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Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Process for preparing an acrylonitrile polymer

Klassifikation / Classification / Classement : C08F 20/44

ENTSCHEIDUNG / DECISION

vom / of / du 17 January 1989

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /
Titulaire du brevet : American Cyanamid Company

Einsprechender / Opponent / Opposant : Hoechst AG

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step (confirmed) -
Common general knowledge (insufficient)"

Leitsatz / Headnote / Sommaire

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Case Number : T 439 /87 - 3.3.1

DECISION
of the Technical Board of Appeal 3.3.1
of 17 January 1989

Appellant : (Opponent)

HOECHST AKTIENGESELLSCHAFT, Frankfurt
-Zentrale Patentabteilung-
Postfach 80 03 20
D-6230 Frankfurt am Main 80

Representative :

Respondent : (Proprietor of the patent)

AMERICAN CYANAMID COMPANY
1937 West main Street
P.O. Box 60 Stammford
Connecticut 06904-0060
USA

Representative :

Diehl, Hermann O., Dr.
Deihl & Kressin
Flüggenstrasse 17
D-8000 M

Decision under appeal : Interlocutory decision of the Opposition Division of the European Patent Office date 13 October 1987 concerning maintenance of European patent No. 0 027 921 in amended form.

Composition of the Board :

Chairman : K. Jahn
Members : R. Spangenberg
L. Mancini

Summary of Facts and Submissions

I. The mention of grant of European patent No. 0 027 921 in respect of European patent application 80 105 983.3 filed on 3 October 1980 and claiming priority of 29 October 1979 of an earlier application in the USA was announced on 16 May 1984 (Bulletin 84/20) on the basis of nine claims relating to acrylonitrile polymers and their preparation.

II. A notice of opposition was filed by the Appellant on 15 February 1985, requesting revocation of the patent in suit in its entirety on the grounds of insufficient disclosure, lack of novelty and lack of inventive step. Seven documents were cited, the most important being

(1) US-A-2 974 123
(3) US-A-2 847 405
(4) US-A-3 025 278
(5) DE-B-1 300 678
(6) DE-A-2 400 043

III. By an interlocutory decision dated 13 October 1987 the Opposition Division maintained the patent in suit in amended form on the basis of 4 claims. The only independent claim read as follows:

"A process for preparing an acrylonitrile polymer which comprises forming, at a temperature in the range of 40°C to 80°C, an aqueous suspension of a monomer content comprising at least 85 weight percent acrylonitrile, the balance being one or more ethylenically unsatured monomers free of acid dyesites copolymerizable with acrylonitrile, said monomer content forming from 25 to 40 weight percent, based on the weight of said suspension, initiating polymerization by adding to said suspension effective amounts of a redox system comprising 4-20 parts per 1,000 parts of monomer, of an oxidizing agent selected from the group consisting of potassium persulfate, potassium chlorate, potassium perchlorate and potassium peroxide, their corresponding sodium and ammonium salts and hydrogen peroxide and a reducing agent, terminating said reaction at a monomer conversion in the range of 75% to 90% and recovering the polymer thus produced characterized in that the reducing agent comprises a mixture of a bisulfite and 0.4-0.8 percent by weight, based on monomers, of a water-soluble mercapto compound, the relative proportions of said bisulfite and said mercapto compound being such as to provide a sulfonic acid end group content in the resultant polymer in the range of 0.20 to 0.35 weight percent, based on the weight of the polymer and to provide a polymer having an intrinsic viscosity in the range of 0.5 to 1.4, and there is added with said redox system a copper activator."

The decision was mainly based on the argument that it was not obvious to modify the process disclosed in document (5) in order to solve the problem of varying the intrinsic viscosity of acrylonitrile polymers while maintaining a constant level of sulfonic acid end groups derived from a bisulfite compound used as a component of the redox initiator system essentially by employing mercaptans as chain transfer agents together with a copper activator in this process. Document (4) was not considered relevant since it relates to the problem of varying the dyability while maintaining a certain molecular weight. Moreover, it does not mention a copper activator. Document (6) was said to require a nitrogen compound as activator which is not used in the claimed process.

The Appellant's argument of insufficient disclosure was not accepted.

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IV. The Appellant filed a notice of appeal against the above decision on 20 November 1987 and paid the appeal fee at the same date. A statement of grounds for appeal was received by telecopy on 22 February 1988, being confirmed on 23 February and relying on further documents, <u>inter alia</u>

(8) Ullmanns Encyclopädie der technischen Chemie (4. Aufl., 1976) Vol. 11, page 328
(9) Ullmanns Encyclopädie der technischen Chemie (4. Aufl., 1976), Vol. 15, pages 187-189,

in order to demonstrate the common general knowledge in the technical field concerned.

V. In the Statement of Grounds and at the oral proceedings held on 17 January 1989, the Appellant argued essentially that the problem underlying the patent in suit was to vary the amount of dyesites and the molecular weight of an acrylonitrile polymer independently. This problem could have been solved by merely applying the common general knowledge as represented by (8) and (9). It was known from (8) that acrylonitrile polymers are made by suspension polymerisation in an aqueous system using a redox initiator system, especially the system $K_2S_2O_8/NaHSO_3$ also encompassed by the claimed process, and that this reaction is catalysed by heavy metal ions. The polymerisation is started by SO_4 and SO_3H radicals. It is also common general knowledge that these radicals are the end-groups of the polymer obtained and represent its dyesites. The function of mercapto compounds as chain transfer agents which lower the molecular weight is also well known in the art, see (9) pages 188-189, the chapter headed "Kettenabbruch, Kettenübertragung". It is evident from the reaction scheme on page 188 that in this case mercapto radicals start new chains and that therefore by using such compounds a decrease of the molecular weight of the polymer can be obtained without increasing the content of sulfonic acid end groups. With respect to this common general knowledge the determination of the exact amounts and ratios of the reagents is a matter of routine experimentation which does not involve an inventive step. Such routine work is also necessary when putting the claimed process into practice because in Claim 1 of the patent in suit these amounts and ratios are only defined by the result to be achieved.

It was further submitted that document (4) does not only teach how to modify the dyeability of polyacrylonitrile fibres while maintaining a certain molecular weight but also what to do if a modification of the molecular weight is required while maintaining a certain level of dyeability.

Reference was also made to document (6), especially page 8, Table 4, lines 4 and 5, and it was stated that only the variation of the mercaptan content and not, as stated in Claim 1 of the patent in suit, the relative proportions of the bisulfite and the mercaptan is responsible for the variation of the molecular weight of the polymer obtained, a fact which can also be derived from Examples 2 and 3 of the patent in suit.

Moreover, the Appellant stated that document (5) discloses all technical features of the claimed process except the amounts of the reagents to be used.

VI. The Respondent filed a new Claim 1 containing the conditions for the determination of the intrinsic viscosity values of the polymer (measured in dimethyl formamide at 40°C). He submitted that the background art

(9) contains very general statements which cannot be applied to the specific process for making acrylonitrile polymers containing sulfonic acid dyesites. He stated that the need for independently varying molecular weight and sulfonic acid end group content of these polymers existed at least for 20 years. Nevertheless the problem has been hitherto solved by quite different approaches, namely either by incorporating dyesite containing comonomers or by varying the amount of the initiator system as disclosed in (4).

Document (6) which relates to a bulk polymerisation process, i.e. a different technique should be considered irrelevant since the molecular weight was modified by a combination of a mercapto compound and a nitrogen containing compound while mercapto compounds alone were ineffective.

The Respondent further stated that the result of the claimed process is not achieved by choosing <u>independently</u> the amount of sulfite as the reducing component of the redox initiator system and the amount of mercapto compound but to use in combination with a copper activator mercapto compounds in a certain range of percentage, based on monomers and to adjust the <u>relative proportions</u> of sulfite and mercapto compound in order to obtain the desired content of sulfonic acid dyesites and the desired range of viscosity. Neither the background art (8) and (9) nor any other document however suggests this approach to solve the problem identified above. He therefore considers the claimed process non-obvious and the Appellant's objections, collecting together various bits of prior art after the invention has been disclosed, being based on hindsight.

VII. The Appellant requested that the decision under appeal be set aside and the patent revoked.

The Respondent requested that the decision under appeal be set aside and the patent maintained with the documents submitted at the oral proceedings.

At the conclusion of the oral proceedings the decision was announced to maintain the patent as requested by the Respondent.

Reasons for the Decision

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

2. At the oral proceedings neither the insufficiency objection nor the objections against the version of the claims under consideration were maintained. As the method for determining the intrinsic viscosity was disclosed in the application documents as filed (see page 6, lines 15 to 17) and the patent as granted, page 3, lines 55-56, no formal objection arises against the current version of the claims.

3. The amounts and proportions of bisulfite and mercapto compound to be used according to the patent in suit are not disclosed in any one of the cited documents. The claimed process is therefore novel. As novelty has no longer been disputed by the Appellant this does not require any detailed explanation.

4. The patent in suit relates to a process for preparing an acrylonitrile polymer free of comonomers containing acid dyesite groups in aqueous suspension in the presence of a

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redox initiator system employing a bisulfite as the reducing agent whereby the content of end groups derived from the bisulfite is controlled in order to maintain uniform dyeing characteristics of the polymer while the intrinsic viscosity, which depends on the molecular weight of the polymer, can be varied independently.

Among the cited prior art document (5) relates to such a suspension polymerisation process having the greatest number of technical features in common with the claimed process since it relates to the use of copper salts as catalysts in a process as described above (see Claim 1) and also mentions the optional use of known chain transfer agents such as mercaptans, especially mercaptoethanol (see column 6, lines 6 to 10 and column 12, Example 8). However, the intended purpose of the copper salt and the mercaptoethanol is to produce colourless and gel-free polymers and the question of dyeability, let alone uniform dyeability irrespective of the molecular weight of the polymer, is not addressed at all. This document does not therefore qualify as closest prior art.

It is true that document (6) relates to acrylonitrile polymers having a constant level of dyeability while the molecular weight is varied (see page 3, lines 10 to 12). However, a bulk polymerisation technique, i.e. a process in the absence of substantial amounts of water is suggested.

The only document cited so far which describes an aqueous suspension polymerisation process in detail and also relates to the variation of the molecular weight of the acrylonitrile without altering its dyeability is (4). According to column 1, lines 57-59 and the sentence bridging columns 1 and 2, this goal can be achieved by shifting the total concentration of the oxidising and reducing agent. The Board therefore finds it appropriate to consider this document as the closest prior art.

5. Vis-à-vis this prior art the technical problem underlying the patent in suit may be seen in providing an alternative method for achieving the same objective.

In order to solve this problem, the patent in suit suggests a process of the kind specified in the precharacterising part of Claim 1 with the further qualifications that the redox system additionally contains a copper activator, and the reducing agent comprises a mixture of a bisulfite and 0.4 to 0.8 percent by weight, based on monomers, of a water-soluble mercapto compound, the relative proportions of said bisulfite and said mercapto compound being such as to provide a sulfonic acid end group content in the resultant polymer in the range of 0.20 to 0.35 weight percent, based on the weight of the polymer, and to provide a polymer having an intrinsic viscosity, measured in dimethyl formamide at 40°C, in the range of 0.5 to 1.4.

6. The Board construes the above expression "the relative proportions" as meaning the ratio of bisulfite and mercapto compound. As can be seen from Examples 1 and 4 of the patent in suit different ratios of the amounts of bisulfite to the amounts of mercaptoethanol, namely a ratio of 3.25 in Example 1 and a ratio of 3.54 in Example 4 result in the same sulfonic acid group content of 0.25% in the resulting polymer while the intrinsic viscosity is increased from 1.01 in Example 1 to 1.2 in Example 4. Both examples relate to the same redox initiator system comprising sodium chlorate as the oxidising component and sodium bisulfite as the reducing component. In Example 1 the concentration of bisulfite (2.08%) is different from the concentration of bisulfite

used in Example 4 (1.7%), thus demonstrating that in order to obtain the desired result, i.e. a constant content of sulfonic acid end groups and a variation of the intrinsic viscosity over a relatively broad range it is indeed necessary to adjust the ratio of the concentrations of bisulfite and mercapto compound and not only to increase the concentration of mercapto compound in order to decrease the intrinsic viscosity. The same rule can be derived from Examples 2 and 3. Therefore, despite the fact that in these examples the amount of bisulfite remains constant and the observed change in molecular weight also corresponds to a variation of the mercaptoethanol concentration only, as submitted by the Appellant, from Examples 1 to 4 the teaching can be derived that by increasing the above ratio the molecular weight of the polymer is increased without altering the concentration of sulfonic acid end groups, i.e. the dyeability. This interpretation was expressly confirmed by the Respondent at the oral proceedings.

In the light of the preceding, the Board holds that the description provides sufficient information to enable a person skilled in the art to determine from the functional definition of the above ratio in Claim 1 the ratios required to obtain any combination of sulfonic acid group concentration and intrinsic viscosity within the ranges indicated in that claim.

Thus the problem underlying the patent in suit is regarded as being plausibly solved.

7. In the Board's view, the claimed process is not obvious in the light of the cited prior art.

7.1 As explained earlier, in document (4) the need for varying the molecular weight without altering the dyeability was already recognised and met. In this document it is also mentioned on column 2, lines 28 to 31 that mercaptans such as <u>inter alia</u> mercaptoethanol may be used in combination with the oxydisable sulfoxy compound, e.g. the bisulfite, as a part of the redox system, but not as a means to influence the molecular weight of the polymer obtained. This purpose of the mercapto compound may nevertheless have been common general knowledge at the filing date of (4). The use of a mercapto compound alone however is not sufficient to bring about the desired result, since according to the patent in suit it is necessary to use in combination

- the mercapto compound in an amount of 0.4 to 0.8% by weight, based on monomers,
- a certain <u>ratio</u> of the concentrations of bisulfite compound and mercapto compound, not a certain amount of mercapto compound alone, for bringing the sulfonic acid end group content and the intrinsic viscosity within the predetermined limits and
- a copper activator.

The Appellant alleged that the first of the above features alone is sufficient to solve the problem and that therefore the other two should be disregarded when assessing inventive step. However he has not provided any evidence that the ratio of bisulfite and mercapto compound is unimportant (see paragraph 6 above) and that other heavy metals than copper would also work. Therefore, in the Board's judgement the three features mentioned above have to be considered together and, since none of them is encompassed by (4), this document does not hint to the claimed alternative solution of the problem addressed therein.

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7.2 In (1), which is already acknowledged in the patent in suit (see page 2, lines 25 to 43), an iron salt as promoter and a concentration of mercaptoethanol within the range of 0.05 to 0.30% (based on total feed to the reactor) are recommended in order to produce a more colour stable polymer without substantial reduction of the conversion rate (see column 1 lines 38 to 50). Nothing is taught by this document about the influence of mercaptoethanol and a copper salt on the molecular weight of the resulting polymer. Thus, for the reasons set out in the preceding paragraph, this document neither alone nor in combination with (4) would have directed a person skilled in the art towards the claimed process.

7.3 In (3) it is stated on column 8, lines 4 to 14 that in a process substantially identical with that according to (1) the use of a catalyst promoter like iron, copper or silver and of chain transfer agents, especially mercaptans, which are sufficiently water soluble but not too volatile, is preferred, since they tend to increase the polymerisation rate. It is also stated that the use of these mercaptans results in the production of a polymer with lower molecular weight. However, this document does not suggest that it is decisive to use copper and not simply any heavy metal as an activator in combination with specified amounts of mercapto compounds and to adjust the ratios of sulfite and mercapto compounds. Moreover, this document is completely silent with respect to the problem of varying the molecular weight without altering the dyeability and cannot therefore teach a skilled person how to solve this problem at all, let alone in the way envisaged by the patent in suit.

7.4 Document (5), too, does not contribute to the solution of that problem. It recommends the combination of a mercapto compound and a copper activator for the same purpose as envisaged in (1), i.e. for obtaining a colorless and stable polymer (see column 6, lines 6 to 10). This document is again silent on the problem which is solved by the patent in suit but is concerned with the solution of quite different problems, i.e. the production of uniform compact, colourless, spherical polymer beads which are capable to produce gel-free spinning solutions, see column 2, lines 33 to 38.

In the Appellant's opinion it was only a matter of routine experimentation not involving an inventive step to find the appropriate amounts of sulfite and mercapto compounds required for the solution of the different problem of varying the molecular weight without altering the dyeability. However this argument must fail since (5) does not provide any incentive to carry out experiments using mercaptan concentrations within the claimed range. The Appellant's submission therefore at most may suggest that a person skilled in the art <u>could</u> have made such experiments. In the Board's jurisprudence however it is necessary to demonstrate that a person skilled in the art <u>would</u> have done so in the expectation to solve the envisaged problem. This however has not been demonstrated and therefore this argument, too, does not render the claimed process obvious.

7.5 Document (6) relates to a bulk polymerisation process being performed in the absence of water and therefore requiring a different redox initiator system comprising an organic hydroperoxide and sulfur dioxide or organic sulfites (see Claim 1). In this document it is stated that it has been tried to solve the problem also underlying the patent in suit, namely to vary the molecular weight without altering the sulfonic acid end group content by adding a mercapto compound to the reaction mixture, however the experiment practically failed (see page 3, Table 1) since large quantities of mercaptoethanol only produced a very small decrease in molecular weight. It was then found that a nitrogen containing compound had to be added in order to obtain the desired result (see page 3, Table 1 and page 5, Table 3). Thus, the reaction system is totally different from that according to the patent in suit and this document cannot therefore provide any incentive for considering the solution envisaged by the patent in suit performing the polymerisation of acrylonitrile in aqueous dispersion in the presence of a copper activator.

7.6 The Appellant's main argument against the inventive step of the claimed process however was that this process was obvious in the light of the common general knowledge as represented by (8) and (9).

This approach however does not take account of the fact that in catalytic processes it is normally impossible to make valid predictions of general applicability. This is especially so for catalytic polymerisations involving redox initiator systems in aqueous dispersion, i.e. in a two phase reaction medium. This is confirmed by the comparison of Examples I and II in (4) which demonstrates that different amounts of the components of the redox initiator system were required at different times of a year in order to bring about substantially identical results (see column 3, lines 36 to 40). Furthermore, it follows from the comparison of (3) and (6) that in aqueous dispersion systems and bulk polymerisation systems the effect of mercaptans on the molecular weight of polyacrylonitrile is quite different despite the use of redox initiators in both cases.

Document (8) relates to the manufacture of acrylonitrile fibers and teaches in general terms that polyacrylonitrile is made by suspension polymerisation initiated by redox systems, especially potassium persulfate/sodium bisulfite, the reaction being started by free radicals from the persulfuric acid as well as from the sulfite ion and accelerated by heavy metal ions. The growing polymer soon becomes insoluble and the reaction then proceeds at insoluble macroradicals under consumption of adsorbed monomers until it is terminated by bimolecular reactions (see page 328, left column, the first complete paragraph). In the first full paragraph on the right hand column the relevant properties of the product, including constant levels of molecular weight and dyeability, are listed. Nothing however is said in (8) about the methods and means by which these parameters are controlled.

Document (9) does not specifically relate to acrylonitrile fibers, but to polymers in general and sets out the mechanism of free radical initiated polymerisation, especially the mechanisms involved in chain initiation, chain termination and chain transfer. In this very general context the action of mercaptans as chain transfer agents is discussed. As it can be seen from the above analysis of (8) the polymerisation of acrylonitrile in aqueous suspension however is a very complex reaction in a two-phase system involving insoluble macroradicals. Therefore in the Board's view it is doubtful whether at all a person skilled the art without additional information would have combined the information available from (8) and (9). However even this combination does not exceed the knowledge already derivable from (3) published 1958. This knowledge however has not enabled the specialists for making acrylonitrile fibers for more than 20 years to solve a problem which has existed for about the same period of time, since (4) was already filed in 1955 and

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published 1962, a circumstance which in the Board's judgement rather points towards the presence of an inventive step than towards the obviousness of the claimed process.

This common general knowledge furthermore would at most have suggested to a person skilled in the art to consider the principle of using mercapto compounds for regulating the molecular weight. However, it has already been demonstrated by (6), see paragraph 7.5 above, that a simple application of the common general knowledge as represented by (9) was not sufficient to solve substantially the same problem of varying the molecular weight without changing the sulfonic acid end group content of an acrylonitrile polymer produced by bulk polymerisation. Thus this prior art would not even have suggested to combine the first and the third of the features set out in paragraph 7.1 above, let alone all three.

The Appellant's arguments therefore must fail also for this reason.

8. Claims 2 to 4 relate to preferred embodiments of the process according to Claim 1 and derive their patentability from this claim.

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 maintain the patent with claims, description and drawing as filed during the oral proceedings.

The Registrar: The Chairman

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22.2.89