DECISION
of 26 November 2002

Case Number: T 0315/00 - 3.5.2

Application Number: 92303472.2

Publication Number: 0512703

IPC: H01B 1/24

Language of the proceedings: EN

Title of invention:
Conductive polymer compositions

Applicant:
CABOT PLASTICS LIMITED

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56, 84

Keyword:
"Claims - clarity (main and auxiliary request 1 (no); auxiliary request 2 (yes))"
"Inventive step (auxiliary request 2 (yes))"

Decisions cited:
-

Catchword:
-
Case Number: T 0315/00 - 3.5.2

DECISION
of the Technical Board of Appeal 3.5.2
of 26 November 2002

Appellant: Cabot Plastics Limited
Gate Street
Dukinfield
Cheshire SK16 4RU (GB)

Representative: Attfield, Donald James
Barker Brettell
138 Hagley Road
Edgbaston
Birmingham B16 9PW (GB)

Decision under appeal: Decision of the Examining Division of the European Patent Office posted 15 October 1999 refusing European patent application No. 92 303 472.2 pursuant to Article 97(1) EPC.

Composition of the Board:
Chairman: W. J. L. Wheeler
Members: F. Edlinger
P. Mühlens
Summary of Facts and Submissions

I. The appeal is against the decision of the examining division refusing European patent application No. 92 303 472.2.

II. With the statement of grounds of appeal, the appellant filed new claims 1 to 8 of a main request. Claim 1 is essentially the same as claim 1 on which the decision under appeal was based, differing therefrom only by the insertion of two words (identified below by being placed in italics), and has the following wording:

"A conductive polymer composition having positive temperature coefficient characteristics comprising at least one thermoplastic polymer providing a matrix throughout which is dispersed a mixture of conductive carbon blacks, the thermoplastic polymer matrix constituting from 20 to 98 per cent by weight of the composition and the mixture constituting from 2 to 80 per cent by weight of the composition and comprising a first conductive carbon black and a second different conductive carbon black, characterised in that each of the carbon blacks has a structure level, as measured by DBP technique, of 40 to 150 cc/100g and each constitutes from 1 to 40 per cent by weight of the composition, the first carbon black comprising particles having average size in the range from 35 to 300 nm and the second carbon black comprising particles having average size in the range from 15 to 25 nm."

Claims 2 to 8 are dependent on claim 1.

III. The following documents, among others, were cited in the decision under appeal:
IV. The reasons given in the decision under appeal may be summarised as follows:

The two carbon blacks specified in claim 1 were only distinguished in (number) average particle size and could each be freely selected in a range between 1 to 40%. Since the particle size distribution of carbon blacks was often not symmetrical, as one could see from D1, the resulting mixture which was contended to have a bimodal particle size distribution, was not clearly distinguishable from that of a (single) third carbon black, especially if only a minor amount of the second carbon black (or if an additional carbon black) was employed in the mixture. Therefore, claim 1 did not comply with Article 84 EPC because it did not clearly and unambiguously specify a mixture of carbon blacks having a bimodal distribution. The applicant had failed to specify how to determine, on the basis of an analysis of the average particle size, whether the carbon black of the composition consisted of one or more fractions or types of carbon black.

Since it was impossible to decide whether a given particle size distribution was attributable to one carbon black or to a mixture of several particle size fractions of various carbon blacks, the composition of claim 1 was anticipated by several known compositions. For instance, the composition of Example 10 of D4 (columns 15 and 16) comprising a carbon black with an
average particle size of 28 nm (with percentages per weight and with DBP values as specified in the claimed ranges) would anticipate a mixture covered by the present claim 1, e.g. when the mixture consisted of a first carbon black of 25 nm and a minor amount of a second carbon black of 35 nm. Also D5 (Table II) disclosed thermoplastic PTC compositions comprising various types of carbon black which were not distinguishable from compositions as claimed.

Even if it were conceded that claim 1 specified compositions containing two distinguishable fractions of carbon black, claim 1 could not be allowed because its subject-matter did not involve an inventive step. Since D4 (column 4, line 61) disclosed the possibility of using mixtures of carbon blacks for obtaining the desired combination of average physical properties, a selection of two or more carbon blacks as claimed was obvious for optimising the physical characteristics of the carbon black known to be necessary for obtaining an intense PTC effect. There was no convincing evidence that the (undisputed) intense PTC effect shown in Figure 1 of the present application was attributable to the fact that two carbon blacks were mixed because the carbon blacks used in the embodiment had further distinguishing features (other than average particle sizes) and the selection of a carbon black with large particle size and a low value of S/D (surface to particle size ratio) would also explain this effect, as explained at the bottom of column 4 in D4.

V. Oral proceedings were held before the Board on 26 November 2002. Two sets of claims according to auxiliary requests 1 and 2 and new pages of the description with adaptations to the amended claims of
auxiliary request 2 were filed in the oral proceedings.

VI. Claim 1 of the auxiliary request 1 is worded as follows:

"A conductive polymer composition having positive temperature coefficient characteristics comprising at least one thermoplastic polymer providing a matrix throughout which is dispersed two conductive carbon blacks, the thermoplastic polymer matrix constituting from 20 to 98 percent by weight of the composition and the two carbon blacks constituting from 2 to 80 percent by weight of composition and comprising a first conductive carbon black and a second different conductive carbon black, characterized in that each of the carbon blacks has a structural level, as measured by DBP technique, of 41 to 120 cc/100g and each constitutes from 1 to 40 percent by weight of the composition, the first carbon black having a BET surface area of from 7 to 42 m\(^2\)/g and the second carbon black having a BET surface area of from 140 to 230 m\(^2\)/g the first carbon black comprising particles having average size in the range of 41 to 148 nm and the second carbon black comprising particles having average size in the range from 15 to 20 nm."

Claims 2 to 8 are dependent on claim 1.

VII. Claim 1 of the auxiliary request 2 is worded as follows:

"A process for preparing a conductive polymer composition having positive temperature coefficient characteristics comprising at least one thermoplastic polymer providing a matrix throughout which is
dispersed a mixture of conductive carbon blacks, by admixing on a conventional mixing machine the thermoplastic polymer matrix in amounts from 20 to 98 per cent by weight of the composition and the mixture of conductive carbon blacks in amounts from 2 to 80 per cent by weight of the composition, the mixture comprising a first conductive carbon black and a second different conductive carbon black, characterised in that each of the carbon blacks has a structure level, as measured by DBP technique, of 40 to 150 cc/100g and each constitutes from 1 to 40 per cent by weight of the composition, the first carbon black comprising particles having average size in the range from 35 to 300 nm and the second carbon black comprising particles having average size in the range from 15 to 25 nm."

Claims 2 to 6 are dependent on claim 1.

VIII. The appellant essentially argued as follows:

A mixture of two different carbon blacks as specified in claim 1 of the main and first auxiliary requests was clearly distinguishable from any previously known carbon black. If a first carbon black was mixed with a second carbon black which was different with respect to the average size of the particles, the resulting mixture was evidently different from both. It could be demonstrated by usual computer analysis for determining particle size distributions that the mixture was different from each of the admixed carbon blacks and from any known carbon black grade, eg with respect to the average size and the relative frequency of particle sizes. These differences were even more pronounced in the case of claim 1 of auxiliary request 1 where the average size ranges of the different carbon blacks were
spaced further apart from each other.

The mixture of carbon blacks with the parameters specified in claim 1 of any of the requests brought about two marked improvements. On the one hand, control of the critical temperature was made easier with the mixture. Table 3 of the application showed that this temperature significantly changed with the percentage by weight of the two different carbon blacks which were admixed. A comparison with a single carbon black with a similar percentage by weight as disclosed in D4, Table II, Examples 6, 7 and 10 (35% carbon black) and Examples 49 and 50 (15% carbon black) showed that this change was much more pronounced when two different carbon blacks were admixed. On the other hand, the influence of the melting point of the polymer matrix on the critical temperature was much less pronounced with a mixture of carbon blacks of the present application as can be seen by comparing Table 5 of the application with the examples of D4. The temperature values $T_{2x}$ at which the resistivity was twice the resistivity at 20°C were about 43% below the melting point (88°C) and were all at or below 50°C in the present application. By contrast, they were only around 18% on an average below the melting point (135°C) in the examples of D4 and only two of the 95 examples in D4, Examples 74 and 75, had a value $T_{2x}$ of as low as 50°C.

D4 did not disclose specific examples of mixtures but only contained a general statement that mixtures of carbon blacks could be used (D4, column 4, line 61). Although D4 gave him no clear incentive to use a mixture in any of the 95 examples, a person skilled in the art would have rather used mixtures, if any, of grades having similar characteristics. There was no
teaching in D4 suggesting that a mixture of two
different carbon blacks as specified in claim 1 of any
of the requests would achieve the benefits exhibited by
the compositions of the present invention. The
theorising, in the decision under appeal, about
possible other causes of the effect achieved by the
present invention was pure supposition. Although the
S/D ratios of the carbon blacks listed in Table 1 of
the present application fell within the range of S/D
ratios of the 95 examples provided in Table 1 of D4,
the compositions of the present application showed a
more pronounced PTC effect than any of the examples in
D4. This effect was attributable to the mixture of two
different carbon blacks as specified in claim 1 of any
of the present requests.

D5 did not disclose or suggest a mixture of two
different carbon blacks and could not, therefore,
render the subject-matter of the present claim 1
obvious.

IX. The appellant requested that the decision under appeal
be set aside and that a patent be granted on the basis of:

- claims 1 to 8 filed with the grounds of appeal
  (main request)

- claims 1 to 8 of auxiliary request 1 filed in the
  oral proceedings

- claims 1 to 6 of auxiliary request 2 filed in the
  oral proceedings;

description, pages 2 to 6 with insert A on page 2
as filed in the oral proceedings;

drawings, Figures 1 to 4 as originally filed.

Reasons for the Decision

1. The appeal is admissible.

2. Main request and auxiliary request 1

2.1 It follows from Article 84 EPC that the claims shall be clear in respect of the matter for which protection is sought. Claim 1 of the main request relates to a conductive polymer composition, i.e. a product, comprising inter alia a thermoplastic polymer matrix throughout which is dispersed a mixture comprising a first conductive carbon black and a second different conductive carbon black. To be clear, the "mixture" of carbon blacks as a characteristic of the product must firstly be distinguishable from a non-mixture, i.e. a carbon black having a known (monomodal) distribution of particle sizes (see e.g. D1, Figure 5). Secondly, at least one parameter characterising the mixture must be clearly defined if protection is sought for a specific mixture.

2.2 As admitted by the appellant in the oral proceedings, the two carbon blacks which are dispersed in the mixture according to claim 1 of the main request may be "different" only in that the first carbon black comprises particles having average size in the range from 35 to 300 nm and the second carbon black comprises particles having average size in the range from 15 to 25 nm. Average size can be determined by standard
tests (see eg page 3, lines 36 to 42 of the present application) and thus constitutes a verifiable parameter for each carbon black. However, neither the average size nor a structure level as specified in claim 1 (before mixture) characterises a particular particle size distribution. But it is generally known that particle size distributions may take various forms, eg a narrow or broad and more or less symmetrical size distribution (D1, Figure 5). Each of the first and second carbon blacks will include particles of a size below and above the average size. Although neither the frequency distribution nor the lower and upper limits of the particles sizes are specified, the first and second carbon blacks would still be distinguishable from each other (and from other known types of carbon black) in respect of their average sizes. However, a considerable overlap of the particle size frequency distribution curves is to be expected.

2.3 When the first and second carbon blacks are mixed and dispersed throughout the polymer matrix and "each constitutes from 1 to 40 per cent by weight of the composition" (claim 1 of the main request), a new particle size distribution and a new average size may be determined for the mixture according to the same standard tests. The new average particle size will normally be different from, and have a value in between those of, the first and second carbon blacks. Nevertheless, the mixture may not be distinguishable from a non-mixture having the same average particle size because neither the particle size distribution of the individual admixed carbon blacks nor that of the mixture is specified. Some embodiments covered by claim 1 would certainly show a bimodal distribution
with local maxima of the frequency near the average sizes of the first and second carbon blacks, eg if approximately equal amounts of first and second carbon blacks having narrow particle size frequency distribution curves were admixed. However, this is not the case for the whole range of the product for which protection is sought because claim 1 of the main request covers a mixture of widely different amounts of two carbon blacks with any arbitrary particle size distribution, eg 1 percent by weight of the one and 40 percent by weight of the other carbon black. It is therefore not clear how such a mixture of carbon blacks could be distinguished from a non-mixture. Moreover, since the particle size frequency distribution of the mixture could only be calculated if those of the first and second carbon blacks were defined (and did not change in the mixing and preparation steps), there is no parameter specified in the claim which clearly characterises the particle size characteristics of the mixture obtained in this way.

2.4 Claim 1 of the auxiliary request 1 specifies narrower ranges of the average sizes of the first and second carbon blacks and a narrower range of structural level. In addition, different ranges of BET surface areas are specified for the two carbon blacks. Although the two average sizes are now separated by a larger gap (maximum average size 20 nm and minimum average size 41 nm), claim 1 has qualitatively the same defects as claim 1 of the main request because the large range (1 to 40%) of the amounts of the two carbon blacks and their unspecified particle size distributions render the characteristics of the mixture unclear. The specification of the BET surface areas gives an indication of the chemically active surface due eg to
surface roughness and porosity of the particles (cf D1, right-hand column below Figure 5). These additional features cannot remove the above lack of clarity concerning the specification of the particle size characteristics of the mixture. Furthermore, the BET surface area of the mixture is not clearly specified by indicating those of the individual admixed carbon blacks of widely different amounts.

2.5 Therefore, claim 1 of both the main request and the auxiliary request 1 do not comply with the requirement of Article 84 EPC.

3. Auxiliary request 2

3.1 Claim 1 of auxiliary request 2 relates to a process for preparing a conductive polymer composition. The amendments of claim 1, in particular the process step of admixing the thermoplastic polymer matrix and the two different carbon blacks, are disclosed in the application as filed (eg page 4, lines 5 to 12). The description has been adapted to the amended claims. The amendments thus do not infringe Article 123(2) EPC.

3.2 The process specified in claim 1 of the auxiliary request 2 does not suffer the same deficiency as the definition of the product because the structural level and the different average sizes of the carbon blacks which have to be mixed in the preparation process to obtain the specified mixture are sufficiently clearly defined. Although a large variety of first and second carbon blacks may be used, it is however clear which carbon blacks may be used, in the preparation process, as the first and the second carbon black and which may not (see point 2.2 above).
3.3 D4 was considered, in the decision under appeal, as reflecting the closest prior art mentioning the possibility of using mixtures of carbon blacks (D4, column 4, line 61). According to the teaching of D4 (eg column 2, lines 25 to 66), the surface area (S), particle size (D) and ratio by volume of the filler (mixture) to the polymer should be such that a quantity calculated from these parameters was less than one, as set out in claim 1 of D4. None of the ninety-five examples of D4 discloses a specific mixture. If a person skilled in the art chose to optimise the physical characteristics of carbon blacks in accordance with the teaching of D4, the particle size would have to be chosen dependent on the surface area and filler to polymer ratio so that the above condition was fulfilled. There is no indication in D4 that a person skilled in the art would mix two carbon blacks having different average sizes, each having a structure level in the range as specified in claim 1 of the auxiliary request 2.

3.4 The appellant has argued that a process for preparing the composition as set out in claim 1 of the auxiliary request 2 made it easier to obtain a more pronounced PTC effect and a better control of the critical temperature of the PTC composition. Although other parameters, such as the surface area or the value of the ratio of the surface area to the particle diameter (D4, column 4, lines 61 to 65), may have a considerable influence on the temperature dependent resistivity, it appears plausible and supported by the examples of the application that such a technical effect may be achieved by mixing the selected types of carbon black because smaller particles could fill interstices between larger particles in the thermoplastic matrix.
3.5 None of the other prior art documents available in the file discloses or suggests the mixing of two carbon blacks as specified in claim 1 of the auxiliary request 2. D5, referred to in the decision under appeal as disclosing compositions which were not distinguishable from the mixture, does not give any hint at preparing a specific mixture of different carbon blacks. The subject-matter of claim 1 of the auxiliary request 2 shall thus be considered as involving an inventive step (Article 56 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 grant a patent on the basis of:

   claims 1 to 6 of auxiliary request 2 filed in the oral proceedings;

   description, pages 2 to 6 with insert A on page 2 as filed in the oral proceedings;

   drawings, Figures 1 to 4 as originally filed.

The Registrar: The Chairman