aspect of the preparation process since none of the process claims states that gellation should be avoided. Finally, the use of the term "*may*" in the quoted sentence makes it even further questionable whether the suggested description is intended to generally apply to all the claimed silicas. In view of the above considerations, the quoted passage cannot be considered as a clear, unambiguous and compelling definition that could justify a narrower reading of the expression "*precipitated silica*" in claim 21, excluding the silicas of examples 18 and 19 of D1 because their preparation involves the formation of a gel in addition to a precipitation step.

- 1.2.2 On the other hand, it is noted that D1 describes a reaction involving bringing together solutions of an alkali metal silicate and a mineral acid at elevated temperature and under stirring and leading to a suspension of silica particles designated as "*precipitated silica*". The preparation of precipitated silicas is generally referred to as a complicated process involving a polymerisation with the occurrence and rupturing of a gel (page 2, lines 6 to 7). The silica products obtained by the method disclosed in D1, like the silicas obtained by other prior art wet processes mentioned on page 2 of D1 are designated as "*precipitated silicas*", as opposed to pyrogenic silica (page 2, lines 52 to 53). Since even according to the appellant's understanding the method of D1 involves,

after the gel formation, further steps with addition of reactants wherein precipitation of silica occurs, the board does not share the appellant's view that the designation of the silica obtained as "precipitated silica" is wrong or imprecise.

As observed by the board during the oral proceedings, the authors of D1, when describing their preparation process and the silicas obtained, were aware of the contents of the textbook from which T3b is extracted. This can be inferred from page 2, lines 43 to 45 of D1. In T3b, a page especially mentioned on page 2, line 11 of the patent in suit, the following is stated under the heading "PRECIPITATED SILICA POWDERS":

"Consideration is given here only to silica powders that consist of aggregates of ultimate particles of colloidal dimensions that have **not at any point existed as macroscopic gel** during the preparation. This is a **purely arbitrary distinction**, but serves to separate the voluminous literature on **silica gel** in **massive, granular, or pulverised** form, from that on all the other types of fine silica powders made by precipitation from the gas phase and from solution."

(emphasis added by the board). The board notes that the cited passage is taken from a standard textbook on the chemistry of silica, whose author however himself considers the distinction he makes to be "arbitrary". Moreover, when considered in its entirety, the quoted passage can be understood to focus the subsequent discussion of silica powders onto those which are obtained by precipitation from the gas phase or from solution as opposed to those obtained by pulverising larger pieces of solid silica gel. This appears to be confirmed by the sentence following the said passage,

which points out differences between "gels that have been pulverised" and "precipitated silica powders". It is thus not surprising that the content of T3b did not keep the authors of D1 from designating their products as "precipitated silica".

- 1.2.3 Moreover, the application leading to the patent D1 having been filed by a chemical company, it can be assumed that the applicant and the inventor were also aware of the common general knowledge in the field in question. From T1, published in 1982, it can be gathered that at the time of its publication the usual industrial preparation of precipitated silica involved forming discrete primary silica particles under strong stirring and at elevated temperature, without passing through an intermediate gel state as in the case of silica gel preparation (see the paragraph bridging pages 465 and 466). However, this does not mean either that the designation "precipitated silica" used later (priority year 1984) by the authors of D1 was somehow erroneous. T2 was published after the priority date of the patent in suit and merely confirms that in 1998 precipitated silica was still prepared industrially in such a manner that "a coherent system and thus a gel state is avoided by stirring and increasing the temperature".
- 1.2.4 Although T1, T2 and T3b show that the preparation of precipitated silicas is usually distinguished from the preparation of silica gel in that the former method does not involve formation of a macroscopic gel, it is not clearly apparent from the patent in suit (see point 1.2.1 above) that the expression "*precipitated silica*"

used in claim 21 does not encompass precipitated silicas of the particular type disclosed in D1.

- 1.2.5 It follows from the above that the subject-matter of claim 21 is not novel. The appellant's main request can thus not be granted.

*First auxiliary request*

2. Novelty

- 2.1 Amended claim 21 now requires that the silica is "*comprised of precipitated aggregates of ultimate particles of colloidal amorphous silica that have not at any point existed as macroscopic gel during preparation*". This additional feature does not, as such, suffice to make the subject-matter of present claim 21 novel, since a product is not necessarily new only for the reason that it is prepared by a new process. Respondent 2 has shown by means of R1 that for the silicas of examples 18 and 19 of D1 the values of those three characterising properties which are quantified in claim 21 fall within the indicated ranges. Under these particular circumstances, the board holds that the burden of proof now rests with the appellant, who has to show by means of suitable evidence that the precipitated silicas obtained in the manner now further specified in amended claim 21 by means of process features are, as such, necessarily different from the ones disclosed in D1. Merely pointing out that the preparation process parameters, and in particular the passing via a gel stage as in D1, will have a direct influence on the properties and structure of the silicas obtained, without even identifying and

quantifying these effects, is not sufficient to this end.

- 2.2 The appellant has also tried to establish novelty by invoking differences in terms of specific properties not addressed in the patent in suit, namely the lower "oil absorption (DBP)" and "Sears surface" of the claimed silicas as compared to the ones of D1.
- 2.2.1 The oil absorption (DBP) of 304.8 ml/100g measured by the appellant for example III of the patent in suit is substantially lower than the value of 410 ml/100g reported in R1 for both the silicas of examples 18 and 19 of D1. However, as pointed out by the board during the oral proceedings, the two silicas according to examples 18 and 19 of D1 differ in terms of their surface area, pore diameter and total intruded volume values, from the silica of example III. In particular, the total intruded volume measured using mercury porosimetry is substantially greater for the former (4.0 and 3.8 cm<sup>3</sup>/g, respectively) than for the latter (3.2 cm<sup>3</sup>/g). It cannot be deduced therefrom that even with a claimed silica having a total intruded volume of e.g. 4 to 4.4 cm<sup>3</sup>/g (higher part of the claimed range) the oil absorption would still remain at the level stated in example III. The appellant could not give any reason why the oil absorption would remain at this level even with a higher total intruded volume and the file contains no evidence in this respect.
- 2.2.2 The board notes that the patent is silent about the oil absorption of the silica produced, let alone about possible effects of the avoidance of an intermediate macroscopic gel formation on this particular property.

No general conclusion can thus be drawn from the patent itself having regard to the oil absorption of all the silicas obtainable by precipitation processes not involving such an intermediate formation of a macroscopic gel. The experimental evidence relied upon by the appellant concerns a single silica (example III) obtained by one very specific process falling under the process claims 1 to 20. Although the opposition division already held that in view of the multiple preparation process parameters involved, the difference noted at the level of a single example was not significant and could not even be extrapolated to all the silicas obtainable by the **claimed** processes (see Reasons, point 12.), the appellant has not provided further experimental evidence. As pointed out by the appellant, D1 aims at achieving high oil absorptions and uses a preparation process with occurrence of a gel. However, D1 does not specifically relate the gel formation to the oil absorption values obtained. It is thus not apparent from D1 whether it is the nature and amount of some more gel-like parts of the silica, rather than other structural features resulting from the preparation process, which are essentially responsible for the oil absorption properties. Under these circumstances, and in view of the low significance the comparison of example III with examples 18 and 19 of D1 (see point 2.2.1), the board is not convinced that all the silicas falling under present claim 21 will necessarily have a lower oil absorption (DBP) than the ones of D1.

- 2.2.3 The appellant also argued that the silicas of examples 18 and 19 of D1, due to their more gel-type structure, would generally have a higher proportion of smaller

pores not accessible to nitrogen in BET surface measurements, and hence a higher Sears surface area than precipitated silicas prepared without occurrence of gel formation. The patent in suit itself does not mention the Sears surface area at all, and it does also not contain any information from which it might be derived that the Sears surface values of the claimed silicas would be relatively low. Even accepting, purely for the sake of argument, that a silica with a more gel-like structure will generally have more smaller pores, the extent to which this interrelation will be reflected in the very specific "hybrid" precipitated silicas of D1 can only be speculated about in the absence of suitable evidence. However, the appellant has not actually measured and compared the Sears surface of silicas obtained as described in examples 18 and 19 of D1 and of a silica obtained by a process as claimed, i.e. without the occurrence of a macroscopic gel. It has thus not convincingly established that silica according to present claim 21 would always have a significantly lower Sears surface than the silicas of D1 because of the differences in terms of the preparation method used.

- 2.3 The appellant has thus not discharged the burden of proof resting on it. Since the silicas according to present claim 21 do not necessarily and unequivocally differ from the ones of examples 18 and 19 of D1 in terms of some further properties caused by their respective preparation processes, the subject-matter of the present product-by-process claim 21 cannot be acknowledged to be novel and the first auxiliary request cannot be granted either.

*Second auxiliary request*

3. Novelty

3.1 In comparison to claims 21 and 33 as granted, present claims 21 and 31 additionally require that the precipitated silica is "*obtainable by the process of any of claims 1 to 20*". By virtue of the referral back to claim 1, present claim 21 was amended to contain more features of the product-by-process type. The opposition division has denied novelty having regard to this claim (see minutes of the oral proceedings point 4., and reasons of the contested decision, points 11 to 13).

3.2 For the same reasons as in the case of the first auxiliary request (see point 2.1 above), the burden of proof rests with the appellant. The fact that the present amended claim 21 refers to the more specific preparation process of claim 1 is not as such a sufficient reason for shifting this burden to the respondents. The opposition division has emphasised that considering the multiple critical parameters having an influence on the properties of the silicas prepared, process claim 1 was formulated broadly (see point 12. of the contested decision). It therefore held that the difference found for the single example III in terms of oil absorption could not be extrapolated to all the silicas covered by a claim 21 referring to the preparation process of claim 1. In the absence of evidence to the contrary, for reasons analogous to those given in points 2.1 to 2.2.3 with respect to claim 21 according to the first auxiliary request, the board comes to the conclusion that the evidence



presented is not sufficient to demonstrate that all the silicas covered by present claim 21 necessarily and unequivocally differ from the ones of examples 18 and 19 of D1 in terms of some further properties caused by their respective preparation processes.

- 3.3 The subject-matter of the present product-by-process claim 21 can thus not be acknowledged to be novel and the second auxiliary request cannot be granted either.

*Third auxiliary request*

4. Allowability of the amendment

In comparison to claims 21 and 33 as granted, present claims 21 and 31 are amended to additionally specify that the precipitated silica has "*an average ultimate particle size of less than 0.1  $\mu\text{m}$  as determined by transmission electron microscopy*". Basis for this amendment can be found on page 16, lines 2 to 6 of the application as filed. The amendment thus meets the requirements of Article 123 (2) and (3) EPC.

5. Novelty

- 5.1 D1, D2 and R1 are silent about the ultimate particle sizes of the silicas referred to therein. The respondents have neither argued nor presented evidence showing that the silicas disclosed in D1 or D2 would inherently have an ultimate particle size of less than 0.1  $\mu\text{m}$ . At the oral proceedings, upon being questioned by the board, the appellant stated that particle growth beyond this size was possible and that greater ultimate particle sizes existed. In the absence of evidence to

the contrary, the board concludes that neither D1 nor D2 clearly and unambiguously discloses an average ultimate particle size of less than 0.1  $\mu\text{m}$ , let alone in combination with surface area and porosity values as required by present claim 21.

5.2 P2 and P3 appear to show that silica products were commercialised by the company Degussa under the trade name FK 300 DS as early as 1965 and still in 1984. P4, a partial copy of a document bearing no apparent indication of its publication date, also refers to a product labelled FK 300 DS, which is a precipitated silica ("Fällungskieselsäure"), see page 2. The alleged publication date of this document (15 May 1969) has not been disputed by the respondents. In P4, the silica FK 300 DS is stated to have a BET surface area of 330  $\text{m}^2/\text{g}$  and an average ultimate particle size ("Mittlere Größe der Primärteilchen") of 0.01  $\mu\text{m}$  ("10 Millimikron"), see the upper table on page 4. However, like P2 and P3, P4 is silent about the porosity of this product. P1, an internal letter of Degussa dated 15 August 1996 relating to the patent in suit, reports the results of tests carried out to investigate the properties (BET surface area, macropore volume, and pore diameter) of different silicas, including a FK 300 DS of unspecified production date, see page 1. The author of P1 additionally asserts that the data determined for the FK 300 DS product fall within the claimed ranges of the patent in suit and that FK 300 DS has been sold since 1965 (see points 1. and 3. on page 2).

5.2.1 In the contested decision, the opposition considered that the opponent had failed to demonstrate that the

product analysed in 1996 was identical with the products prepared and sold before the priority date of the patent in suit. Despite this finding, respondent 1 has not submitted evidence to this effect in the appeal procedure but merely referred to its written submissions made during the opposition procedure. In the absence of such evidence, even assuming that documents P1 to P4 were sufficient to show a public prior use of a precipitated silica bearing the trade name FK 300 DS and having the properties mentioned in P4 in the period from 1965 to 1984, the board sees no reason to diverge from the opinion of the opposition division. The arguments that respondent 1 had submitted in point 6. of its letter of 12 September 2003 are not convincing for the following reasons: As emphasised by the appellant, identical trade names of products are generally not sufficient to establish the identity of all their properties, since the latter may vary over the years without the trade name being changed. In view of such possible variations the close similarity, in terms of BET surface, of the FK 300 DS described in P4 and the one subjected to measurements in 1996 does not imply that the other properties of the two products would also be closely similar, let alone properties which are not even specified in P4. Points 1. and 3. on page 2 of P1, do not, as alleged in the said letter of 12 September 2003 (see point 6. second paragraph), contain any statement concerning the identity of any FK 300 DS actually sold and the one subjected to measurements in 1996.

5.2.2 Summarising, P1 to P4 do not establish that silicas having all the properties required by present claim 21

have been sold or otherwise disclosed before the priority date of the patent in suit.

- 5.3 The subject-matter of independent claim 21, and of claims 22 to 42 which refer back to and hence comprise all the features of the silicas of claim 21, is thus novel.
6. Inventive step
- 6.1 D1 can be considered to represent the closest prior art. Taking D1 as the starting point, the technical problem to be solved by the claimed silicas can be seen in providing further precipitated silicas having properties making them suitable for use in many different applications, in particular for clarifying beer, for reinforcing silicon rubber, but also as fillers and extenders in toothpaste, as carriers for vitamins, as paper extenders and brighteners, and in a multitude of other uses, see the patent in suit, the paragraph bridging pages 2 and 3; and page 3, lines 7 to 12. In the absence of evidence to the contrary, the board accepts that this technical problem is solved by the silicas of claim 21.
- 6.2 The respondents have not substantiated any inventive step objection against the claims according to the present request. In the absence of any evidence to the contrary, the board accepts the information provided by the appellant, i.e. that average ultimate particle sizes of more than 0.1  $\mu\text{m}$  were possible and existed. None of D1 and D2 addresses the average ultimate particle size, the pore diameter at the maximum of the volume pore size distribution or the total intruded

volume of the silicas disclosed. Moreover, D2 is merely concerned with silicas for use as reinforcing fillers for organo-silicic compositions, and D1 only mentions the use of the silicas disclosed as filler, in particular reinforcing filler, for polymers and as thickening agent. The board thus comes to the conclusion that neither D1 nor D2 suggest silicas suitable for various purposes and having, in combination with the other properties required by present claim 21, an average ultimate particle size of less than 0.1  $\mu\text{m}$ .

6.3 P4 mentions a mean ultimate particle size ("Mittlere Größe der Primärteilchen") of 10nm for the FK 300 DS silica. However, P4 also does not address the porosity of the silica at all and contains no details concerning the preparation method used. Moreover, it only relates to the use of precipitated silica in paper coating. Hence, it can also not suggest obtaining silica particles suitable for various applications and having all the properties required by present claim 21.

6.4 The subject-matter of independent claim 21, of independent claim 33 which relates to an elastomeric composition comprising a silica with all the features of the silica of claim 21, and of dependent claims 22 to 32 and 34 to 42 depending on either of these two independent claims, is thus based on an inventive step.

The scope of the opposition by respondent 2 was expressly restricted to product claims 21 to 42 of the patent in suit. Although the notice of opposition of respondent 1 was formally directed against all the claims of the patent as granted ("Widerruf des

erteilten Patents für alle Ansprüche"), respondent 1 has not at any point of the opposition and appeal procedures raised or substantiated specific objections against process claims 1 to 20 as granted, which claims are identical with claims 1 to 20 according to the present request. The board considers that the subject-matter of process claims 1 to 20 is not objectionable under Article 100(a) EPC in view of the prior art relied upon by the respondents. Under the present circumstances, and since this was not disputed, detailed reasons need not be given.

7. The complete set of claims filed as third auxiliary request during the oral proceedings corresponds the third auxiliary request filed earlier in writing, which was presented in a more condensed form referring to the granted claims 1 to 20, 22 to 32, 34 to 42. Since the respondents have been given the opportunity to comment on this request, the decision complies with the requirement of Article 113(1) EPC.

**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 maintain the patent on the basis of the set of claims 1 to 42 according to the third auxiliary request filed during the oral proceedings and a description to be adapted.

The registrar:

The chairman:

C. Vodz

M. Eberhard