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Bezeichnung der Erfindung / Title of invention / Titre de l'invention : Gas-phase olefin polymerisation process and system for operating it.

Klassifikation / Classification / Classement : C08F 10/00

ENTSCHEIDUNG / DECISION

vom / of / du 29 March 1989

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent / Titulaire du brevet : ICI PLC

Einsprechender / Opponent / Opposant : BP Chemicals Limited

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Art. 54, 56

Schlagwort / Keyword / Mot clé : "Novelty (yes) - use of a known apparatus" "Inventive step (yes) - process feature not suggested by the prior art".

Leitsatz / Headnote / Sommaire

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Case Number : T 329 /87 - 3.3.1

DECISION
of the Technical Board of Appeal 3.3.1 of 29 March 1989

Appellant :
(Opponent)

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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. 0 004 417 in amended form.

Composition of the Board :

Chairman : K. Jahn
Members : C. Gérardin
J. Stephens-Ofner

Summary of Facts and Submissions

I. The mention of the grant of the patent No. 4417 in respect to European patent application No. 79 300 292.4 filed on 26 February 1979 and claiming priority of 23 march 1978 from an earlier application GB 11676/78 was announced on 14 September 1983 on the basis of 9 claims.

The two independent claims read as follows:

Claim 1: "A gas phase olefin polymerisation apparatus including a gas-phase polymerisation reactor, wherein optionally a stirrer is present in the gas-phase polymerisation reactor, characterised in that the polymerisation apparatus also includes means to detect, within the polymerisation reactor, an undesirable or a potentially undesirable, reaction condition which is an excessive temperature and/or pressure or is failure of any stirrer present in the gas-phase polymerisation reactor, a source of gaseous carbon dioxide, means connecting said source of gaseous carbon dioxide to the polymerisation reactor, means for preventing the flow of gaseous carbon dioxide to the reactor during normal operation of the polymerisation reaction and means operative by said detecting means to automatically permit the flow of gaseous carbon dioxide to the reactor only upon detecting said undesirable, or potentially undesirable, reaction condition and to thereby at least reduce the rate of polymerisation."

Claim 6: "A process of operating an olefin polymerisation process in a gas-phase polymerisation reactor using a catalyst system based on a transition metal compound, characterised in that the process is monitored to detect an undesirable, or potentially undesirable, reaction

.../...

condition which is an excessive temperature and/or pressure, or is failure of any stirrer present in the gas-phase polymerisation reactor, the process is operated in the absence of any gaseous carbon dioxide as long as said undesirable, or potentially undesirable, reaction condition does not occur and, when the undesirable, or potentially undesirable, reaction condition is detected, gaseous carbon dioxide is introduced into the polymerisation reactor in a quantity which is sufficient to at least reduce the rate of the polymerisation reaction."

II. The Appellant (Opponent) filed an opposition against the grant of the patent on 14 June 1984 on the grounds of lack of novelty of the apparatus claim with regard to the teaching of following document

(1) FR-A-1 386 079

and lack of inventive step of the process claim with regard to the teaching of following documents

(2) US-A-3 269 997

(3) Proceedings of the International Congress of Catalysis, Miami Beach, 20 to 26 August 1972, Comparative study of the mechanism of olefin polymerisations in the presence of one- and two-component systems based on $TiCl_2$ by Y.I. Ermakov, published in 1972, volume 5, pages 23-399 to 23-406

(4) Die Makromolekulare Chemie, 1977, volume 178, pages 261 to 266

(5) BE-A-628 272.

A further ground of opposition was that the wording of Claim 1 contravened Article 123(2) EPC.

III. By a decision dated 30 June 1987 the Opposition Division maintained the patent in amended form, the amendment consisting basically in the incorporation into Claim 1 of Claim 3 as granted.

The reasons in this decision can be summarized as follows:

The objection under Article 123(2) EPC has been overcome by the combination of Claims 1 and 3.

The subject-matter of Claim 1 is novel since the apparatus described in document (1) does not comprise means to detect failure of rotation of the stirrer. This feature, which is not suggested by the prior art, is advantageous in that it provides prompt information that undesirable reaction conditions are about to occur; therefore, it is inventive as well.

The process according to Claim 5 is novel since document (1) does not mention the addition of carbon dioxide when an undesirable reaction condition occurs. Furthermore, this process is inventive with regard to the gas-phase polymerisation process described in FR-A-2 307 822 (document (8)) wherein, as in the other documents cited dealing with this type of polymerisation, the use of a quenching agent to control the temperature is not disclosed, let alone the selection of carbon dioxide as quenching agent.

IV. A notice of appeal was lodged against this decision on 27 August 1987 with payment of the prescribed fee. The statement of grounds of appeal was filed on 10 November 1987.

Regarding novelty, the Appellant argued that the nature of the gas was not a feature of the apparatus, since the apparatus remained the same whatever gas was used in it. In addition, the Appellant maintained that novelty could not be derived from the fact that means to detect failure of rotation of the stirrer were not explicitly described in document (1), because such means were self-evident.

As far as the inventive step was concerned, since the Opposition Division admitted that the use of carbon dioxide as quenching agent was known from document (2), this feature could not by itself contribute to the inventiveness of the process. In this regard, the difference construed by the Opposition Division between gas-phase processes and other processes was artificial, as was apparent from the declaration of Dr. Peter Tait. Consequently, the teaching of the other documents on file, wherein the use of carbon dioxide as quenching agent was mentioned, became relevant and made the selection claimed by the Respondent even more obvious.

V. The arguments presented by the Respondent in the counter-statement filed on 23 March 1988 and during oral proceedings held on 29 March 1989 can be summarized as follows:

The Appellant's argument, that a source of carbon dioxide was not an apparatus feature, was mere quibbling, since there had to be something which introduced the gas under the conditions specified in the claims. As to the use of carbon dioxide in document (2), this teaching was totally irrelevant, since it related to discolorising polymers.

The same applied to the other documents already cited in opposition proceedings, which gave contradictory information about the influence of carbon dioxide on polymerisation catalysts.

There were substantial differences between gas-phase and liquid-phase processes as regards quenching the reaction in an emergency leading to the formation of fused lumps of material. Furthermore, in a liquid-phase process, the use of a signal from the stirrer as a means of initiating the injection of carbon dioxide would be unacceptable, since by the time there was a significant effect on the stirrer, the reaction would be well out of control. The situation was entirely different in a gas-phase process, as shown in Example 2 of the patent in suit.

VI. During oral proceedings, a new set of claims was submitted, wherein Claim 1 corresponded to Claim 6 as granted (process claim) and Claim 5 was drafted as a dependent apparatus claim, but otherwise corresponded to Claim 1 of the version as amended by the Opposition Division for carrying out the process as claimed in any of the proceedings claims.

VII. The Appellant requested that the impugned decision be set aside and the patent be revoked in its entirety.

The Respondent requested that the appeal be rejected and the patent be maintained on the basis of the documents submitted during oral proceedings.

At the end of oral proceedings the decision was announced that the patent was maintained as requested by the Respondent.

Reasons for the Decision

1. The appeal complies with Article 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 EPC.

Claim 1 corresponds to the independent process Claim 6 as granted and dependent process Claims 2 to 4 correspond to original Claims 7 to 9. The apparatus Claim 5 is merely a combination of Claims 1 and 3 as granted which has been drafted as a dependent claim, whilst Claims 6 to 8 correspond to original Claims 2, 4 and 5.

3. The patent in suit concerns a gas-phase olefin polymerisation process and a system for operating it. Such a process is disclosed in document (8) which the Board, like the Opposition Division, considers as the closest prior art. This process is characterised by a high heat transfer to the walls of the polymerisation reactor (page 4, line 24 to page 5, line 12). Cooling of the polymerisation is normally effected by circulating a cooling liquid, such as water, through the walls of the reactor; additional cooling may be obtained by introducing the monomer into the reactor as a liquid which vapourises within the reactor by extracting the heat of evaporation from the reactor and its contents. Further cooling may be achieved by locating additional surfaces, for example cooling coils, within the polymerisation reactor in the vapour space above the solid polymer. The temperature and pressure within the polymerisation reactor are continuously monitored and the rate of supply of coolant and monomer may be varied in dependance on the measured temperature and/or pressure. In practice, the rate of supply of coolant can be adjusted in dependance on the

temperature, whilst the rate of monomer supply can be adjusted in dependance on the pressure. Although this controlled heat exchange process works satisfactorily as long as the polymerisation reaction proceeds in a steady state, it could not cope with a rapid increase of temperature and/or pressure resulting from run-away reaction conditions or the failure of the stirrer. Such localised overheating of the polymer within the reaction bed would lead to the overheating and fusing of the polymer particles into a large lump mass which would be detrimental to the continuity of the gas-phase polymerisation. The problem underlying the patent in suit can thus be seen in providing a gas-phase olefin polymerisation process which can cope with such emergency conditions without impairing the quality of the polyolefin.

The Respondent proposes to solve this problem essentially by introducing carbon dioxide into the polymerisation reactor in order at least to reduce the rate of polymerisation or even to quench the reaction altogether. More specifically, the polymerisation process claimed by the Respondent is carried out under normal conditions in the absence of carbon dioxide and is monitored to detect an undesirable, or potentially undesirable, reaction condition or the failure of the stirrer; when any such reaction condition is detected, gaseous carbon dioxide is introduced into the polymerisation reactor in a quantity sufficient to reduce the rate of the polymerisation reaction.

In view of the Examples 1 to 4 in the patent in suit, the Board is satisfied that the above defined technical problem has been plausibly solved.

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

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

5.1 The use of an inert gas has already been suggested in order to control the temperature in a polymerisation reactor. In document (1), which deals with a slurry polymerisation process of olefins, the removal of heat is achieved by means of an inert gas which is introduced into the reactor, is then eliminated together with olefin vapours from the reactor, and is subsequently separated from these vapours, which are then recycled into the reactor, whilst the inert gas after compression is recycled into the reactor so as to maintain the temperature nearly constant inside it (page 1, column 2, paragraph 2 to page 2, column 1, paragraph 1; page 3, column 1, paragraph 3). According to a preferred embodiment, the circulation of the gas is controlled by means of a valve, which automatically opens when the temperature exceeds a certain value and closes when the temperature drops (page 2, column 2, lines 3 to 10). The inert gas is defined as a non-condensable gas free of oxygen or any substance which would interfere with the polymerisation; it is preferably selected from hydrogen, nitrogen, argon, helium and neon (page 2, column 2, paragraph 5). In the case of hydrogen, this compound will act as well as a chain transfer agent controlling the molecular weight of the polymer, but its main function remains the removal of heat (page 3, column 1, lines 2 to 6).

The essential feature of this process is thus the removal of heat under normal operating circumstances by circulating a non-condensable gas; this gas is cooled and routinely recirculated to the polymerisation reactor to assist in cooling, not to slow down or stop the polymerisation reaction. This mechanism can be regarded as essentially continuous, unless the temperature becomes too low. It is self-evident that this disclosure, which is neither concerned with the emergency situation resulting from run-away reaction conditions, nor with the risk of overheating polymer particles in a gas-phase polymerisation reactor, cannot contribute to the solution of the above defined technical problem.

Even if the skilled man regarded this teaching as relevant to the problem underlying the patent in suit and tried to find a solution to run-away conditions in the reactor along this line, he would choose a cooling gas which would be basically inert and have no influence on the catalyst activity, except perhaps on the molecular weight, and would reduce the monomer concentration in the gas phase, first by increasing the relative amount of inert gas as much as possible, then, as a last resort, by closing the inlet pipe of the monomer. Clearly, none of these steps corresponds to the subject-matter of the patent in suit.

5.2 The Appellant regards the use of carbon dioxide to deactivate the catalysts suitable for the gas-phase polymerisation of olefins mentioned in document (2) as pointing at the solution claimed by the Respondent.

According to this document, olefin polymers which are normally obtained as dark coloured products are treated with specific agents in order to produce white or colourless polymers (column 1, lines 10 to 13 and 41/42). These treating agents are said to have both a polymer-decolorizing and a catalyst-inactivating action (column 2, lines 33/34); carbon dioxide is quoted among a long list of compounds suitable for this purpose (column 2, line 69 to column 3, line 46).

Experimental results, however, do not suggest optimal efficiency for carbon dioxide. According to Example III, a small amount of solid carbon dioxide is added to 30 grams of a dark brown solid ethylene polymer swollen with benzene and the mixture is allowed to stand overnight and warm up to room temperature. After being filtered from the benzene and dried in a vacuum oven, the colour of the dry polymer is a light tan. By contrast, a contact for about 30 minutes between the same polymer and several other treating agents yields a polymer having a residual colour corresponding to the colour of the treating agent (Examples II and IV) or a white polymer (Examples V to VIII); similarly a white polyethylene can be obtained from a dark brown product by treatment with butyl alcohol (Example X) or from a dark product by treatment with methyl alcohol and aqueous hydrochloric acid (Example XI). Such results cannot, obviously, be an incentive to select carbon dioxide as a treating agent for deactivating purposes.

Moreover, carbon dioxide and methyl alcohol which are generally described in document (2) as appropriate catalyst-inactivating and polymer-decolorizing treating agents are not at all equivalent in the context of the process claimed in the patent in suit. In the latter, the introduction of carbon dioxide into the autoclave gives rise to a rapid fall in the temperature without any build-up or lump formation after the stirrer is stopped (Example 4); by contrast, the addition of methanol to inhibit the catalyst results in a drop in the temperature of the vessel contents as expected, but the polymer product is then found to be in the form of a fused lump formed after the stirrer had been stopped (Comparative Example A).

In fact, both in view of the difference in the way of incorporating the treating agent, i.e. into an already prepared product according to document (2) and during the polymerisation reaction according to the patent in suit, and of the rather poor results obtained for carbon dioxide in this prior art, it is doubtful whether the skilled man would even consider this teaching as being relevant.

5.3 In addition, the documents wherein the ability of carbon dioxide to inhibit olefin polymerisation reaction is mentioned, all underline the shortcomings of this compound or the side effects caused by it.

5.3.1 Document (5) suggests the carrying out of olefin polymerisations in the presence of carbon dioxide and a ternary catalyst system comprising

(a) a metal, a hydride or an organometallic compound of a metal from the groups IV, V and VI of the Periodic Table

(b) an anorganic compound of a polyvalent metal at least trivalent

(c) a metal halogenide wherein the metal is from the groups III or V of the Periodic Table

(Claim 1). The use of carbon dioxide in amounts corresponding to molar ratios carbon dioxide:polyvalent metal between 20 and 300 results in an increased activity, a slight reduction of the molecular weight and a narrower molecular weight distribution (page 2, paragraph 4 to page 3, paragraph 3; Figure). Detrimental effects on the catalyst activity appear only for larger amounts of carbon dioxide (page 2, paragraph 5; page 4, Table, last line; page 5, paragraph 3); these amounts which correspond to the deactivation of the catalyst are considerably higher than in the patent in suit which specifies that effective quenching can be achieved by the addition of carbon dioxide in an amount of at least five times, on a molar basis, relative to the total effective catalyst system (compare column 3, lines 48 to 52).

During oral proceedings, both parties agreed that this dual effect of carbon dioxide on the catalyst activity, i.e. activation for molar ratios between 20 and 300 and inhibition beyond this limit, was probably related to the unusual combination of titanium, tin and aluminium in the only exemplified catalyst system, and that this teaching was actually superseded by that of document (4).

5.3.2 Document (3) is a comparative study of the mechanism of olefin polymerisation in the presence of a catalyst system comprising titanium dichloride, optionally together with diethyl aluminium chloride. For the determination of the number of propagation centers and of the propagation rate constant, the method of stopping the polymerisation with carbon monoxide and carbon dioxide labelled with ^{14}C was used (page 399, Abstract). These inhibitors were introduced in the amount necessary for the complete stopping of polymerisation (page 399, point 2, Experimental procedure). The kinetic curves for the reaction of polymerisation provide clear evidence of the ability of these compounds to quench the reaction almost completely (page 400, Figure 1).

5.3.3 Document (4) can be regarded as an attempt to apply the investigative technique mentioned in document (3) to the polymerisation of ethylene and propylene, using classical heterogenous Ziegler-Natta catalytic systems (page 261, paragraph 3 to page 262, paragraph 3). The authors of this academic article conclude that the suitability of an agent for the determination of active centers depends not only on its selectivity and high efficiency, but also on the irreversibility on the reaction with the active center. The kinetic curves of Figure 1 which show the influence of carbon monoxide and carbon dioxide upon the rate constant of propylene polymerisation, demonstrate that this latter requirement is not met; in practice, upon the removal of the quenching agent by evacuation, the polymerisation activity is restored to at least 80% of the initial value (page 262, paragraph 4 to page 263, paragraph 1). This reversibility suggests that the retardation influence on olefin polymerisation should be interpreted as a sorption equilibrium (page 263, paragraph 7).

This conclusion would, clearly,deter the skilled man from using carbon dioxide as quenching agent for olefin polymerisation reactions catalyzed with such catalyst systems. In view of the problem to be solved, i.e. in view of the necessity to cope with the high temperatures associated with run-away conditions, it is self-evident that the skilled man would not want an agent which would give rise to a reversible situation like a sorption equilibrium, which is a highly temperature dependent phenomenon, but would choose an inhibitor which eliminated any possibility of reactivation of the catalyst. Apart from the fact that carbon dioxide is even less active than carbon monoxide (page 263, paragraph 2) and that any excess thereof could lead to side reactions (page 265, paragraph 1), the mechanism proposed for the interaction between the quenching agent and the active centers of the catalyst system would thus invite the skilled man to look for a conventional active catalyst poison, like an alcohol stream or a sulphur compound, as indeed the Respondent argued during the oral proceedings.

5.4 In conclusion, neither the experimental results from document (2) which demonstrate that carbon dioxide is less efficient than other deactivating agents well known in the art, nor the theoretical considerations in document (4) from which it appears that an effective inhibition should not involve a reversible adsorption of the quenching agent on the active centers of the catalyst system, would be an incentive to select carbon dioxide for use in emergency conditions. Furthermore, the use of carbon dioxide would be incompatible with the teaching of document (1) wherein normal polymerisation conditions are insured by continuous removal of heat with an inert gas, not by preventing formation of heat by inhibiting the reaction with a quenching agent. For all these reasons, the use of carbon dioxide in the process claimed by Respondent cannot be held to be obvious. On the contrary, since such use confers an unexpected advantage over the conventional quenching agents regarding lump formation in the reactor, the requirements for inventive step are met.

6. Claim 1 being acceptable, the same applies to the dependent process Claims 2 to 4, which merely relate to preferred embodiments of the process according to Claim 1 and derive their patentability from this claim.

7. The Appellant has repeatedly objected the lack of novelty of the apparatus claim with regard to the teaching of document (1). Although novelty could not be acknowledged for an independent claim by merely specifying that the apparatus contains a source of carbon dioxide, since the nature of the gas is a process feature, not an apparatus

feature, this objection cannot be maintained against the present dependent form for the apparatus claim. The Board regards Claim 5 as a use claim of a known apparatus for carrying out the patentable process according to Claim 1; because of this restrictive interpretation of the apparatus claim, Claim 5 and consequently the subsequent apparatus Claims 6 to 8 derive their patentability from that of Claim 1 and are thus acceptable as well.

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 on the basis of the documents (description, 8 claims, 1 drawing) submitted at oral proceedings.

The Registrar:

F. Klein

The Chairman:

K. Jahn

Cb

27.iii.