Europäisches Patentamt
Beschwerdekammern

European Patent Office
Boards of Appeal

Office européen des brevets
Chambres de recours

Veröffentlichung im Amtsblatt	~~Ja~~/Nein
Publication in the Official Journal	~~Yes~~/No
Publication au Journal Officiel	~~Oui~~/Non

001543

Aktenzeichen / Case Number / N° du recours : T 471/88 - 3.3.1

Anmeldenummer / Filing No / N° de la demande : 83 103 220.6

Veröffentlichungs-Nr. / Publication No / N° de la publication : 0 091 101

Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Anionic polymerization of cracker stream styrene

Klassifikation / Classification / Classement : C08F 212/08

ENTSCHEIDUNG / DECISION
vom / of / du 30 May 1989

Anmelder / Applicant / Demandeur : The Dow Chemical Company

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence : Styrene polymerisation/Dow

EPÜ / EPC / CBE Articles 56, 84 and 123(2)

Schlagwort / Keyword / Mot clé : "Inventive step (confirmed)" -
"Added subject-matter (No)"

Leitsatz / Headnote / Sommaire

Case Number : T 471/88 - 3.3.1

DECISION
of the Technical Board of Appeal 3.3.1
of 30 May 1988

Appellant :

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Decision under appeal :

Decision of Examining Division 011 **of the European Patent Office** dated 5 April 1988 **refusing European patent application No.** 83 103 220.6 **pursuant to Article** 97(1) **EPC**

Composition of the Board :

Chairman : F.J. Antony
Members : R.W. Andrews
W. Moser

Summary of Facts and Submissions

I. European patent application No. 83 103 220.6 (publication No. 0 091 101), filed on 31 March 1983 and claiming priority of 2 April 1982 from a prior application in the United States of America, was refused by a decision of the Examining Division 011 of the European Patent Office dated 5 April 1988. The decision was based on Claims 1 to 6 filed on 16 August 1986.

II. The stated ground for the refusal was that the claimed subject-matter did not involve an inventive step in the light of the disclosure in US-A-2 813 089 (1). The Examining Division considered that the skilled person faced with the problem of polymerising the present precursor fraction would be aware of its phenylacetylene content and of its deleterious effect on the molecular weight of the resulting polymer. Therefore, it would be obvious to remove or at least reduce the amount of phenylacetylene by selective hydrogenation.

The use of polymerisation feedstocks containing 15 to 25% by weight of styrene in document (1) would not deter the skilled person from submitting feedstocks containing 30 to 60 parts by weight of styrene since no prejudice existed against the anionic polymerisation of feedstocks containing larger amounts than 25% by weight of styrene.

Objections were additionally raised against allegedly new subject-matter in Claim 4 (Article 123(2) EPC) and alleged lack of clarity (Article 84 EPC) of Claim 1.

III. A notice of appeal was filed on 5 May 1988 and the prescribed fee duly paid. A statement of grounds of appeal was filed on 4 August 1988. In this statement and during the oral proceedings held on 30 May 1989 the following submissions were made on the behalf of the Appellant:

Styrene is produced commercially on a large scale by dehydrogenating ethylbenzene obtained by the ethylation of benzene. Document (1) discloses an economic alternative to this process insofar as the separation and purification of either the ethylbenzene or the styrene is avoided. After a lapse of 25 years the present process is an alternative process which is more economic than that disclosed in document (1). In his discussion of document (1) the Appellant emphasised the differences between this prior art process and the presently claimed one.

The Appellant also contended that it was not known prior to the present invention that the feedstock contained phenylacetylene or that phenylacetylene inhibited the polymerisation of styrene. Furthermore, document (1) did not mention phenylacetylene and from the documents cited by the Rapporteur's letter of 10 March 1989 it was impossible to predict that the small amounts of phenylacetylene present in the feedstock could be selectively hydrogenated without hydrogenation of the styrene taking place.

IV. The Appellant requests that the decision of the Examining Division be set aside and a patent granted on the basis of either of the texts in accordance with the main and auxiliary requests submitted during the oral proceedings. The only independent claim in accordance with the main request reads as follows:

"A process for the preparation of styrene polymers from a petroleum byproduct stream from the cracking of petroleum products, the stream containing primarily C_8 compounds of an aromatic nature and comprising 30 to 60 parts by weight of styrene, 40 to 70 parts by weight of mixed isomers of xylene, and 0.5 to 2 parts by weight of phenylacetylene, characterised by a direct process including the steps of

(1) selectively hydrogenating in the presence of a catalyst the phenylacetylene to styrene so that no phenylacetylene is detectable by gas chromatography,

(2) polymerizing the styrene in the stream with an organolithium initiator whereby at least 99 percent of the styrene in the stream is converted to polymer having a weight average molecular weight in the range of 150 000 to 300 000 and

(3) recovering the styrene polymer.

VI. At the conclusion of the oral proceedings the decision of the Board was announced.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.

2. There are no formal objections to the present claims since they are adequately supported by the original disclosure. Thus, Claim 1 finds support in Claim 1 as originally filed, page 2, lines 5 to 10, page 3, lines 3 to 11 and lines 24 to 26 and page 5, lines 14 to 18. Claims 2, 3, 5 and 6 correspond to original Claims 3, 4, 2 and 5 respectively. In the Board's judgement Claim 4 is adequately supported by the Example in which after selective hydrogenation of phenylacetylene, the filtered reaction mixture is distilled before polymerisation. Any water present in the reaction mixture would be distilled over in the first fraction of

the distillate. Since the first 50 ml of the distillate was discarded this would result in the remainder of the distillate being free from water. Moreover, it is clear that the second fraction of the distillate must be free from water, since it polymerised after being charged into a dry round bottom flask.

In the Board's judgement the present Claim 1 satisfies the requirement of Article 84 EPC with respect to clarity insofar as the phrase "at least the major portion" has been deleted and the definition of the petroleum by-product stream from the cracking of petroleum products used as starting material in terms of parts by weight of styrene, xylenes and phenylacetylene is sufficiently clear in the present circumstances.

The description has been amended to bring it into agreement with the amended statement of claim and to correct clerical errors.

3. The application relates to a process for directly preparing styrene polymers of moulding grade from a petroleum byproduct stream from the cracking of petroleum products containing aromatic C_8 compounds. Document (1), which is considered to represent the closest state of the art, discloses a process for the direct production of moulding grade polystyrene from a C_8 aromatic fraction obtained in the hydroforming of naphtha.

In the light of this closest prior art, the technical problem underlying the application in suit may be seen in providing an alternative process for the direct production of moulding grade polystyrene from a petroleum byproduct stream.

According to the patent application this problem is essentially solved by using as feedstock for the process a petroleum byproduct stream comprising 30 to 60 parts by weight styrene, 40 to 70 parts by weight of mixed isomers of xylene and 0.5 to 2.0 parts by weight of phenylacetylene. The phenylacetylene is removed by selective catalytic hydrogenation and the styrene in the thus treated stream is polymerised with an organolithium initiator to yield a polymer having a weight average molecular weight in the range of 150 000 to 300 000.

In the light of the Example, the Board is satisfied that this technical problem is plausibly solved.

4. After examination of the cited prior art, the Board has reached the conclusion that this technical teaching is not disclosed therein and the claimed subject-matter is, therefore, novel. Since novelty is not disputed it is not necessary to consider this matter in detail.

5. It still remains to be examined whether the requirement of inventive step is met by the claimed subject-matter.

5.1 The starting material for the process described in document (1) is a C_8 aromatic fraction comprising 10 to 40% of ethylbenzene and mixed isomers of xylene (cf. column 1, lines 52 to 60). The ethylbenzene in this feedstock is dehydrogenated to provide, after removal of water and tar, a solution of styrene in xylenes which contains some ethylbenzene, toluene and benzene. This product, after drying but without any further purification or separation is introdued into a polymerisation reactor wherein the styrene monomer is converted to high molecular weight polymer by contact with a catalyst consisting of finely divided metallic sodium (cf. column 2, lines 40 to 53). According to this prior art document the use of sodium as the catalyst for the polymerisation step is essential since thermal polymerisation, free radical polymerisation or polymerisation with acid type catalysts did not yield "polystyrene of desirable properties in feasible yields" in the xylene solution (cf. column 2, lines 53 to 58).

Thus, although this prior art document teaches that moulding grade polystyrene may be prepared directly from a petroleum byproduct stream, the presently claimed process differs from this earlier process in several respects. Insofar as the amounts of styrene in the feedstock to the polymerisation reactor are different in the two processes, it is considered that the skilled person would not be discouraged from attempting to polymerise the present feedstock merely by the fact that it contained a higher concentration of styrene than that used in the process disclosed in document (1).

5.2 Phenylacetylene, which is present in small amounts in the feedstock used in the present process, is not mentioned in document (1). Thus, the skilled person would not be able to draw any conclusion with regard to the effect of this compound on the polymerisation of styrene from this document. Furthermore, the Board is not in a position to refute the Appellant's statement made during the oral proceedings that the presence of phenylacetylene in the present starting material was unknown until the development of the present invention. However, in the Board's opinion the skilled person contemplating using a starting material which is a mixture of compounds would almost certainly have the material analysed before employing it. In the event that this analysis had not been carried out, the skilled person would automatically have the material analysed if the desired result of the process in which the material was used as feedstock was not achieved. Therefore, at least at that stage, the skilled person would become aware of the presence of phenylacetylene in the present feedstock.

5.3 The Appellant made the presently irrefutable statement that the adverse effect of phenylacetylene on the molecular weight of polystyrene was unknown before its disclosure in the present patent application (cf. the paragraph bridging pages 2 and 3). Thus, if the skilled person seeking a solution to the above-defined technical problem decided to use the present feedstock and to follow the teaching of document (1), the resulting polystyrene would be unsatisfactory insofar as its weight average molecular weight would not fall within the claimed range. In view of the teaching of document (1) with respect to the importance of the choice of catalyst, the skilled person would consider replacing the sodium catalyst by other anionic catalysts or abandoning the use of anionic polymerisation conditions in favour of thermal or free radical polymerisation or polymerisation with cationic catalysts. However, even the use of an organolithium in place of the sodium catalyst as taught by document (1) would not solve the problem underlying the application in suit.

5.4 In the light of this failure, the skilled person would turn his attention to the components of the feedstock. Although there is no prior art describing the adverse effect of phenylacetylene on the polymerisation of styrene, the skilled person would consider removing it in a further attempt to solve the present technical problem, particularly since this compound is not mentioned as being present in the feedstock used in the process described in document (1) and in view of the fact that it contains a triple bond. Of the methods available for the removal of the phenylacetylene from the present feedstock, the skilled person could have considered selective hydrogenation.

However, the skilled person would have been unable to predict whether such a procedure would be successful in view of the small amount of phenylacetylene present in the mixture and the requirement that hydrogenationshould be so selective that substantially none of the styrene should be reduced to ethylbenzene.

Although the selective hydrogenation of acetylenes to the corresponding olefins is known (cf. Catalytic Hydrogenation in Organic Syntheses, Paul Rylander, pages 13 to 19 (2); Journal of the American Chemical Society, Volume 86, pages 4358 to 4363, 1964 (3); and Chemische Technik, Volume 25(10), pages 604 to 608, 1973 (4)), none of these documents disclose the selective hydrogenation of small amounts of phenylacetylene in the presence of styrene.

Document (2) discusses the effects of various factors, such as catalyst loading and agitation, temperature, solvent, catalysts, and metal additives, on the selectivity in the hydrogenation of acetylenes. From the teaching of this document the skilled person would not find any encouragement to adopt selective hydrogenation for removing the phenylacetylene from the present feedstock.

Document (3) describes the homogeneous reduction of pure phenylacetylene to styrene by chromous sulphate in a mixture of dimethylformamide and water (cf. third paragraph of the right-hand column on page 4362) However, in view of the use of a polar solvent mixture and the effects of solvents on selectivity (cf. document (2), page 16), the skilled person would not be immediately led by the teaching of this document to the claimed method for removing the small amounts of phenylacetylene in the feedstock.

Document (4) discloses the selective hydrogenation in the gas or liquid phase of unsaturated hydrocarbons having 2 to 4 carbon atoms. According to this document, losses of butadiene are unavoidable during the selective hyrogenation of the C_4 fraction in the liquid phase. Similarly, if the amounts of methylacetylene and propadiene in the C_3 fraction are small and to be reduced to a low level, the yield of propylene is reduced (cf. left-hand column on page 604). However, the removal of acetylene from ethylene by selective hydrogenation in the gaseous phase can be successfully accomplished (cf. right-hand column on page 606). Thus, the teaching of this document would not allow the skilled person to draw any conclusions regarding the feasibility of using selective hydrogenation to remove the small amounts of phenylacetylene in the present starting material.

5.5 Therefore, in the Board's judgement, the cited prior art would not have suggested to the skilled person that the solution to the above-defined technical problem lay in using the present feedstock as a starting material in a process for the preparation of moulding grade polystyrene whereby the phenylacetylene present is removed by selective hydrogenation and the thus purified stream is polymerised in the presence of an organolithium initiator.

6. In summary, the subject-matter of Claim 1 in accordance with the main request involves an inventive step. Dependent Claims 2 to 6, which relate to preferred embodiments of the process according to Claim 1, derive their patentability from this claim.

7. In view of the above, the Appellant's auxiliary request may be disregarded.

Order

For these reasons, it is decided that:

1. The decision of the Examining Division is set aside.

2. The case is remitted to the first instance with the order to grant a patent on the basis of the documents according to the main request.

The Registrar: The Chairman:

F.Klein F.Antony