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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 322/87 - 3.3.3

Anmeldenummer / Filing No / N° de la demande : 79 300 772.5

Veröffentlichungs-Nr. / Publication No / N° de la publication : 0 005 913

Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Method of improving the processability of rigid polymers; melts, solutions and shaped articles prepared according to this method.

Klassifikation / Classification / Classement : C08J 5/00

ENTSCHEIDUNG / DECISION
vom / of / du 25 April 1990

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /
Titulaire du brevet : Imperial Chemical Industries Limited

Einsprechender / Opponent / Opposant : Bayer AG
BASF AG

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Art. 54, 56, 69(1), 83, 84, 113(1)

Schlagwort / Keyword / Mot clé : "Novelty after incorporation of a disclaimer (yes)"
"Inventive step (yes) - prior art concerned with laboratory investigations"

Leitsatz / Headnote / Sommaire

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Case Number : T 322/87 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 25 April 1990

Appellant 01: (Opponent 01)
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Decision under appeal :
Interlocutory decision of the Opposition Division of the European Patent Office dated 30 June 1987 concerning maintenance of European patent No. 0 005 913 in amended form.

Composition of the Board :

Chairman : F. Antony
Members : C. Gérardin
W. Moser

Summary of Facts and Submissions

I. The mention of the grant of the patent No. 5 913 in respect of European patent application No. 79 300 772.5 filed on 4 May 1979 and claiming priority of 26 May 1978 of two earlier applications in Great Britain, was published on 20 July 1983 on the basis of 11 claims.

Claim 1 reads as follows:

"A method of improving the processability of rigid polymers which are capable of exhibiting thermotropic or lyotropic behaviour characterised in that a melt or solution of a rigid polymer is subjected to shear between relatively moving surfaces at an apparent shear rate of at least 100 sec^{-1}, the rigid polymer being in a thermotropic or lyotropic state prior to shearing or being caused to exhibit thermotropic or lyotropic behaviour as a result of the applied shear."

II. Notices of opposition were filed on 9 September 1983 by Appellant 1 (Opponent 1) and on 30 November 1983 by Appellant 2 (Opponent 2) against the grant of the patent on the grounds that the subject-matter of the patent in suit was not novel and did not involve an inventive step. It was further objected that, because of the presence of obscure and ambiguous wording in Claim 1 objectionable under Article 84 EPC, the requirements of Article 83 EPC were not met.

These various objections which were emphasised and elaborated in several later submissions were based essentially on the following documents:

(1) Journal of Polymer Science, Polymer Chemistry Edition, August 1976, Volume 14, No. 8, pages 2048 to 2058

(9) Der Extruder als Plastifiziereinheit, VDI-Verlag, 1977, pages 51 to 61

(12) Contemporary Topics in Polymer Science, 1977, Volume 2, pages 109 to 137.

III. By a decision of 30 June 1987, the Opposition Division maintained the patent in amended form on the basis of new claims filed on 3 May 1985, the amendments consisting in the introduction of the upper limit of 1000 s^{-1} for the shear rate and in the further indication that a shaped article was fabricated from the melt or solution while the viscosity was reduced as indicated.

Regarding the objection of insufficiency it was stated in the said decision that neither the selection of a suitable material, i.e. a polymer capable of exhibiting thermotropic or lyotropic behaviour, nor the treatment to which the starting material was to be subjected, would present any difficulty to the skilled man.

As far as novelty was concerned, although both the effect of various shear rates on the viscosity of liquid crystal polymers and the fabrication of test pieces from such polymers using a reciprocating screw injection moulding machine were disclosed in document (1), the fact that these features appeared in different sections of the article did not allow the conclusion to be drawn that the fabrication of test pieces inevitably involved subjecting the polymers to shear rates within the specific range as claimed; besides, no evidence for the performance characteristics of conventional extruders was provided in this respect.

Finally, although it was known that shear-induced orientation and the resulting viscosity reduction did not relax instantaneously, the fabrication of shaped articles utilising such shear effects could not be inferred from the prior art.

IV. The Appellants lodged notices of appeal against this decision on, respectively, 27 August 1987 and 28 August 1987 by telex confirmed in writing on 29 August 1987 and paid the prescribed fee at the same time. The arguments presented in the Statements of Grounds of Appeal filed, respectively, on 26 October 1987 and 30 October 1987, as well as in later submissions, can be summarised as follows:

The exact scope of the process as claimed was unclear regarding both the choice of the starting material and the amount of polymer subjected to shear; moreover, the wording of Claim 1 did not take the viscosity reduction caused by the increase in temperature into account.

As far as the objections of lack of novelty and inventive step were concerned, emphasis was put on the arguments already presented in opposition procedure. During oral proceedings held on 25 April 1990, a document summarising data from previously filed citations and showing the performance characteristics of conventional extruders available at the date of priority of the patent in suit was filed to demonstrate that shear rates of 100 to 1000 s^{-1} were actually usual in the art, thus implicitly used in the experimental studies referred to in document (1).

V. Following an objection of accidental anticipation by document (1) raised by the Board during oral proceedings the Respondent filed two sets of 7 claims each,

respectively as main and auxiliary requests. In Claim 1 according to the main request it was specified that "the use of a 6-oz New Britain 175-TP reciprocating screw machine and a Newbury HV1-25T reciprocating screw machine" was excluded; in Claim 1 according to the auxiliary request it was specified that "such shear as occurs in a reciprocating screw injection moulding machine" was excluded. Moreover, it was specified in these two claims that, as a result of the shear being applied, the viscosity was reduced relative to its value measured at a given temperature without pre-shear.

The Appellants strongly objected to the wording of these claims, especially to Claim 1 according to the main request, whose wording incorporated trade names of machines which were no longer available on the market and whose performance characteristics, therefore, were not exactly known.

VI. The essence of the arguments put forward by the Respondent can be summarised as follows:

The new claims were addressed to a skilled man who would have no difficulty in choosing a suitable polymer, nor in verifying the critical reduction in viscosity. Document (1) did not provide evidence that shear rates within the range of 100 to 1000 s^{-1} were actually employed. Even if some prior art documents showed that the orientation induced in liquid crystal polymer melts relaxes more slowly than in isotropic polymers, the advantages to be gained from preparing shaped articles by a process involving shear rates within a critical range had not been previously recognised.

VII. The Appellants requested that the decision under appeal be set aside and the patent be revoked in its entirety.

The Respondent requested that the patent be maintained on the basis of the claims filed as main request or on the basis of the claims filed as auxiliary request during oral proceedings.

Reasons for the Decision

1. The appeals comply with Articles 106 to 108 and Rule 64 EPC and are, therefore, admissible.

Main Request

2. The current version of the claims does not give rise to objections under Article 123 EPC.

As compared to the granted version, Claim 1 firstly differs by being directed to a method of fabricating a shaped article from rigid polymers; this object of the process is disclosed on page 3, lines 3 to 6 of the specification as granted, corresponding to page 4, lines 4 to 10 of the original documents. The change of "shear" into "pre-shear", which aims at identifying the first step of the process more clearly, is justified in view of page 4, lines 12/13 and 22, respectively page 7, lines 10 to 12 and 29, as well as the Tables of Examples 2, 3 and 5 to 9, respectively Examples 2, 3, 5, 6 and 8 to 10. The upper limit of 1000 s^{-1} for the shear rate is the preferred value mentioned on page 4, line 4 and Claim 9, respectively page 6, line 32 and Claim 10. The resulting reduced viscosity relative to the viscosity measured at a given temperature without pre-shear can be regarded as adequately supported in view of page 3, lines 3 to 9, respectively page 4, lines 4 to 15, and the values of the apparent viscosity with and without pre-shear in the

Tables of Examples 3, 5 and 6, respectively Examples 3, 5 and 6. Last, the exclusion of "the use of a 6-oz New Britain 175-TP reciprocating screw machine and a Newbury HV1-25T reciprocating screw machine" is an admissible disclaimer corresponding to the machines mentioned in document (1) on page 2056, Table VII, symbols a and b.

Claim 2 results from the combination of Claims 3 and 9 of the patent, corresponding to original Claims 4 and 10 of the application as filed. As to Claims 3 to 7, they correspond to Claims 4 to 8 of the patent as granted, respectively Claims 5 to 9 of the originally filed documents.

3. The introduction into Claim 1 of disclaimers for the use of reciprocating screw machines identified by their trade names makes the wording of this claim fundamentally unclear. The presence of such trade names is objectionable insofar as these words merely denote the origin of the machines, but not the actual shear rates resulting from the use thereof; moreover, since these machines are no longer available on the market, it is impossible to know their exact performance characteristics and therefore the exact scope of what is disclaimed, and consequently the scope for which protection is sought.

For this reason, the wording of Claim 1 is objectionable under Article 84 EPC, thus certainly not clearly allowable within the meaning of the Decision T 153/85, "Alternative claims/AMOCO", of 11 December 1986 published in OJ EPO 1988, 001. Following the principles set out in that decision regarding the filing of alternative claims at a late stage (points 2.1 and 2.2), the Board refuses to consider the set of claims submitted as Main Request.

Auxiliary Request

4. Claim 1 according to the Auxiliary Request differs from that according to the Main Request solely in that its disclaimer generally excludes the use of such shear as occurs in a reciprocating screw injection moulding machine. Such a disclaimer is permissible since it corresponds to the disclosure in document (1) (page 2054, paragraph 2, lines 4 to 6). Since Claims 2 to 7 are identical to Claims 2 to 7 according to the Main Request, they are formally acceptable for the reasons given above.

This means that no objection arises having regard to Article 123 EPC.

5. The objections raised by the Appellants under Article 83 EPC in combination with Article 84 EPC cannot be accepted, since neither the selection of an appropriate polymer, nor the treatment to which the latter is to be subjected would present any difficulty for the skilled man carrying out the teaching of the patent in suit.

5.1 From the wording of Claim 1 it appears that the patent in suit concerns a two-step method of fabricating a shaped article. The first step comprises shearing a polymer melt or polymer solution between relatively moving surfaces at an apparent shear rate of 100 to 1000 s^{-1} to reduce its viscosity; the second step consists in the fabrication of a shaped article while the viscosity still has a reduced value induced by shearing. In the Board's view, this cannot be construed as applying to "at least part of the melt", but relates unambiguously to the whole melt or solution. As to the starting material, it is generally defined in the description of the patent in suit as a solution or melt of a rigid polymer capable of exhibiting thermotropic or lyotropic behaviour, i.e. of a polymer

which is in a thermotropic or lyotropic state prior to shearing or which acquires this behaviour as the result of being subjected to shear within the range of 100 to 1000 s^{-1} (page 2, lines 33 to 37; page 3, lines 15 to 20). More specifically, the description mentions (page 3, lines 24 to 43) that rigid polymers suitable for use in the claimed process are based on the recurring unit

$$+ X\text{-}A +$$

wherein X is an aromatic radical, optionally substituted, and A represents either an atom or a group of atoms and assumes a configuration wherein its outgoing bonds are either parallel or form an angle of at least 120°; it further indicates the requirements in terms of glass-rubber transition temperature and intrinsic viscosity which the rigid polymers to be used as solutions and melts have to meet (page 3, lines 51 to 53; page 3, line 64 to page 4, line 1). On this basis the skilled man would have no difficulty in selecting a suitable rigid polymer. As to the treatment to which the thermotropic melt or lyotropic solution is subjected, it only involves conventional processing within a relatively narrow range of shear rates, as apparent from the various examples.

5.2 The fact remains that the wording of the claims, even interpreted in the light of the description within the meaning of Article 69 EPC, does not provide more than structural and physical requirements which are necessary, but not sufficient to identify which polymer melts and solutions are likely to respond to the shearing operation and which are not. As can be implied from the description (page 2, lines 4 to 6 and 55 to 58), not all rigid polymers are capable of exhibiting thermotropic behaviour, whether induced or natural. The feature "capable of exhibiting thermotropic or lyotropic behaviour" in the

preamble of Claim 1 corresponds in fact to a functional characterisation of the polymer. Such functional features are permissible in a claim if a more precise definition is not otherwise possible without restricting the scope of an invention, and if they provide sufficiently clear instructions for the skilled person to reduce them to practice without undue burden (cf. Decision T 68/85, "Synergistic herbicides/CIBA GEIGY", OJ EPO 1987, 228). In the present case, the aforementioned prerequisites are met, since, as the Respondent put forward in the observations filed on 3 May 1985 in opposition procedure (page 1, paragraph 3), all the skilled man, wishing to determine whether a given polymer falls within the claim or not, has to do is to observe the behaviour of that polymer at shear rates between 100 and 1000 s^{-1} between relatively moving surfaces.

5.3 Further, by specifying that as the result of the shear being applied the viscosity is reduced relative to the viscosity measured at a given temperature without pre-shear, the wording of Claim 1 avoids any ambiguity between temperature-induced viscosity reduction and pressure-induced viscosity reduction. It is thus clear that the subsequent fabrication of shaped articles is based on the latter phenomenon only.

5.4 In view of the foregoing, the objection of insufficient disclosure within the meaning of Article 100(b) EPC is unfounded.

6. The issue of novelty with regard to the teaching of document (1) was raised on the basis of new evidence submitted during oral proceedings.

6.1 Document (1) can be regarded as an investigation of various properties, especially mechanical properties, of

injection moulded polyethylene terephthalate (PET) modified with p-hydroxybenzoic acid (PHB). The melt viscosity of these copolymers is determined for various amounts of PHB and at shear rates between 1 and nearly 10^5 s^{-1} (page 2049, Figures 1 and 2). It appears that PET containing 60 mole% PHB is particularly shear-sensitive and that at a shear rate of 1000 s^{-1} the melt viscosity is less than 5% that of non-modified PET; this behaviour is attributed by the authors to the presence of liquid crystals (page 2049, line 9 to page 2050, line 6). It is further specified that the type of injection moulding machine used to mould copolyesters modified with up to 90 mole% PHB affects the mechanical properties of these polymers (page 2054, paragraph 2, lines 1 to 6). Three different injection moulding machines are mentioned, including two reciprocating screw injection moulding machines, for which the cylinder temperatures are indicated (page 2056, Table VII, footnotes a and b). This raises the question whether the various properties listed in Table VII were obtained using conditions falling within the scope of Claim 1 regarding both the shear rate and the screw back time.

6.2 The evidence whether the screw inevitably rotates at a speed sufficient to generate a shear rate within the terms of Claim 1 cannot be provided directly from the screw driven injection moulding machines mentioned in document (1), because these types of apparatus are no longer available on the market. It is therefore necessary to work out their performance characteristics on the basis of data and information related to the equipment commonly used at the date of publication of document (1), i.e. in 1976.

6.2.1 According to document (9) (page 52, equation (31)), the shear applied to the whole melt or minimum shear rate in a screw injection moulding machine is given by the equation

$$\dot{\gamma} = f_g \frac{\pi D n}{h}$$

wherein D and h are respectively the diameter and the pitch of the screw, n is the screw speed (rpm) and f_g is a correction factor which takes the shear rate distribution in the main channel into account.

This equation was applied to the screw driven injection moulding machine described in a technical information sheet published on 21 November 1974 and submitted by the Appellants during oral proceedings. The experts representing the parties agreed that a value of 1.0 or 1.5 for f_g would be a fair approximation and that 120 rpm for n would be reasonable, but took different values for the ratio D:h. Whereas the Respondent assumed that this ratio would be about 8 on the basis of the drawing on page 3, the Appellants took the actual figures for these parameters from the Table on page 4 of this document. In the Board's view, the latter interpretation is undoubtedly correct, since the Respondent's calculation is based on a schematic representation of the screw which does not necessarily respect the exact proportions thereof.

On the basis of the Appellant's calculation, one obtains shear rates of at least 150 s^{-1}, which falls within the range envisaged by the Respondent.

6.2.2 From a procedural point of view the Respondent raised the question of the admissibility of this new evidence during oral proceedings. In the Board's view, although the technical information sheet is a new document as such, it does not disclose more than the features of a standard screw driven injection moulding machine available at the date of publication of document (1). The new document only helps to demonstrate that the reciprocating screw machines

referred to in document (1) could not be distinguished by their performance characteristics from conventional screw injection moulding machines, as was repeatedly alleged by the Appellants. Moreover, it must be emphasised that the result of the calculations made during oral proceedings is not the basis of a new argument or objection, but merely confirms the results obtained by using formula (31) of document (9) already filed in the Annex to the Statement of 20 December 1986.

As the Board appreciated in the non-published decision T 324/88 (points 5 and 8) of 8 February 1989, a submission filed at a late stage which merely confirms an evidence already on file cannot be regarded as a new or late filed evidence and therefore does not contravene the requirements of Article 113(1) EPC. For this reason, the Board regards the evidence provided during oral proceedings not as a new argument, but as a confirmation of results previously filed.

6.2.3 The Board is aware that the evidence provided by the Appellants is nothing more than an indirect estimation of the shear rates produced by the reciprocating screw injection moulding machines mentioned in document (1) on the basis of the technical features of similar machines. However, the demonstration makes it sufficiently evident that the shear rate to which the melt is subjected during the moulding operation described in document (1) comes within the terms of Claim 1.

6.3 As the Respondent put forward in the counterstatement filed on 23 March 1990 (page 3, paragraph 4), in the screw driving injection moulding process the screw is used as a pump to deliver material into a reservoir before it is injected into the mould. After a shot has been injected into the mould, the screw is screwed back to pump the next

shot into the reservoir. It is during the screw back step that shear between relatively moving surfaces is applied. In conventional injection moulding, the screw back time is of the order of one or two minutes, which means that the first portion of the melt to enter the reservoir has a period of one or two minutes to relax prior to injection into the mould.

This period should be compared with the actual relaxation period, i.e. the time interval before the viscosity reverts to its original pre-sheared level. According to the patent in suit (page 3, lines 10 to 14), typical relaxation periods can vary between 1 and 10000 seconds, but are normally in the range of from 10 to 100 seconds.

These figures are of the same order of magnitude as the screw back time and overlap to a large extent, which means that the moulding operation is carried out whilst the viscosity may well be in the viscosity reduced state. Contrary to the Respondent's argument in the above counterstatement, abnormally short screw back times are thus not at all necessary to operate within the terms of Claim 1.

6.4 In view of these conclusions, it must be assumed that in the injection moulding process disclosed in document (1) the reciprocating screw machines were operating at a speed sufficient to generate a shear rate within the range as claimed and that the treated melt was fabricated whilst the shear induced viscosity reduction was still available. The subject-matter as defined in Claim 1, according to which the shear as occurs in a reciprocating screw injection moulding machine is explicitly excluded, is thus novel.

7. It still remains to be examined whether the subject-matter of the patent in suit involves an inventive step with regard to the teaching of the cited documents.

7.1 The patent in suit concerns a method of fabricating a shaped article from rigid polymers which are capable of exhibiting thermotropic or lyotropic behaviour. The fabrication of shaped articles is disclosed in document (1) which the Board regards as the closest state of the art. As noted above, that document deals first with the variation of melt viscosity according to the degree of modification of PET with PHB as well as shear rates applied (Figures 1 and 2), then with various aspects of injection moulding, including the mechanical properties of the copolyesters (pages 2050 to 2057). Emphasis is put on tensile strengths and flexural moduli of injection moulded specimens of PET modified with 40 to 90 mol% PHB (page 2050, paragraph 3 and Table II). Even if one regards the two sections of this article as relating to the successive steps of a single process, nothing suggests a necessary continuity between these steps, i.e. that the moulding operation should follow the mechanical treatment of the polymer immediately. In this respect, thus, the prior art process is merely conventional.

With regard to this teaching the problem underlying the patent in suit may thus be seen in providing a process wherein the processability of the polymer would be improved without impairing the good mechanical properties of the moulded article.

According to the patent in suit this problem is solved by subjecting a melt or solution of a rigid polymer capable of exhibiting thermotropic or lyotropic behaviour to pre-shear between relatively moving surfaces at a specific shear rate between 100 and 1000 s^{-1} and carrying out the

moulding operation by normal techniques while the viscosity is still reduced as a result of the applied shear.

In view of the undisputed advantages put forward in the description (page 4, lines 23 to 26) and the examples in the patent in suit, the Board is satisfied that the above defined technical problem is plausibly solved.

7.2 The data relative to the melt viscosity for various shear rates mentioned in document (1) only record the stage of material whilst the particular shear rate is applied, but give no indication of what happens when the shear rate ceases to be applied. In particular, they do not suggest that the material retains a memory of pre-shearing history; moreover, there is no discussion of the relaxation period and thus no recognition that the benefit of reduced viscosity persists when the shear is removed (Appendix to Respondent's observations filed on 20 September 1984 during opposition procedure, page 1, paragraph 3). For this reason, this document cannot induce the skilled man to exploit the benefit of low viscosity in injection moulding processes.

7.3 It is not disputed that document (12) mentions in general terms that the orientation induced in liquid crystal polymers relaxes more slowly than in isotropic polymers (page 132, paragraph 2). However, as argued by the Respondent during oral proceedings in opposition procedure (minutes, page 6, paragraph 2), it is not clear whether the shear forces referred to in the middle of the paragraph in the above document are directed to the phenomenon of pressure driven flow as the result of the passage of the melt or solution through the injection nozzle, or to relatively moving surfaces. Moreover, in the Board's view, even if the skilled man considered

relatively moving surfaces to produce the shear forces as required, there is no reason why he should not follow the teaching of document (1) regarding the advantage resulting from the use of reciprocating screw injection moulding machines. Furthermore, it is specified in document (12) that the observed average molecular orientation in a moulded article depends actually upon the processing conditions which may be the polymer's melt temperature, the mould's temperature and mass, the article's shape and dimensions, and the polymer's flow pattern and injection rate into the mould (last sentence of the paragraph); this can only mean that the authors of this article failed to recognise the advantages to be gained by preparing shaped articles by means of a process involving the step of preshearing the melt at a shear rate between 100 and 1000 s^{-1} between relatively moving surfaces.

For these various reasons, document (12) neither in isolation nor in combination with document (1) can suggest the operative features of the method as claimed.

7.4 The correlation between persisting reduced viscosity as a result of applied shear and improved processability is not to be found in any of the numerous documents relied upon by the Appellants. The fact that none of the nearly thirty documents on file is a patent document, is evidence that the prior art was more concerned with laboratory investigations on the basis of theoretical considerations than with the practical aspects of moulding operations such as processability.

7.5 For these reasons, in the Board's view, the subject-matter of Claim 1 involves an inventive step.

8. Claim 1 being allowable, the same applies to Claims 2 to 7 which represent preferred embodiments of Claim 1, the patentability of which is supported by that of the main claim.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The main request is rejected.

3. The case is remitted to the first instance with the order to maintain the patent on the basis of Claims 1 to 7 submitted during oral proceedings as auxiliary request and a description to be adapted accordingly.

The Registrar: The Chairman:

M. Beer F. Antony