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D E C I S I O N
of 16 May 1995

Case Number: T 0646/91 - 3.3.3

Application Number: 83105506.6

Publication Number: 0096384

IPC: C08G 75/02

Language of the proceedings: EN

Title of invention:

Recovering granular poly(arylene sulfide) particles from a poly(arylene sulfide) reaction mixture

Patentee:

PHILLIPS PETROLEUM COMPANY

Opponent:

Celanese Engineering Resins, Inc.
HOECHST AKTIENGESELLSCHAFT
Kureha Kagaku Kogyo Kabushiki Kaisha

Headword:

-

Relevant legal provisions:

EPC Art. 54, 56, 100(a), 107

Keyword:

"Novelty (yes) - no implicit disclosure"
"Inventive step (yes) - no incentive"
"Change of the specific ground of opposition within Article 100(a) EPC by a party as of right - allowable"

Decisions cited:

G 0009/92, G 0004/93

Catchword:

-



Case Number: T 0646/91 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 16 May 1995

Appellant:
(Opponent 03)

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Decision under appeal: Interlocutory decision of the Opposition Division of the European Patent Office dated 23 April 1991, posted on 26 July 1991 concerning maintenance of European patent No. 0 096 384 in amended form.

Composition of the Board:

Chairman: C. Gérardin
Members: H. H. Fessel
W. M. Schar

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 096 384 in respect of European patent application No. 83 105 506.6 filed on 3 June 1983 claiming a US priority of 4 June 1982 (US 385 223) was announced on 6 May 1987 (cf. Bulletin 87/19).

II. Three Notices of Opposition were filed on 4 February 1988 by Celanese Engineering Resins Inc. (Opponents 1) and on 5 February by Hoechst Aktiengesellschaft (Opponents 2) and Kureha Kagaku Kogyo Kabushiki Kaisha (Opponents 3), respectively, alleging lack of novelty and of inventive step (Article 100(a) EPC) as well as insufficiency of disclosure (Article 100(b)EPC). The oppositions were supported inter alia by the following documents:

- (1) US-A-3 354 129;
- (2) US-A-3 919 177;
- (3) US-A-4 116 947;
- (4) US-A-3 308 211; and
- (8) US-A-4 071 509.

III. By an interlocutory decision given at the end of Oral Proceedings on 23 April 1991 and issued on 26 July 1991 the Opposition Division held that there were no grounds of opposition to the maintenance of the patent in an amended form on the basis of a set of 11 claims, of which Claim 1 was submitted during oral proceedings and Claims 2 to 11 had been received on 19 December 1988. The only independent Claims 1 and 11 read as follows:

"1. A method for recovery of granular poly(arylene sulfide) from a polymerization reaction mixture comprising polar organic solvent, poly(arylene sulfide)

and alkali metal halide by-product, said reaction mixture being at a temperature above that at which poly(arylene sulfide) is in molten phase characterized by said method being carried out by adding said polymerization reaction mixture with an amount of a separation agent being soluble in said polar solvent but not being itself a solvent for said poly(arylene sulfide), to cause a phase separation of said molten poly(arylene sulfide) from said polar organic solvent, and subsequently reducing the temperature of said separated phases to produce a slurry comprising particulate poly(arylene sulfide) in polar organic solvent.

11. The use of poly(arylene sulfide) obtained by any of claims 1 to 10 for making shaped articles, in particular in the form of fibres, films or sheets."

The other claims are directed to preferred methods according to Claim 1.

In that decision it was first stated that the requirements of Article 83 EPC were met, since the description, especially the examples, gave detailed information how to carry out the process as defined in Claim 1. Novelty was no longer a matter of dispute between the parties. An inventive step could not be denied, since the combined advantages obtainable by the claimed process - granular poly(arylene sulfide) (PAS hereinafter) having both enlarged particle size and lower ash content - could not be expected from the documents relied upon by the Opponents.

IV. An appeal was lodged on 23 August 1991 together with payment of the prescribed fee by the Appellants Kureha Kagaku Kogyo Kabushiki Kaisha (Opponents 3). The

Statement of Grounds of Appeal was filed on 28 October 1991.

i) In this statement and in subsequent submissions the Appellants disputed that the process as defined in Claim 1 involved any inventive step. They argued that the technical problem had been shifted from its original formulation, which was the definition of a process for the preparation of granular or particulate PAS, to a more ambitious one, which was the definition of a process for the recovery of a product having simultaneously larger particle size and lower ash content. That new technical problem had already been solved in the prior art, as evident from the following additional documents:

- (22) US-A-4 025 496;
- (23) US-A-3 697 487 (a document already cited in the Search Report);
- (24) US-A-3 865 794;
- (25) New Techn. Coll. on Particulate Process (1974) pages 289 to 291 (with translation); and
- (26) Jap. Patent Appln. No. 47-39233 (1972) also with translation into English.

ii) It appeared in particular from documents (22), (23) and (24) that it was known to add water to PAS polymerisation reaction mixture; although there was no explicit mention of a phase separation in these citations, it was reasonable to assume that such separation actually occurred and that the same benefits in terms of particle size and ash content as in the patent in suit were obtained.

iii) A reproduction of Example 28 of document (2) and Example 20 of document (3) showed that the ash content was not controlled by the phase separation, but largely by the washing process.

V. Opponents 2, a party as of right to the appeal proceedings, submitted on 13 April 1995 a further document:

(27) R. Gabler und J. Studinka, Neue Polyphenylensulfone-Reaktionen an festen Polymeren, Chimia 28 (1974), pages 567 to 575.

They contended that a combination of that reference, which disclosed the use of water for the work-up of polyarylene thioether, with the process according to document (1) or (2) rendered obvious the claimed subject-matter.

VI. In their written submissions the Respondents (Patentees) first objected to the filing of several documents at such late stage. In substance, they relied on the arguments presented before the Opposition Division that nowhere in the prior art there was mention of a PAS reaction mixture separating into two organic phases by adding water, whereby a polymer having improved particle size and lower ash content could be obtained. The experiments carried out by the Appellants were not a true replication and, consequently, could not cast any doubt on the validity of the beneficial effects observed by the Patentees.

VII. Oral Proceedings were held on 16 May 1995. From Opponents 1 and 2, although duly summoned as parties as of right to the procedure, only Opponents 2 attended the hearing.

i) First, Opponents 2 raised the issue of novelty on the basis of documents (2), (3) and (22), which all mentioned the presence of water during the final recovery stage of PAS. In their reply, the Respondents referred to the wording of Claim 1, which required (1) water to be added to cause phase

- separation, followed by (2) a temperature reduction to produce a slurry; none of the citations, either explicitly or implicitly, mentioned such features.
- ii) Subsequently, the parties emphasized their previous arguments regarding, on the one hand, the ash content and, thereby, the definition of the technical problem, and, on the other hand, the inventiveness of the process.
 - iii) No objection was raised against the patentability of the subject-matter of independent Claim 11.

VIII. The Appellants requested that the decision under appeal be set aside and that the patent be revoked.

The Respondents requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.
2. As it appears from points IV i) and V above, the Appellants and Opponents 2 relied on several new documents ((22) to (27)) to support their written arguments of lack of inventive step on a basis different from the approach followed before the Opposition Division. Although the Respondents first objected to the lateness of the filing without justification of these citations, they subsequently provided detailed counterarguments showing that the teaching of these citations could not affect the issue of inventive step and, thereby, the ultimate outcome of the case. For this reason, the Board, although it had come to the same finding, did not regard it appropriate to exercise its discretion under Article 114(2) EPC and did not exclude any of the late-filed documents from the discussion during oral proceedings.

During oral proceedings, however, only document (22) was extensively discussed, so that there will be no reference to documents (23) to (27) hereinafter.

3. Only the issue of lack of inventive step of the process was raised in the Statement of Grounds of Appeal (cf. point IV above). This was in line with the statements made during oral proceedings before the Opposition Division wherein, following the amendments of Claim 1, all parties agreed that the claimed subject-matter was novel with regard to the prior art which had been considered, especially documents (2) and (3), in that "none of the documents cited by any of the Opponents discloses a process for the recovery of PAS, in which the separating agent (normally water) is added after the beginning (normally at the end) of the polymerisation reaction at a temperature at which PAS is in its molten state" (cf. decision under appeal, point 7). During oral proceedings before the Board, however, the non-appealing Opponents 2 raised the issue of lack of novelty of the method as claimed on their quality as a party as of right.

Although such course of action is close to going against the course of the present appeal procedure (Article 11(3)RPBA), it is held allowable under the present circumstances because the novelty objection falls under the ground of opposition pursuant to Article 100(a) EPC which was raised initially; moreover that objection did not go beyond the scope of the procedural rights of a party as of right pursuant to Article 107 EPC, which is considered to be limited insofar as its requests may not go beyond those of the appealing party (cf. G 9/92, OJ EPO 94, 875; G 4/93 of 14 July 1994, to be published).

4. That objection of lack of novelty was based on specific embodiments disclosed, on the one hand, in documents (2) and (3), and, on the other hand, in the late filed document (22).

4.1 In particular, Opponents 2 argued that in the process according to Example XXVIII of document (2) the amount of water, which comprises both water of hydration and free water, could be calculated as being 0.3 mole water/mole solvent or 0.054 water/kg N-methylpyrrolidone (NMP), which fell within the range required in Claim 3 of the patent in suit. The same conclusion would arise from document (3), in particular from Example 20 (Table I). In both cases, thus, one had to assume that phase separation occurred like in the claimed process together with the same beneficial effects.

However, as pointed out by the Respondents, it is not proper to consider the presence of water only in quantitative terms and conclude that the same amounts will produce the same effects. In the prior art, water is added either as water of hydration or as free water and is thus present together with the unreacted initial products, or it is used to wash the polymer after separation as the final step; none of these embodiments can be equated with the controlled addition of water to the polymerisation reaction mixture, thus after the reaction started, as required in the claimed process. This is thus to be regarded as a distinguishing feature which confers novelty over the teaching of documents (2) and (3).

4.2 The further objection against novelty was based on document (22), especially on the teaching given in Example 1 said to be inherently novelty destroying. In particular, the amount of water added as steam to the reactor was found to correspond to the amount required in the patent in suit to achieve phase separation.

In fact, as demonstrated by the Respondents in the Counterstatement of Appeal (pages 5 to 8, point B. Appellants' computations) and during oral proceedings, this objection cannot be accepted for the following reasons. The first is that the computation by the Appellants is based on the assumption of equilibrium conditions, which in fact do not exist. This clearly appears from not only Example I, but equally from Example II, wherein it is stated that "the reactor ... was quickly pressured to 150 psig using steam" (Example I, column 6, lines 17 to 19; Example II, column 7, lines 20 to 22); this also appears from the next sentence in Example II (column 7, lines 22 to 24), which specifies that the pressure of the reactor when heating it to 540°F actually fell so it had to be pressured again with nitrogen subsequently. The second reason is that document (22) is silent as to the creation of phase separation of molten polymer and polar organic solvent as well as on subsequent reduction of the temperature of the separated phases to produce a slurry comprising particulate PAS in polar organic solvent. Thus, even if one assumed for the sake of argument that amounts of water as envisaged or even required in the patent in suit were implicitly used in document (22), this could not objectively be related to the effects achieved in the claimed process. Such an interpretation of the citation can only be made with the benefit of the teaching of the patent in suit, as evident not only from the Appellants' allegation on

page 7, second paragraph of the Statement of Grounds of Appeal reading:

"As seen from the description on page 4, lines 51 to 61 of the European Patent No. 96 384, it is clear that the phase separation is caused in Examples I and II of US patent No. 4, 025, 496",

but as well from the assumption made by Opponents 2 during oral proceedings that phase separation must have occurred.

- 4.3 For these reasons the Board comes to the conclusion that none of the cited documents discloses either explicitly or implicitly a method for the production of granular PAS within the terms of Claim 1 of the patent in suit, so that novelty is acknowledged.
5. The patent in suit relates to a process for recovery of granular PAS from a polymerisation reaction mixture comprising polar organic solvent, PAS and alkali metal halide by-product. As acknowledged in lines 16 to 19 of page 2 of the patent in suit, such a process is already known from document (1), in which the particulate product is recovered as fine powder by flash evaporation.
- 5.1 The discussion of the issue of novelty in point 4 above has shown that further processes of that kind are known from prior art documents (2), (3) and (22). Document (2) relates to a process as disclosed in document (1), but using at least one alkali metal carboxylate whereby a polymer of higher molecular weight is produced (see e.g. Examples XXI to XXVIII). Ash contents of the resulting polymers are given in Tables II, III and IV, but there is no information about particle size distribution.

Example XXVIII, said to demonstrate the effect of lithium benzoate on molecular weight, i.e. on inherent viscosity, indicates, however, that the reaction product consists of fine balls.

Since the ash content is not mentioned in documents (3) and (22), the Board considers document (2) to represent the closest prior art.

- 5.2 As specified in the introduction of the patent in suit (page 2, lines 29 to 33), powdery resin of PAS prepared by conventional solvent flashing process is not only difficult to filter from the arylene sulfide oligomers which are formed as by-products of the polymerisation, but also results in practical difficulties related to its relatively low bulk density. There is thus a need to provide a granular PAS which would be free from these disadvantages.
- 5.3 The problem underlying the patent in suit may thus be seen in the definition of a process for the recovery of a product with simultaneously lower ash content and larger particle size.
- 5.4 According to the patent in suit this technical problem is to be solved by contacting the polymerisation reaction mixture with an amount of a liquid, which is not a solvent for PAS, sufficient to cause phase separation, and subsequently reducing the temperature to produce a slurry of particulate PAS in polar organic solvent, as specified in Claim 1.
6. The Appellants and Opponents 2 have objected that the experimental data in the patent in suit did not show that the desired improvements in terms of particle size and ash content were achieved, i.e. that the features of

the claimed process did not provide an effective solution to the above defined problem.

6.1 In particular, the Appellants and Opponents 2 criticised the conclusions drawn from the ash contents in the examples provided during oral proceedings before the Opposition Division, since the ash content in comparative Run 4 (0.36%) was less than in Run 2 (0.40%), and the ash content in comparative Run 8 (0.07%) was less than in Run 5 (0.08%) and Run 6 (0.15%). Further, as compared with Examples I, II and V of the patent in suit, the ash content in Example VII in which 1,2,4-trichlorobenzene was used was very low, even in the case of no addition of water. It followed that Claim 1 should be restricted to the embodiments ensuring the desired low ash content.

6.2 In the Board's view, on the contrary, it is evident from Runs 1 and 2 of the patent in suit that a coarser product with a lower ash content (0.40 or 0.338 vis-à-vis 0.61) is produced when water is added to the hot polymerisation mixture to accomplish phase separation into liquid PAS and NMP solution. Runs 2 and 4 were not made under comparable conditions (absence of sodium acetate during polymerisation in Run 2) and the results cannot be compared as alleged by the Appellants.

6.3. Similarly, the results of Runs 4, 5 and 6 in Table III of the patent in suit made under comparable conditions with and without addition of water after the CO₂ treatment show that the product of Runs 5 and 6 (water added) is clearly coarser than that of Run 4 (no water added) and has also a lower ash content.

The experimental data provided by the Appellants showing that the ash content of Run 5 is higher than that of comparative Run 4, i.e. 0.336 versus 0.114, cannot cast

any doubt on the conclusiveness of the results of Runs 4, 5 and 6. The discrepancy between these results was attributed by the Appellants (cf. point IV iii) above) to the fact that the ash content is controlled to a large extent by the washing process, not by phase separation, and thus fluctuates in each experiment, even under substantially identical conditions of PAS preparation and washing. This argument, however, has not been supported by any experimental evidence showing the relative influence of washing process and phase separation on ash content and cannot, therefore, be considered.

6.4 Because of the presence of 1,2,4-trichlorobenzene in Run 8, no direct comparison with Runs 5 and 6 is possible. The conclusions drawn by the Appellants are thus not acceptable. As to the similar ash content of Runs 8 (comparative) and 9 (0.07 versus 0.06), the difference, although small, does show an improvement in the right direction and cannot consequently be used as evidence to the contrary.

6.5 In view of the experimental data in the patent in suit the Board is thus satisfied that the combination of features according to Claim 1 provides an effective solution of the above defined technical problem. By the same token these results show that the reformulation of the technical problem underlying the patent in suit was appropriate.

7. It has now to be decided whether the claimed subject-matter involves an inventive step vis-à-vis the cited prior art.

7.1 No hint is given in document (2) as to how coarser particles with a lower ash content may be produced. The same applies to the teaching given in document (3) which

is directed to the production of branched PAS of low melt flow which can be moulded, extruded or spun into fibres without prior curing by carrying out the polymerisation in the presence of controlled small amounts of water (column 1, lines 56 to 62). The method for producing PAS disclosed in document (22) is based on a specific sequenced addition of components to produce a reaction mixture which is subjected to distillation and extraction with water to produce PAS solids recoverable by filtration (Claim 1). No hint is given to carry out these final steps by phase separation with water and cooling in order to get a slurry of coarse relatively pure PAS (low ash content) in NMP solution. It follows that a person skilled in the art faced with the above defined problem would not obtain any information from these citations for the solution of that problem.

- 7.2 Document (4) relates to a process for producing granules of thermoplastic materials from molten plastic and is particularly concerned with the production of synthetic, organic, thermoplastic resin particles such as polyolefins, especially polyethylene, and with the virtual elimination of any solvent therefrom (column 1, lines 9 to 14 in conjunction with the Example). According to the Example a polyethylene resin having a methyl ethyl ketone solvent content of 90% by volume, is poured in about three times its volume of water held liquid at a temperature of 200°F under pressure. As pointed out by the Respondents, such conditions would be totally inappropriate in the case of PAS, since they would lead to dump a reaction mixture of molten PAS containing all the impurities of the reaction into an "ocean" of water; it is self evident that such a step cannot be concerned with the recovery of a clean product. A further point to be considered is that the general process disclosed in this citation aims at the production of small particles (column 2, lines 29 to 31;

column 3, line 8), which are even described as free flowing powder (column 3, line 42); the shrinking process involved (column 3, lines 38 to 41) is clearly the opposite of the effect desired in the patent in suit, which means that neither the general teaching, nor consequently the specific process features disclosed in document (4) would be regarded as relevant by the skilled person.

- 7.3 Document (8) relates to the purification of PAS containing ash forming impurities without extensive degradation of the polymer. This is achieved by heating the polymer with an organic amide, such as NMP, and a controlled amount of an alkali metal salt selected from the group consisting of alkali metal carboxylates, alkali metal carbonates and lithium halides under conditions of temperature and time to appreciably reduce the amount of ash-forming impurities without extensive degradation of the polymer (Claim 1). As specified in column 2, lines 31 to 45, PAS which can suitably be employed in that purification process is prepared by the process disclosed in US-A-3 919 177, i.e document (2). In the process according to Example X, a branched PAS first purified according to the method of Example V (column 6, line 25 to column 7, line 5), the latter corresponding to the general teaching of the citation, is molten in the presence of water. Apart from the fact that, contrary to the claimed process which is a one-step process, such a process is a two-step process, there is no mention of a phase separation occurring; moreover, since the ash value content measured in Example X (0.11 weight %) is the same as in Example VI where no water is added, there would be no incentive for a skilled person to operate according to this specific embodiment in order to lower the ash contents.

Furthermore, in view of the absence of any reference to particle size, which is the second aspect of the technical problem underlying the patent in suit, this citation cannot lead to the claimed subject-matter.

7.4 It follows that the combination of features of the process as specified in Claim 1 cannot be derived in an obvious manner from the documents cited and, therefore, it involves an inventive step.

8. Since the subject-matter of Claim 1 is allowable, the same applies to the subject-matter of dependent Claims 2 to 10 relating to preferred embodiments of the subject-matter of Claim 1.

Order

For these reasons it is decided that:

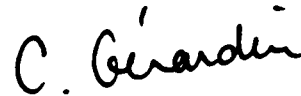
The appeal is dismissed.

The Registrar:



E. Görgmaier

The Chairman:



C. Gérardin