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

Aktenzeichen / Case Number / N° du recours : T 198/89 - 3.3.3

Anmeldenummer / Filing No / N° de la demande : 82 102 422.1

Veröffentlichungs-Nr. / Publication No / N° de la publication : 0 062 230

Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Method for preventing polymer scale deposition in the polymerization of ethylenically unsaturated monomers

Klassifikation / Classification / Classement : C08F 2/00

ENTSCHEIDUNG / DECISION

vom / of / du 22 June 1990

Anmelder / Applicant / Demandeur : Shin-Etsu Chemical Co. Ltd.

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step (denied)"

Leitsatz / Headnote / Sommaire

Europäisches Patentamt
Beschwerdekammern

European Patent Office
Boards of Appeal

Office européen des brevets
Chambres de recours

Case Number : T 198/89 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 22 June 1990

Appellant : Shin-Etsu Chemical Co., Ltd.
6-1, Otemachi 2-chome
Chiyoda-ku, Tokyo
(JP)

Representative : Jaeger, Klaus, Dipl.-Chem. Dr.
Jaeger & Partner
Patentanwälte
Pippinplatz 4a
D-8035 Gauting

Decision under appeal : Decision of Examining Division 011 of the European Patent Office dated 5 October 1988 refusing European patent application No. 82 102 422.1 pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : F. Antony
Members : C. Gérardin
J. Stephens-Ofner

Summary of Facts and Submissions

I. European patent application No. 82 102 422.1, filed on 23 March 1982, claiming priority of 23 March 1981 from an earlier application in Japan and published on 13 October 1982 under the publication No. 62 230, was refused by a decision of the Examining Division dated 5 October 1988.

The rejection was based on a set of 7 claims, Claim 1 filed on 16 October 1987 reading as follows:

"A method, in the suspension polymerization of an ethylenically unsaturated polymerizable monomer in an aqueous medium contained in a polymerization reactor, for preventing deposition of polymer scale on the surfaces of the inner walls of the polymerization reactor and other parts coming into contact with the monomer during the polymerization, which comprises, prior to the polymerization, providing a coating layer on the surface formed of an oxidative condensation product formed of an aromatic amine compound, characterized in that the condensation product is made by the condensation reaction of at least one aromatic amine compound and at least one aromatic nitro compound, the amount of aromatic nitro compound being at least in the range from 0.10 to 0.50 mole per mole of the aromatic amine compound, in the presence of the mineral acid and a condensation catalyst at a temperature in the range from 100 to 250°C."

II. The ground for this decision was that the subject-matter of the application in suit was not novel or, in any case, did not involve an inventive step with regard to the teaching of following documents:

(1) BE-A-756 008

(2) The Chemistry of Synthetic Dyes by K. Venkataraman, Vol. II, page 775, Academic Press, 1952,
(3) Ullmanns Encyklopädie der technischen Chemie, 4th edition, Vol. 8, pages 230-231, Verlag Chemie, Weinheim,
(4) JP-A-53/13 689 (English translation).

More specifically, it was stated in the decision that a method for preventing the deposition of polymer scale on the inner walls of polymerisation reactors was described in document (1), wherein said walls were coated prior to polymerisation with organic nitrogen-containing polar compounds, such as nigrosine. Although the conditions concerning the method of preparation of these compounds, especially the oxidative conditions, the identification of the starting compounds, the molar ratios and the general condensation conditions, were not explicitly mentioned, they could be regarded as implicitly disclosed.

It was further specified that the essential operative features were in fact common in the art and actually taught in documents (2) and (3). Furthermore, reference was made to the English translation of document (4) provided by the Appellant and discussed in detail in a previous communication to support the general objection of lack of inventive step.

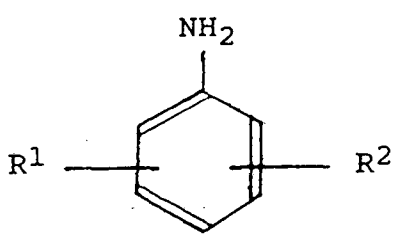
III. A Notice of Appeal was lodged against that decision on 2 December 1988 with payment of the prescribed fee. The arguments presented by the Appellant in the Statement of Grounds of Appeal filed on 9 February 1989, together with a new set of 7 claims, can be summarised as follows:

Document (1) should be regarded as a broad teaching encompassing a large number of nitrogen-containing compounds, unlike new Claim 1 of the application in suit

which was now directed to the use of specific condensation products prepared under specific conditions. These conditions resulted in improved efficiency against deposition of polymer scale on the surface of the inner walls of polymerisation reactors with regard to similar commercial products; this technical effect was evidence of novelty. Moreover, since this improvement was unexpected, it should confer an inventive step to the claimed subject-matter. Besides these substantive arguments, it was objected from a procedural point of view that the combination of the teaching of document (1) with features taken from other prior art documents in order to demonstrate lack of novelty was not permissible.

IV. The new Claim 1 filed in accordance with these arguments differs from Claim 1 as rejected mainly by the introduction into the characterising part of the claim of specific definitions for the aromatic amine and the aromatic nitro compound by means of general formulae.

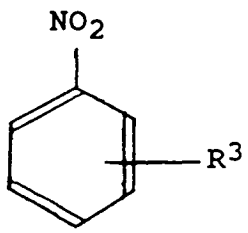
The aromatic amine is defined as having "the following general formula



wherein R^1 is a hydrogen atom, a chlorine atom or a group selected from the class consisting of amino, phenylazo ($-N=N-C_6H_5$), hydroxy, acetyl, methoxy, phenylamino, aminophenylamino, methoxyphenylamino, dimethylamino, hydroxyphenylamino and acetylamino groups as well as alkyl groups having from 1 to 3 carbon atoms, and

R^2 is a hydrogen atom, an amino group, a hydroxy group or a methyl group, ..."

Likewise, the aromatic nitro compound is defined as having "the following general formula



wherein
R^3 is a hydrogen atom, a chlorine atom or a group selected from the class consisting of hydroxy, methoxy, ethoxy, amino, carboxyl (-COOH) and sulfo ($-SO_2OH$) groups, ..."

Further, the term "at least" before "in the range from 0.10 to 0.50 mole..." has been omitted.

Claims 2 to 7 concern preferred embodiments of the method according to Claim 1.

V. Together with the Notice of Appeal oral proceedings were requested in case the Board could not allow the appeal in writing. Accordingly the Appellant was summoned to oral proceedings scheduled to take place on 12 June 1990.

On 7 June 1990 the Appellant withdrew his request for oral proceedings and informed the EPO that he would not attend any oral hearing. He further requested that a decision be issued on the basis of the documents on file. The oral proceedings scheduled were cancelled accordingly.

VI. The Appellant requests that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 7 filed on 9 February 1989.

Reasons for the Decision

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

2. The wording of the claims does not give rise to objections under Article 123(2) EPC.

Claim 1 differs from Claim 1 as originally filed by the introduction into the characterising part of the claim of the definition of the aromatic amine and the aromatic nitro compound used to prepare the oxidative condensation product and further by the relative amounts of the two reactants. The general formulae of the two reactants are disclosed respectively on page 7, paragraph 2 and page 8, paragraph 2 of the original application. As to the relative amounts of the two reactants, they correspond to the subject-matter of original Claim 5.

Claims 2 to 7 correspond respectively to Claims 2 to 4 and 6 to 8 of the application as originally filed.

3. The application in suit concerns a method for preventing polymer scale deposition in the polymerisation of ethylenically unsaturated monomers. Such a method is disclosed in document (1) which the Board, like the Examining Division, considers to be the closest state of the art. That document describes a process for the suspension polymerisation of vinyl chloride, wherein the inner walls of the reactor are coated prior to the polymerisation reaction with certain organic compounds in

order to reduce or even prevent the deposition of polymer scale (Claim 1). The compounds suitable for this purpose are generally defined as polar compounds selected from an extremely broad class of compounds (page 9, line 5 to page 11, line 7), among which "nigrosine" is explicitly mentioned (page 9, line 19). Experimental data show that the amount of polymer scale deposited on the inner walls of the reactor, and consequently the number of polymerisation runs likely to be performed after one single coating operation, varies within broad limits according to the compound applied.

In the light of this prior art, the technical problem underlying the application in suit can be regarded as providing a class of compounds of optimal efficacy in preventing scale deposition, thereby allowing a large number of polymerisation runs.

According to Claim 1 of the application in suit this problem is solved by using a coating of an oxidative condensation product prepared by reacting 0.1 to 0.5 mole of at least one aromatic amine of given formula with 1 mole of at least one aromatic nitro compound of given formula in the presence of a mineral acid and a condensation catalyst at a temperature in the range from 100 to 250°C.

In view of the experimental results mentioned in the application in suit, particularly those in Table 7 showing that up to 80 polymerisation runs can be performed in the same reactor by merely discharging the polymerisate slurry out of the reactor and washing the reactor walls with water, the Board is satisfied that the above technical problem has been effectively solved.

4. As stated above, document (1) does mention "nigrosine". The fact that this compound is only one out of a long list, therefore, cannot detract from the fact that the use of "nigrosine" simply is not novel, there being no question of a selection of a compound specifically mentioned in the prior art and actually used in several examples.

However, as appears from document (2), the term "nigrosine" does not relate to an individual compound, but stands for a whole class of compounds obtainable by simple condensation in the presence of hydrochloric acid of aniline with an aromatic nitro compound; this reaction is carried out either under oxidising conditions involving the use of ferric chloride, or under reducing conditions involving the use of iron metal. It is further specified that nigrosines have good dyeing properties, the actual shade depending on the degree of phenylation which is determined by the relative amounts of the two reactants and the time of heating (page 775, lines 5 to 19).

The structural complexity of the "nigrosine" class of dyestuffs is mentioned in document (3) as well, which also confirms the criticality of the molar ratio of the two reactants for the properties of the condensation product (page 230, point 2.2.4, paragraphs 1 and 2).

Documents (2) and (3) both reflect common general knowledge and were thus rightfully taken into consideration by the Examining Division in its investigation on novelty over the teaching of document (1); however, in the absence in document (1) of any reference to the particular condensation conditions applied to the preparation of "nigrosine", the use of any specific "nigrosine" compound cannot be regarded as individually disclosed in this document; for this reason,

the subject-matter of Claim 1 according to the application in suit must be regarded as novel.

5. The issue of inventive step thus boils down to the question whether for a skilled person, knowing from document (1) that scale formation during the suspension polymerisation of, e.g., vinyl chloride can be reduced by coating the inner walls of the reactor with "nigrosine" prior to the polymerisation reaction, and faced with the problem of even further suppressing such scale formation, the combination of reaction conditions and choice of reactants as set out in Claim 1 can be regarded as inventive.

More specifically, this question reduces to the obviousness of choosing following starting materials and operative features:

(a) aromatic amine and aromatic nitro compound as defined in Claim 1;
(b) condensation reaction under oxidative conditions;
(c) at a molar ratio of from 0.1 to 0.5 mole of nitro compound per mole of amine;
(d) in the presence of mineral acid and a condensation catalyst;
(e) at a temperature in the range from 100 to 250°C in order to define a relatively narrow class of "nigrosine" compounds having an effectiveness in preventing polymer scale deposition on the reactor walls higher than the unspecified "nigrosine" compounds referred to in document (1).

5.1 In this connection document (4), which deals with a closely similar problem, is of the greatest relevance. This document teaches that the scale preventing effect of aromatic amine condensation products coated on the inner walls of polymerisation reactors depends to a large extent

on the method of preparation of these additives. More specifically, this document describes a method of polymerisation, especially suspension polymerisation, of ethylenically unsaturated monomers, wherein the reactor walls are coated with an oxidative condensation product of an aromatic amine and optionally with an electron-accepting organic compound in order to reduce polymer scale deposition (Claim 1). It is further specified that an aromatic amine before condensation, thus per se, has a low scale preventing effect, that an aromatic amine condensed using a reducing agent has no effect with a water-soluble catalyst, but is effective with an oil-soluble catalyst, and that an aromatic amine condensed using an oxidising agent is effective with oil soluble as well as water-soluble catalysts (page 3, paragraphs 3 and 4). This enhanced effectiveness is attributed to the fact that the oxidative condensation product is scarcely susceptible to the influence of the oxidising activity caused by the initiator, and is thus not likely to be decomposed by oxidation.

In the Board's view, the high effectiveness and stability of the condensation products prepared by the oxidative route can only be a direct incentive for the skilled man to adopt these operative features for the preparation of the condensation products disclosed in document (1).

The list of aromatic amines suitable for this purpose (page 4, paragraph 5 to page 5, paragraph 1) comprises aniline as well as various compounds which can be regarded as substitution derivatives therefrom, wherein the substituent(s) can be a chlorine atom or an amino, hydroxy, methyl or methoxy group or a combination thereof, which all correspond to the definition of the radicals R^1 and R^2 in Claim 1 of the application in suit. Likewise, the list of electron-accepting compounds includes nitrobenzene (page 6, line 32) and nigrosine as an

oxidative condensation product is actually used in several experiments (32,37,38,42,46,51,59,65 and 70). This teaching corresponds to the features identified above under (a) and (b).

5.2 As far as the features (c) to (e) are concerned, although they are not specifically disclosed as such in document (4), they are encompassed within the broad scope of its teaching, which mentions, respectively, a weight ratio of 1:20 to 20:1 for the two reactants (page 7, paragraph 3 to page 8, paragraph 1), various mineral acids to carry out the oxidative condensation reaction (page 5, paragraph 2) and temperatures of 50°C or more for this reaction (page 6, paragraph 1 and page 9, paragraph 2).

In fact, in the light of the teachings of documents (2) and (3) above, features (c) to (e) can be regarded as common general knowledge. In document (3), which illustrates three embodiments of the preparation of a nigrosine compound, it is indicated that the temperature normally rises as the result of the exothermic reaction between aniline and nitrobenzene in acid medium, and is maintained at about 180°C according to the three examples of preparation of nigrosine. Furthermore, the molar ratios of the two reactants used in these examples fall within the range as required in the application in suit. Likewise, document (2) mentions the use of a strong mineral acid in the oxidative condensation reaction of aniline and nitrobenzene.

Accordingly, in the Board's view, there can be no doubt that features (c) to (e) were well known in the art and that the skilled man would have had no difficulty in modifying these operative features according to the starting compounds, i.e. according to the meaning of the radicals R^1, R^2 and R^3.

5.3 In view of the above considerations, both the operative features of the condensation reaction and the choice of the compounds suitable for this reaction must be regarded as obvious for the solution of the problem underlying the application in suit. The advantageous results put forward by the Appellant in the Statement of Grounds of Appeal (page 5, paragraph 1 to page 6, paragraph 4) regarding polymer scale deposition and number of polymerisation runs which can be performed after one single coating operation with condensation products prepared by the oxidative route, are not disputed. However, in the Board's view, they merely confirm the greater effectiveness conferred by the oxidative route to these condensation products mentioned in document (4) and hence cannot be regarded as surprising. For this reason, the subject-matter of Claim 1 of the application in suit does not involve an inventive step.

6. Claim 1 not being allowable, the same applies to the dependent method Claims 2 to 7 which merely represent preferred embodiments of the subject-matter of Claim 1 and thus fall with it.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

M. Beer F. Antony

03350