Datasheet for the decision
of 22 August 2006

Case Number: T 0623/03 - 3.3.05
Application Number: 93920405.3
Publication Number: 0609433
IPC: C09C 1/50
Language of the proceedings: EN
Title of invention:
Carbon blacks
Patentee:
CABOT CORPORATION
Opponent:
Degussa-Hüls Aktiengesellschaft
Headword: -
Relevant legal provisions:
EPC Art. 56
Keyword:
"Novelty, inventive step (yes; after amendment)"
Decisions cited: -
Catchword: -
Case Number: T 0623/03 - 3.3.05

DECISION
of the Technical Board of Appeal 3.3.05
of 22 August 2006

Appellant: CABOT CORPORATION
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Decision under appeal: Decision of the Opposition Division of the European Patent Office posted 16 April 2003 revoking European patent No. 0609433 pursuant to Article 102(1) EPC.

Composition of the Board:
Chairman: M. Eberhard
Members: H. Engl
J. Willems
Summary of Facts and Submissions

I. European patent EP 0 609 433 B1 was granted based on European patent application 93 920 405.3, claiming the priorities of 27 August 1992 (US 935794) and 25 June 1993 (US 81881).

II. The European patent was subsequently revoked in opposition proceedings by a decision of the opposition division posted on 16 April 2003. The decision was based on the claims of the main request filed with letter of 28 February 2003 and a first and second auxiliary request filed during oral proceedings. The claims of the main request corresponded to the granted claims 1 - 12, except for a disclaimer added to claim 7.

III. The following documents were inter alia cited during the opposition procedure:

D2: WO A 93/10194, published on 27 May 1993
D8: Abstract of JP A 03 014848 and partial translation thereof
D9: Abstract of JP A 01 229074
D10: Abstract of CS A 255823
D11: ASTM D1765-73a (1975)
D13: Abstract of JP A 51 004088
D15a: "Arbeitsanweisung" "Bestimmung der Aggregatgrößenverteilung durch Photosedimentometrie DCP" by Dr Roller, dated 4 December 2002
D15b: Sample characterisation by Dr W Niedermeier, dated 4 December 2002
D16: Declaration of E. Sroka, dated 15 January 2002
D16a: Experimental Report on Sapex 20, dated 7 November 2002
D17: Declaration of Roger Albright, dated 31 May 2002
D17a: Measurement results N762 Lot No. A103061B3
D17b: Measurement results N774 Lot No. A102221B1
D17c: Measurement results N754 Lot No. A199093327
D17d: Measurement results N787 Lot No. A107110B4
D19: US A 4 221 772

Documents D15a, D15b, D16, D16a, D17 and D17a-d were relied upon as evidence for alleged public prior uses of different carbon blacks.

IV. The grounds for the revocation were as follows:

The subject matter of claim 1 in accordance with the main request lacked novelty having regard to D8. The subject matter of claim 1 according to the first auxiliary request lacked novelty having regard to document D11. Alleged prior uses of carbon furnace blacks based on documents D15, D15a, D16 and D16a were found inconclusive by the opposition division. However, the subject matter of claims 3 and 5 in accordance with the second auxiliary request was considered not to involve an inventive step having regard to the disclosures of documents D2 and D19, respectively.

V. An appeal was filed against the revocation of the patent by the patentee (henceforth: the appellant) with letter of 2 June 2003.

With the statement of the grounds for appeal, received on 26 August 2003, the appellant submitted new claims as a main and an auxiliary request. Also submitted were
arguments on the novelty and inventiveness of the subject matter of the claims having regard in particular to documents D2, D8, D11, D13 and D19.

VI. With his letter of reply, the respondent (opponent) filed literature regarding the ASTM and Japanese standards for determination of the dibutylphthalate absorption number (DBP) and the iodine number (I₂No.) which were, in his submission, directly comparable. He relied in particular upon documents


Also filed were the respective translations into English D8a, D9a, D10a and D13a of documents D8, D9, D10 and D13.

VII. In the annex to the summons for oral proceedings, the board raised the question of added subject matter concerning claim 7 of the main request and claims 1 and 5 of the auxiliary request. The board commented on the questions of novelty of the subject matter of claims 1, 2, 5, 7 and 12 of the main request; and of claims 2 and 10 of the auxiliary request. Concerning inventive step, D2 was considered as the closest prior art with respect to the carbon blacks of claims 7 and 8
of the main request and a technical problem was formulated taking into account that no comparison with the carbon blacks of D2 had been made.

VIII. In reply to the summons for oral proceedings the appellant submitted new arguments and new claims as a main and seven auxiliary requests.

Regarding the alleged prior use based on the Sapex 20 samples, *inter alia* documents

D24: G. Joyce, "the CARBON AGGREGATE", Vol. 12, Nr. 2, 2005, pages 1 - 8 (an ASTM International Publication); and

D26: Declaration of J. R. Wilson, dated 21 July 2006

were submitted to show that iodine numbers would substantially change over time. The appellant argued that the recent analysis of these old samples could not, therefore, prove lack of novelty of claim 5.

On inventive step, the appellant defined the technical problem to be solved, apparently starting from D2, as providing a furnace carbon black with physical properties that render it particularly advantageous for use in rubber and plastic compositions where compound processing is important. In connection with D11 the appellant pointed to the significance of the M-Ratio as evidenced in Table 8 of the patent specification showing the improvements in terms of mixing energy and extrusion shrinkage. Said improvements could be attributed to the claimed M - Ratio which was not known from D11.
IX. Oral proceedings took place on 22 August 2006. At the beginning of the oral proceedings, the appellant filed four sets of amended claims as a main request and three auxiliary requests to replace all previous requests. He withdrew the main and the first auxiliary requests later on and made the second and third auxiliary requests the main and the first auxiliary requests respectively.

The claims of said main request read as follows:

1. A furnace carbon black having an I$_2$No. of 51-62 mg/g according to ASTM Test Procedure D1510, and a DBP of 61-125 cc/100g, and an M-Ratio of 1.25-2.00.

2. A composition of matter comprising a material from the group consisting of rubber and plastics, and a furnace carbon black of claim 1.

X. The essential arguments of the parties may be summarized as follows:

The appellant:

Having regard to inventive step, the appellant discussed during oral proceedings the experimental data contained in Tables 10 and 13 of the patent specification, reporting the properties of carbon blacks incorporated in EPDM rubber. Example 8 of Table 13 illustrating the invention as claimed in claim 1 of the main request would show a lower extrusion shrinkage and a higher extrusion rate and therefore superior properties in comparison with EPDM
samples comprising conventional carbon blacks (Control A and B). Comparing Control B and the invention, the rebound (a measure of the rubbery properties) and compression set of these samples would surprisingly be similar to those of Control B in spite of the large differences in iodine numbers. This was, according to the appellant, an indication of the presence of an inventive step. The beneficial effect attributable to the M - Ratio in the claimed range could be studied from Table 10 of the specification. Both the rebound value and the compression set (a measure of the permanent deformation of the rubber after release of the compressive force) of sample 6, which has an M - Ratio in the claimed range, would be better than in Control D which exhibits an M-Ratio value lying outside the claimed range.

These results show, according to the appellant, that the problem of the invention, defined in paragraph [0007] of the patent in suit, and consisting in providing carbon blacks which impart rubbers and plastic compositions advantageous processing properties and mechanical properties, was actually solved.

These beneficial effects would be attributable to the particle size distribution, characterized in the claims by the M - Ratio. The invention deliberately deviated from a conventional symmetric particle size distribution. It should therefore be considered as non-obvious.
The respondent:

The respondent accepted novelty of the subject matter of claims 1 and 2 of the main request.

The respondent considered the furnace carbon black N440 listed in document D11 to represent the closest prior art. Said ASTM standard furnace black would exhibit a typical I_2No. of 50 (determined in accordance with ASTM D1510) and a DBP absorption of 60 cm$^3$/100g (in accordance with ASTM D2414). Said standard test procedures ASTM D1510 and ASTM D2414 would typically exhibit an experimental error such that the I_2No. and DBP values of N440 would be indistinguishable from the furnace blacks claimed in claim 1 (see the standard test methods in D22b and D22c, respectively). The only difference was thus to be seen in the claimed M-Ratio, which was not reported in D11.

The respondent defined the technical object of the claimed invention, having regard to standard black N440, as providing an alternative furnace carbon black.

The respondent disputed that any improvement could be acknowledged from the experimental data contained in the opposed patent. He pointed out that particular properties of the EPDM compositions (extrusion rate and extrusion shrinkage) for which a positive effect could be seen in example 8 of Table 13 were counterbalanced by negative results in example 6 of Table 6. Therefore, the claimed subject matter should not be seen as an improvement of the art. Moreover, the underlying technical problem was not clear, as no comparison with the closest prior art had been made.
The respondent's main argument on inventive step was that furnace carbon blacks having an M-Ratio in the claimed range were known in the art. This was shown in reports D17a-d giving the results of measurements for furnace carbon blacks N762, N774, N754 and N787, respectively. According to the declaration by R. Albright (D17), these furnace carbon blacks were produced and sold before the first priority date of the opposed patent.

Another furnace carbon black designated as Sapex 20 and having an M-Ratio of 1.64 was manufactured and sold in 1984 (D16 and D16a). The respondent concluded that furnace carbon blacks having an M-Ratio in the claimed range were usual in the art, all the more so as the products N762, N774, N754 and N787 were standard carbon blacks as shown in D11. In the appellant's own submission, the M-Ratio could be chosen independently from iodine number and DBP. There was thus no reason why the skilled person should not consider carbon blacks having the claimed M-Ratio as an alternative to known standard carbon blacks such as N440.

XI. The appellant requested that the decision under appeal be set aside and that the patent be maintained in amended form on the basis of the set of claims of the main request or alternatively of the auxiliary request, both filed during oral proceedings.

The respondent requested that the appeal be dismissed.
Reasons for the Decision

1. The appeal is admissible.

2. Amendments (main request)

Claim 1 of the main request is based on claim 13 and the description, page 3, lines 4 - 15, page 10, lines 1 and 2, of the PCT application WO-A-94/05732 in its published version.

Claim 2 is based on claims 13 and 22 and the description, page 3, lines 4 - 15 and page 10, lines 1 and 2, of the said PCT application.

New claims 1 and 2 correspond to claims 10 and 12 of the granted patent, with the additional definition of the I_2No. test method in the new claims.

The claims therefore meet the requirements of Art. 123(2) and (3) EPC.

3. Novelty (main request)

Novelty of the subject matter of claim 1 (which mainly corresponds to granted claim 10) was not contested during the appeal or opposition procedure.

The board is also not aware of prior art disclosing all the features of claim 1 in combination.

Document D2 was published on 27 May 1993 with the priority of 13 November 1991. Because current claim 1 is entitled to the priority right as of 27 August 1992.
and D2 meets the requirements of Art. 158(1) and (2) EPC, this document constitutes prior art under the provisions of Article 54(3)(4) EPC. D2 discloses carbon blacks having an \( I_2 \)No. of 26 - 34 mg/g and thus outside the claimed range.

D8/D8a discloses carbon blacks having an \( I_2 \)No. of between 18 and 28 mg/g, outside the claimed range.

The subject matter of claim 2 derives its novelty from claim 1.

The claims of the main request thus meet the requirements of Art. 54 EPC.

4. **Inventive step (main request)**

4.1 Claim 1 is directed to a furnace carbon black which is characterized by the combination of three parameters:

- \( I_2 \)No. (iodine number) of 51 - 62 mg/g, as determined in accordance with ASTM Test Procedure D1510;

- DBP (dibutyl phthalate) absorption value of 61 - 125 cm\(^3\)/100g; and

- M - Ratio of 1.25 to 2.00.

Standard test procedures for measuring the iodine number and DBP absorption have been developed (see D22a: ASTM D1510-85; D22b: ASTM D1510-85e1; D22c: ASTM D2414-86; and D22d: Japanese standard JIS-K-6221-1982). Iodine number and DBP absorption are routinely used for
characterizing carbon blacks (see D2, D8, D9, D10, D13 and D19).

The M - Ratio is defined in the patent in suit (paragraphs [0003], [0043] and [0044]; Figure 2) as the ratio \( M = \frac{D_{st}}{D_{mode}} \), wherein \( D_{st} \) is the median Stokes diameter (i.e., the Stokes diameter of the carbon particles of the sample where 50% by weight of the sample is either larger or smaller) and \( D_{mode} \) is the Stokes diameter at the peak (maximum) of the Stokes particle size distribution curve. As pointed out by the appellant, the M - Ratio may thus be roughly said to reflect the asymmetry of the Stokes particle size distribution (its deviation from a Gaussian distribution).

4.2 Closest prior art

The respondent has identified the furnace carbon black N440 listed in document D11 (page 406, Table 1, last but one entry) as the closest prior art. Said ASTM standard furnace black is reported to exhibit a typical \( I_2N_{o.} \) of 50 g/kg (determined in accordance with ASTM D1510) and a DBP absorption of 60 cm\(^3\)/100g (determined in accordance with ASTM D2414).

Inspection of said standard test procedures ASTM D1510 and ASTM D2414 (in D22b and D22c, respectively) reveals that the \( I_2N_{o.} \) and DBP values of N440 fall within the ranges of claim 1, when one takes into account the experimental margins of error of ± 1.8 g/kg and ± 2 % (or ± 2.1 %), respectively, associated with said standard test procedures (see D22b, page 385, chapter 12.4.2.; and D22c, page 618, chapters 12.3.2. and 12.4.2.).
12.4.2.). Applying this experimental uncertainty, the carbon black N440 becomes indistinguishable from the subject matter of current claim 1, as far as I₂No. and DBP absorption are concerned. The board is therefore of the opinion that these two parameters cannot establish novelty of the claimed subject matter.

The claimed subject matter is novel, however, because D11 is silent on the M - Ratio of furnace carbon black N440. D11 does also not report any processing characteristics or mechanical properties of rubber and plastics compositions comprising said furnace carbon black N440.

4.3 Technical problem and solution

According to paragraph [0007] of the patent in suit, the latter aims at providing new classes of furnace carbon blacks which are advantageous for use in rubber and plastic compositions where compound processing properties such as mixing energy, viscosity, extrusion shrinkage, and mechanical properties of the compound, such as hardness, compression set and resistivity, are important. At the oral proceedings, the appellant argued on the basis of the data in Tables 8 and 13 of the patent in suit that the claimed carbon blacks, when incorporated in EPDM (ethylene propylene diene polymethylene) rubber compositions led to improved extrusion rate and extrusion shrinkage and thus to a better dimensional control during the extrusion operation. This was, however, disputed by the respondent.
The opposed patent contains experimental data on the properties of carbon blacks incorporated in EPDM rubber (Tables 7 to 13). Of the examples provided in the patent specification, only example 8 illustrates the invention as claimed in claim 1 of the main request. The results are summarized in Table 13, compared with EPDM samples comprising conventional carbon blacks (Control A and B). The comparison shows that sample Control A (having a \( I_2 \) No. of 35.4, a DBP of 91 and an M-Ratio of 1.07, the latter being outside the claimed range), exhibits a higher extrusion shrinkage and a lower extrusion rate and is therefore inferior with respect to these parameters to inventive sample 8. However, rebound and compression set of Control A are improved over example 8.

Table 10 compares EPDM samples comprising two carbon blacks which differ essentially only in the M-Ratio, \( I_2 \) No. and DBP absorption being comparable. It is to be noted that the \( I_2 \) Nos. of example 6 and Control D fall outside the claimed range. Therefore, the results have only limited significance for the presently claimed subject matter. Here, Control D with the M-Ratio outside the claimed range exhibits higher (better) extrusion rate and less extrusion shrinkage. However, rebound is lower (worse), and compression set is increased (worse). These figures show the opposite trend of Table 13.

The data of both tables 10 and 13 are furthermore deficient insofar, as no comparison with a carbon furnace black in accordance with the closest prior art (N440 of D11) has been made, and because the Control
samples D and A cannot be considered as illustrative of the furnace carbon black N440.

The board concludes from the above that the available experimental evidence is inconclusive and insufficient to demonstrate an improvement of the claimed carbon blacks, in terms of extrusion rate, extrusion shrinkage, rebound and compression set, over the closest prior art.

It can be deducted, however, from the synoptic presentation of examples 1 - 8 in Table 7 that the carbon black in accordance with claim 1 of the main request, represented by example 8, achieves a relatively high extrusion rate together with a relatively low extrusion shrinkage during compound processing, when incorporated in EPDM rubber compositions.

Therefore, starting from D11 as the closest prior art, the technical problem which has actually been solved by the claimed carbon black can be seen in the provision of other furnace carbon blacks suitable for use in rubber compositions which, when incorporated in particular in EPDM compositions, lead in particular to a high extrusion rate and a low extrusion shrinkage during compound processing.

4.4 It remains to be decided whether the claimed solution is obvious in the light of the available prior art.

The respondent's main argument on this point was that furnace carbon blacks having an M - Ratio in the claimed range were known in the art. He relied on
documents D15a, D15b, D16, D16a, D17 and D17a-d as evidence for several prior uses.

Based on these alleged prior uses, the respondent asserted that furnace carbon blacks having an M - Ratio in the claimed range were usual in the art. He further pointed out that in the appellant's own submission the M-Ratio could be chosen independently from iodine number and DBP absorption. According to the respondent, there was thus no reason why the skilled person should not consider such carbon blacks as an alternative to standard carbon blacks such as N440.

The appellant has argued that the properties of the samples of SAPEX 20 would have changed in the time elapsed since their production in 1984. The values analyzed in 2002 would thus not reflect the actual properties of the furnace black produced. The opposition division had accepted this argument and dismissed the evidence of prior use as inconclusive. In appeal proceedings, the appellant has supported his argument inter alia by document D24 and declaration D26.

4.5 From the evidence and arguments the board concludes the following:

4.5.1 Prior use Gliwickie Zaklady Chemiczne "Carbochem"

According to declaration D16, a furnace carbon black designated Sapex 20 was manufactured and sold by Gliwickie Zaklady Chemiczne "Carbochem" in 1984. During the year 1984, 14198 tons of Sapex 20 were manufactured and sold to different customers. A sample (20 kg) of
that production was retained. According to the respondent, the analysis dated 7 November 2002 was carried out on the sample retained from that production. It exhibited an M - Ratio of 1.64, an I₂No. of 19.2 and a DBP absorption of 127.2 (D16a).

The board has no reason to question the veracity of the declaration D16 as regards the production and the sales of the furnace carbon black Sapex 20. Thus it considers that important amounts of this product were sold to customers before the priority date of the patent in suit. However, concerning the properties of the product sold, it is observed that the designation ZN84/MPCHOiL\-G/Ca-1/09/Sapex 20 in D15a and D15b is different from the designation in D16 (ZN-84/MPCHiL/Ca-1/09). Assuming in favour of the respondent that there is a typing mistake in D15a and D15b as argued by the respondent during oral proceedings, and that the measurements (D15a, D15b, D16a) were actually performed on the sample retained from 1984, the question arises whether the combination of properties measured on 7 November 2002, i.e. about 18 years after the production of the sample, are identical to those of the product sold in 1984. In this context documents D24 and D26 have been provided by the appellant in order to show the instability over time of the I₂ number of furnace carbon blacks.

The board finds the appellant's argument based on document D24 conclusive. Said document reports a significant instability of the iodine number observed on carbon blacks over a period of time of 12 - 14 years (see D24, Tables 1, 3 and 7). The period of time elapsed between the production of the sample of Sapex
20 (in 1984) and the date of the measurement of the I₂No. on this sample is about 18 years. The board thus considers that the actual I₂ number of the Sapex 20 product manufactured in 1984 remains unknown. For these reasons, it cannot be concluded with the required certainty that the Sapex 20 carbon black sold in 1984 had in fact the combination of characteristics reported in D16a.

4.5.2 Prior use Ashland / Degussa

According to the declaration D17, furnace carbon blacks N762, N774 and N787 were produced by Ashland / Degussa at its plant in Aransas Pass, Texas, in significant amounts of between 2500 and 5000 MT/year and sold in the years 1990 and 1991, i.e. before the priority date of the patent in suit, some of the customers being Bridgestone/Firestone, Cooper Tire and Carbogal de Portugal. As the date of production of carbon black N754 (14 September 1999) is after the priority date of the patent in suit, the alleged prior use of this product is not taken into account.

Experimental reports D17a, D17b and D17d relate to measurements on furnace carbon blacks N762, N774 and N787, respectively. The particle distribution measurements carried out on these samples on 16 July 2002 revealed M - Ratios of 1.63; 1.63; and 1.68, respectively, that is, in the claimed range. The values for the DBP absorption were determined as 64.9 cm³/100g; 69.2 cm³/100g and 78.8 cm³/100g, respectively, and thus also fall within the claimed range. The values of the I₂No. are 27.4 mg/g; 27.1 mg/g and 28.3 mg/g,
respectively, which is considerably removed from the claimed range of 51 - 62 mg/g.

However, it is not clear whether the properties indicated in D17a, b and d were measured on samples retained from the production in 1990 and 1991 or on samples available to the public in 2002, before the date of the measurement on 16 July 2002. In the first case a period of time of about eleven or twelve years would have elapsed between the date of production and the date of measurement and therefore, in view of D24 and D26, considerations similar to those indicated in point 4.5.1 above would apply likewise to this prior use as regards the iodine number instability. In the second case, the characteristics of the product sold eleven or twelve years earlier, in particular the iodine number, would be unknown. Therefore, as in the preceding prior use of Sapex 20, it cannot be concluded from the evidence on file that the furnace carbon blacks N762, N774 and N787 sold to customers in the years 1990 and 1991 actually exhibited the combination of characteristics reported in D17a, D17b and D17d, respectively.

4.6 However, even if one assumes in favour of the respondent that the carbon blacks sold by Gliwickie Zaklady Chemiczne "Carbochem" and by Ashland / Degussa had the combined characteristics as determined in experimental reports D16a and D17a, b and d, respectively, the claimed subject matter would still not derive therefrom in an obvious manner, for the following reasons.
The carbon blacks of the prior art do not immediately reveal to the public the special significance of a particular parameter, such as the M - Ratio. It should be borne in mind that the M - Ratio is not a conspicuous property of a carbon black sample, but is calculated from the shape of the Stokes particle size distribution curve after measuring the same. But even if the skilled person would have determined this parameter, the samples of the prior art would have disclosed particular M - Ratios in combination with specific values for the I₂ number which are much lower than the claimed range. Neither document D11 nor the prior use carbon blacks suggest to maintain an I₂ number and a DBP absorption value as disclosed in D11 (carbon black N440) while changing the particle size distribution of the product, expressed by its M - Ratio, and choosing M - Ratios as indicated in D16a or D17a,b,d, for the prior use carbon blacks, in order to solve the problem defined above.

4.7 D8/D8a reports values for $\Delta \text{Dst} / \text{Dst}$ (median diameter divided by mode diameter) which appears to correspond to the M - Ratio of the opposed patent. Of the seven samples analyzed, only one has an M - Ratio in the claimed range (Table 2). This sample No. 3 differs considerably from the claimed carbon black in its I₂No. of 18. Neither D11 nor D8/D8a contain information which would give the skilled person an incentive to produce a carbon black having both the I₂No. and the DBP absorption maintained at the level disclosed for N440 while changing the particle size distribution so that the M - Ratio would be equal or similar to that of sample No. 3 of D8/D8a in order to solve the problem stated above.
The other documents cited during the opposition and appeal procedures were not relied upon by the respondent during the oral proceedings. In the board's view, these documents do not contain additional relevant information which would, in combination with the documents discussed above, render the claimed subject matter obvious.

4.8 The subject matter of claim 1 of the main request is therefore based on an inventive step. Claim 2 refers back to claim 1 and is likewise allowable.

5. Since the main request can be allowed, there is no need to consider the auxiliary request.
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 in amended form on the basis of the set of claims of the main request filed during the oral proceedings, the drawings as granted and a description to be adapted.

The Registrar             The Chairman

C. Vodz                    M. Eberhard