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DECISION
of 10 January 1996

Case Number: T 0814/93 - 3.3.3

Application Number: 88102914.4

Publication Number: 0280325

IPC: C08G 75/20

Language of the proceedings: EN

Title of invention:
Melt stabilization of poly(arylene sulfide sulfone)s

Applicant:
PHILLIPS PETROLEUM COMPANY

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56, 123

Keyword:
"Novelty - (yes - disclaimer)"
"Inventive step - (yes - specific polymer non-obvious over generic formula)"

Decisions cited:
-

Catchword:
-

Case Number: T 0814/93 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 10 January 1996

Appellant:

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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 31 March 1993 refusing European patent application No. 88 102 914 .4 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: C. Gérardin
Members: P. Kitzmantel
J. A. Stephens-Ofner

Summary of Facts and Submissions

I. This appeal, which was filed on 9 June 1993, lies against the decision of the Examining Division announced orally on 2 March 1993, with written reasons posted on 31 March 1993, refusing European patent application No. 88 102 914.4 in the name of Phillips Petroleum Company filed on 26 February 1988, claiming a US-priority of 27 February 1987, and published under No. 280 325. The appeal fee was paid together with the Notice of Appeal and a Statement of Grounds of Appeal was filed on 9 August 1993.

II. The decision under appeal was based on a set of 31 claims filed on 2 March 1993, independent Claims 1, 21, 26 and 31 reading as follows:

"1. A method for preparing a melt stabilized polymer consisting of a poly(arylene sulfide sulfone), a melt flow stabilizer, and optionally at least one additional component selected from pigments, fillers, extenders, plasticizers, stabilizers and strengtheners, characterized by contacting a poly(arylene sulfide sulfone) homopolymer with a zinc source as said melt flow stabilizer in the absence of sulfur to obtain a zinc cations-containing poly(arylene sulfide sulfone) homopolymer having improved melt stability."

"21. A method for preparing a polymer having improved melt stability consisting of a poly(arylene sulfide sulfone) and a melt flow stabilizer, and optionally at least one additional component selected from pigments, fillers, extenders, plasticizers, stabilizers and strengtheners, characterized by contacting in a polar organic solvent a polyhaloaromatic sulfone monomer, an alkali metal sulfide and a soluble zinc source as said melt flow stabilizer in the absence of sulfur under polymerization conditions to obtain a homopolymer having incorporated zinc cations."

"26. A melt stable polymer composition consisting of a poly(arylene sulfide sulfone), a melt flow stabilizer and optionally at least one additional component selected from pigments, fillers, extenders, plasticizers, stabilizers and strengtheners, characterized by a content of zinc cations in the absence of sulfur as said melt flow stabilizer and in that said poly(arylene sulfide sulfone) is a homopolymer."

"31. A method for improving the melt stability of poly(arylene sulfide sulfones), characterized by incorporating zinc cations in the absence of sulfur into said poly(arylene sulfide sulfones), wherein said poly(arylene sulfide sulfones) are homopolymers."

Claims 2 to 20, 22 to 25 and 27 to 30 were dependent, respectively, on Claim 1, Claims 21 and 26.

III. The decision under appeal held that by the exclusion of the presence of sulfur according to independent Claims 1, 21, 26 and 31 their subject-matter was rendered novel over

D1: US-A-3 725 362.

However, these claims were not inventive having regard to

D2: DE-A-3 143 992,

which disclosed the melt flow stabilization of arylene sulfide polymers (hereinafter PAS) by their treatment, in the absence of oxygen, with water soluble chemicals, including zinc nitrate. It would have been obvious to apply this teaching to poly(arylene sulfide sulfones) (hereinafter PASS), which belonged generically to the class of PAS, as confirmed by

D3: US-A-3 354 129 and

D4: Comprehensive Polymer Science, vol. 5, 1989, Pergamon Press, pages 558 to 559.

Without experimental evidence of a different melt flow behaviour of PAS and PASS these polymers were considered to be equivalent in this respect.

IV. Against this decision the Appellant argued that, with respect to melt flow stabilization, D2 referred only to partially cured, and not to uncured PAS. Furthermore, although belonging to the generic class of PAS, with regard to their melt flow stability, PASS behaved differently from other members of this class. In support of this contention he referred to

D5: US-A-4 301 274

which demonstrated that PASS suffered from poor melt flow stability. Moreover, the Appellant submitted an experimental report (Scoggins Report) comparing the melt flow behaviour of uncured and cured poly(phenylene sulfide) (hereinafter PPS) and poly(phenylene sulfide sulfone) (hereinafter PPSS) after addition of, on the one hand sodium hydroxide, and on the other hand zinc acetate. In his opinion, the Scoggins Report showed that sodium hydroxide, a preferred melt flow stabilizer according to D2, was ineffective in both, PPS and PPSS, while zinc acetate performed satisfactorily in PPSS, but was only moderately effective in PPS.

V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 31 as filed on 2 March 1993.

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments*

Apart from purely editorial amendments the following features have been incorporated into Claim 1 which did not appear in its original version:

(i) "melt flow stabilizer" (twice): basis on page 2, lines 17 to 34, page 5, lines 26 to 29 and page 6, line 33 to page 7, line 1 of the original application;

(ii) "in the absence of sulfur": disclaimer establishing novelty over D1; see particularly Claims 1, 4 and 5 of D1, according to which disclosure D1 comprises PASS compositions (G = SO_2 in Claim 4, formula V) containing as a melt flow modifier sulfur and zinc oxide;

(iii) "optionally at least one additional component selected from pigments, fillers, extenders, plasticizers, stabilizers and strengtheners": basis on page 10, lines 27 to 30 and page 11, lines 1 to 4 of the original application;

(iv) "homopolymer": preferred polymer according to page 11, lines 5 to 6 of the original application.

Independent Claims 21, 26 and 31 are based, respectively, on original Claims 17, 21 and 26 and comprise the same amendments as Claim 1 (except for Claim 26 which does not comprise feature (iii) as set out above).

Dependent Claims 2 to 20 correspond to original Claims 2 to 4 (4 split into new Claims 4 to 6), 5 (split into new Claims 7 and 8), 6 (split into new Claims 9 and 10) and 7 to 16.

Dependent Claims 22 to 25 correspond to original Claims 18, 19 (19 split into new Claims 23 and 24) and 20.

Dependent Claim 27 is based on a combination of features of original Claim 22 and of the statement on page 11, lines 1 to 4 of the original application, and dependent Claims 28 to 30 correspond to original Claims 23 to 25.

The amendments meet, therefore, the requirements of Article 123(2) EPC.

3. *Novelty*

3.1 D1 discloses a process for decreasing the melt flow of PAS by heating the polymer in the presence of a melt flow modifier system comprising sulfur and a melt flow modifier rate accelerator such as zinc oxide (Abstract; Claims 1, 2 and 5; column 1, line 37 to column 3, line 6). According to the definition of the unit G in formula V (Claim 4; column 2, lines 25 to 60) and the disclosure of 4-bromophenyl-3-n-butyl-4-chlorophenyl sulfone as starting compound (column 4, line 38) PASS is within the meaning of the generic term "PAS" as used in D1. However, since according to this citation the presence of sulfur is essential for achieving the desired decrease of the melt flow (Claim 1; column 5, lines 25 to 27; column 6, Example 1, lines 58 to 61; column 7, Example II, lines 4 to 7), by disclaiming the presence of sulfur novelty of the subject-matter of the otherwise anticipated Claims 1, 21, 26 and 31 over D1 has been established.

3.2 According to one of its two embodiments D2 teaches that the melt flow behaviour of at least partially cured PAS may be stabilized by heating the polymer, preferably in the essential absence of oxygen, in the presence of water and a water soluble substance, possibly a group IA, II or IIIA metal salt (Claims 1 and 5; page 7, line 33 to page 8, line 3; page 8, lines 21 to 35). Among a considerable number of alternatives for the water soluble substance zinc nitrate is mentioned (page 7, line 13). At page 5, line 26 to page 6, line 23 of D2 the PAS to be used is specified by its melt index range and by reference to its preferred method of preparation from polyhaloaromatic compounds, alkali sulfides and organic amides. PPS is indicated as a typical representative and used in all worked examples. There is no explicit or implicit disclosure in D2 of the possible use of PASS. Therefore, D2 does not anticipate the subject-matter of present independent Claims 1, 21, 26 and 31.

4. *Inventive step*

4.1 D1 requires the presence of sulfur as an ingredient of the melt flow modifier for PAS, which is not needed by and must not be present according to the present invention. This document is not, therefore, an appropriate starting point for the development of the presently claimed method of the melt stabilization of PASS.

4.2.1 The manner of treatment of PAS disclosed in D2 (see Section 3.2 above) corresponds to the treatment of PASS carried out according to the present invention. Since PASS belongs generically to the class of PAS polymers, this citation is highly relevant for the assessment of the obviousness of the subject-matter now being claimed.

4.2.2 The problem to be solved by the present invention vis-à-vis D2 was the adaptation to PASS of the process for an improvement of the melt flow stability of PAS disclosed in D2.

According to the independent process Claims 1, 21 and 31 and the independent product Claim 26 this problem can be solved by incorporation of zinc cations into the PASS.

On the basis of the information contained in Tables I, III and IV of the application in suit the Board is satisfied that the above problem has been effectively solved by the measures set out in the independent claims.

For the assessment of an inventive step, the crucial question is therefore whether the skilled person would have expected that zinc salts, like the nitrate exemplified in D2, would be able to act as melt flow stabilizers for PASS.

4.2.3 D2 relates to PAS, which polymer class is characterized by repeating units of the structure "arylene-sulfide-arylene". Since PASS comprises such units it can be considered to belong to the class of PAS polymers. This is confirmed by Tables 12 and 13 of D4, which list a number of PAS polymers, among them PASS.

While D2 contains no express mention of PASS, it points at page 6, line 1 to document D3 by saying that therein "a suitable process [for the preparation of PAS] was described". From this statement it cannot, however, be reasonably inferred that with regard to their melt flow behaviour all PAS species coming under the broad definition set out in D3 (see Claim 10) would behave in an equivalent way. On the contrary, the skilled person would, on the basis of his common general knowledge, not expect the same thermal stability from aromatic polymers having bonding units as different in their thermodynamic stability as -O-, -S-, -CO- and SO_2-, neither would he, in view of the different chemical reactivity of these units, expect a similar behaviour towards the chemicals enumerated in Claim 1 of D2; even less would he expect similar results with respect to the melt flow stabilization of polymers containing such units. The different character of different PAS polymers is also emphasized by their very differing physical properties (softening temperature, crystallinity, solubility) as disclosed in D4, Tables 12 and 13).

Moreover, from the information contained in D2 it is impossible to draw any conclusions as to the mechanism leading to the desired melt flow stabilization, from which it could then be deduced that - under the prevailing circumstances - the differences between those units would be irrelevant to the desired effect. Indeed the only relevant concrete information in D2, which is contained in Example 2 (page 13, line 28 to page 16, line 2; particularly Table II), is rather confusing: according to Experiment 21 by treatment of PPS (partially cured by oxygen) with NaOH the melt index after 5 min at 316°C (load 5 kg) is considerably reduced (from 232g to 87g), but increased again by 15% after 30 min; according to Experiment 22 the melt index of the same PPS, when treated with $NaHCO_3$, decreased after 5 min to only 105g, but decreased further by 37% after 30 min. These results demonstrate that chemically rather similar compounds (NaOH and $NaHCO_3$) may affect the melt flow of PPS in a quite different way. Any forecast of the effect of a chemically much more different compound (like zinc nitrate) would therefore be impossible.

This conclusion is reinforced by a comparison of Experiments 23 and 26 in said Table II. In these experiments a PPS was used that was partially cured by oxygen in the presence of $Na_2CO_3.H_2O$. This starting material showed a melt index change (difference between melt indices after 5 min and after 30 min) of 13%. After treatment with NaOH in order to "stabilize the melt flow" (Experiment 26) the melt flow change **increased** to 16%, indicating a **worsening** of the melt flow stability. Only the treatment of the same starting PPS with acetic acid and calcium acetate (Experiments 24 and 25) brought about the desired improvement of the melt index change (reduction to 1% and 3,3%, respectively). These data show that the improvement of the melt index stability depends not only on the kind of stabilizer used, but also on the curing history of the starting PPS.

One may therefore conclude that according to D2 zinc nitrate was just a possible candidate as PAS melt index stabilizer, whose actual effectivity was, however, completely unknown and furthermore dependent on the precuring treatment of the starting PAS. On the basis of the information contained in D2 any assumption of the effectivity of zinc compounds as melt flow stabilizers for PASS was therefore purely speculative.

Even for the preferred PAS polymer, i.e. PPS, D2 does not offer any rules for achieving success (effective melt flow stabilization) in a systematic way. The skilled person wishing to apply the teaching disclosed in D2 for PPS to other related polymers (like PASS) would therefore be left without any, let alone any reliable guidance. Since the number of chemicals proposed as melt flow stabilizers in D2 is very high, and since zinc nitrate is not prominent among them, it would have involved quite a large number of experiments to find out that, under certain conditions, zinc cations are effective in stabilizing the melt flow of PASS. Such a time-consuming, random procedure is beyond the concept of obviousness.

The solution of the existing problem by incorporation of zinc cations into PASS was therefore non-obvious over the disclosure in D2.

4.2.4 In the appealed decision it was held that Example II of the application in suit was inappropriate to support the Appellant's contention that different types of PAS would react differently with the same melt flow stabilizers, because, in contrast to the "inventive" examples, in this comparative Example II the PAS used, i.e. poly(phenylene sulfide ketone) (hereinafter PPSK), had been pre-treated with NaOH before the treatment with zinc acetate.

While it is true that for the indicated reason Example II does not provide a basis for a correct comparison of PPSK and PPSS, it nevertheless shows that under the conditions described in D2 zinc acetate is essentially ineffective as melt flow stabilizer for PPSK: according to the first experiment in Table II, which is a control experiment without after-treatment with metal salt, the melt index decreases by 85% from 88.70 g (after 5 min) to 13.33 g (after 15 min); according to the third experiment, with NaOH/zinc acetate after-treatment, the melt index decreases by 83,8% from 72.02g to 11.7g.

Since, according to Table II of D2, from the three series of experiments (20-22; 23-26; and 27-29) the latter two work with Na_2CO_3 pre-treated PPS, and since according to Claim 3 of D2 Na_2CO_3 and NaOH are equally preferred agents, the conditions used in Example II of the application in suit are fully in line with one of the preferred embodiments of D2.

Thus, said Example II shows that at least under otherwise favourable conditions zinc acetate is not an effective melt flow stabilizer for PPSK.

4.2.5 Even if, despite of what is set out in Section 4.2.3 above, one would assume that PASS is a polymer close enough to PPS to make its use in the melt stabilization process according to D2 prima facie obvious to one skilled in the art, the data submitted in Table I of the Scoggins Declaration attached to Appellant's letter of 9 December 1993 demonstrate that such assumption is at variance with the facts. Table I shows that NaOH, one of the preferred melt flow stabilizing agents of D2, is - under the chosen conditions - ineffective as melt flow stabilizer as well in PPS as in PPSS, both uncured and partially cured: compare Experiment 1 with 6 and 7, on the one hand, and Experiment 8 with 12 and 13, on the other hand. Table I shows furthermore that zinc acetate causes a continuous increase of the melt flow of uncured PPS (compare Experiments 1 and 3) and a decrease of the melt flow of partially cured PPS (compare Experiments 1 and 5); in both cases the numerical change of the melt flow (5 min/30 min) was about 20% (-19.8% in the case of Experiment 3; +23% in the case of Experiment 5). As compared therewith the treatment of uncured and partially cured PPSS with zinc acetate does not considerably affect the melt flow after 5 min and provides good melt flow stability (Experiment 9: change in melt flow of uncured PPSS (5 min/30 min) was 8.0%; Experiment 11: change in melt flow of partially cured PPS was 2.8%).

These data show in the first place that, because of the low melt flow stabilities of both PPS and PPSS obtained by the use of NaOH, one of the preferred treatment agents according to D2, there was no incentive for the notional skilled person, wanting to improve the melt flow stability of PASS to pursue the route suggested by D2. Since in the case of PPS the melt flow increases from 5 min to 30 min (change -57.2% and -64.6%: see Experiments 6 and 7), while in the case of PPSS the melt flow decreases (change +63.6 and +80.0%: see Experiments 12 and 13), there was also no reason for the skilled person to assume an analogous reactivity of NaOH with PPS and PPSS.

In the second place, if the skilled person had nevertheless found it worthwhile to test the suitability of a zinc salt (here zinc acetate) as melt flow stabilizer, he would not only have discovered its low melt flow stabilizing effectivity in PPS, but also its very considerable depolymerizing effect on uncured PPS and its curing effect on partially cured PPS. This behaviour cannot be regarded as an incentive for testing the melt flow stabilizing effect of zinc salts in PASS. The fact that zinc salts do indeed provide a considerable melt flow stabilizing effect in both, uncured and partially cured PASS, shows only that PAS and PASS cannot be considered to belong, with regard to their melt flow behaviour, to the same class of polymers.

4.2.6 It was argued by the Appellant that under their normal processing conditions the melt flow stability of PASS was worse than that of PAS, because the latter would be able to be processed at much lower temperatures (see page 1, line 14 to page 2, line 2 of the original application). This assertion, which was meant to emphasize that there was a considerable qualitative difference between the melt flow stability of PAS and PASS, was not backed up by literature, nor has the Appellant himself provided relevant experimental evidence in support of this assertion. This argument cannot, therefore, be taken into account in an objective assessment of inventive step.

D5, which has been cited by the Appellant in this context, confirms, however, that the lacking melt flow stability of PASS was known in the art. According to this citation (see Abstract; Claims 1, 2) the melt flow stability of PASS can be improved by end-capping of the polymer with dihaloaromatic sulfone.

This is a solution to the problem underlying the application in suit which is completely different from that of the present invention, i.e. the incorporation of zinc cations. D5 cannot, therefore, contribute anything to the question of its obviousness or not.

4.2.7 The subject-matter of independent Claims 1, 21, 26 and 31 therefore involves an inventive step.

In view of their appendancy the same conclusion applies also to the subject-matter of the dependent claims.

5. The reasons in the appealed decision do not, therefore, prejudice the grant of a patent on the basis of Claims 1 to 31. The amended description complies with the requirements of the EPC, particularly Article 84 and Rule 27(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 grant a patent with the following claims and the following amended description:

   Claims: No. 1 to 31 as filed on 2 March 1993;

   Description: Pages 3 to 4, 8 to 10, 13 and 17 as originally filed.
   Pages 1 and 1a as filed on 1 December 1995.
   Pages 2, 5 to 7, 11, 12, 14 to 16 and 18 as filed on 1 April 92.

The Registrar:

E. Görgmaier

The Chairman:

C. Gérardin

C. Gérardin