DECISION
of 9 December 2002

Case Number: T 0633/98 - 3.3.3
Application Number: 94306168.9
Publication Number: 0640627
IPC: C08F 210/16

Language of the proceedings: EN

Title of invention:
Ethylene-alpha-olefin copolymers and molded article thereof

Applicant:
Sumitomo Chemical Company, Limited

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 83, 84, 123(2)

Keyword:
"Sufficiency (yes)"

Decisions cited:
-

Catchword:
-
Case Number: T 0633/98 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 9 December 2002

Appellant: Sumitomo Chemical Company, Limited
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 23 December 1997
refusing European patent application
No. 94 306 168 pursuant to Article 97(1) EPC.

Composition of the Board:
Chairman: R. Young
Members: C. Idez
U. Tronser
Summary of Facts and Submissions

I. European patent application No. 94 306 168.9, filed on 22 August 1994, claiming the priority of the earlier Japanese patent application No. 207 496/93 of 23 August 1993 and published under No. 0 640 627 on 1 March 1995, was refused by a decision of the Examining Division announced orally on 25 November 1997 and issued in writing on 23 December 1997.

II. The decision was based, as main request, on Claims 1 to 8 as originally filed and, as auxiliary request, on a set of Claims 1 to 8 filed on 28 October 1997. Claim 1 read as follows:

"An ethylene-\(\alpha\)-olefin copolymer in which
(A) a density is 0.870 to 0.945 g/cm\(^3\),
(B) a relation between an activation energy of flow \(E_a\) (J/mole K) obtained by measurement of viscoelasticity at at least three temperatures in the molten state and a melt flow rate MFR (g/10 min) satisfies the following equation (1):
\[
\log E_a = 4.6 - 0.04 \times \log MFR \quad (1)
\]
(C) a coefficient \(C_x\) of variation of chemical composition distribution represented by the following equation (2) is 0.40 to 0.80:
\[
C_x = \sigma/SCB_{ave}(2)
\]
wherein \(\sigma\) is a standard deviation of chemical composition distribution (1/1,000 C) and \(SCB_{ave}\) is the average of short chain branchings per 1,000 C (1/1,000 C), and
(D) a ratio of a weight average molecular weight \(M_w\) to a number average molecular weight \(M_n\) (\(M_w/M_n\)) is 3 to 20."
Claims 2 to 7 were dependent on Claim 1. Independent Claim 8 related to a molded article formed from the ethylene-alpha-olefin copolymer according to Claim 1 or 2.

Claim 1 of the auxiliary request read as follows:

"A process for producing an ethylene-α-olefin copolymer in which
(A) a density is 0.870 to 0.945 g/cm³,
(B) a relation between an activation energy of flow $E_a$ (J/mole K) obtained by measurement of viscoelasticity at at least three temperatures in the molten state and a melt flow rate MFR (g/10 min) satisfies the following equation (1):
$$\log E_a \geq 4.6 - 0.04 \times \log MFR \quad (1)$$
(C) a coefficient $C_x$ of variation of chemical composition distribution represented by the following equation (2) is 0.40 to 0.80:
$$C_x = \sigma/SCB_{ave} \quad (2)$$
wherein $\sigma$ is a standard deviation of chemical composition distribution (1/1,000 C) and $SCB_{ave}$ is the average of short chain branchings per 1,1000 C (1/1,000 C), and

(D) a ratio of a weight average molecular weight $M_w$ to a number average molecular weight $M_n$ ($M_w/M_n$) is 3 to 20; comprising copolymerising ethylene with an α-olefin in the presence of an olefin-polymerising catalyst comprising (a) a titanium compound having at least one titanium-nitrogen bond, (b) an organomagnesium compound and (c) a halogen-containing aluminium compound."
Claims 2 to 7 were dependent on Claim 1. Independent Claim 8 related to a molded article formed from the ethylene-alpha-olefin copolymer produced according to Claim 1 or 2.

III. The Examining Division refused the application on the grounds that it did not meet the requirements of Article 83 EPC. More precisely, the decision stated that the parameter (B) of the pending Claims 1 of both requests could not be determined by a person skilled in the art using only the disclosure of the application and his general knowledge, since the conditions, i.e. the temperature and the load, under which the melt flow rate MFR should be measured, were not specified.

The decision further stated that the parameter (C) of the pending Claims 1 of both requests could not be determined by a person skilled in the art using only the disclosure of the application and his general knowledge, in particular, for the following reasons:

(a) the formula (15) did not give a value for $H_1$, (b) the formula (17) gave values for the SCBi's which were only dependent on $T_i$ and independent of the copolymer in question, (c) the method for determining the SCB in the document (iii), i.e. a partial English translation of "Rheology, pages 52 to 54, edited by Nippon Rheology Society, published by Kobunshi Kankokai in 1992" was different from the method for determining the SCBi and SCB mentioned in the description of the application, (d) it was not indicated in the description what kind of standard deviation of what plotting of the SCBi's was meant in the formula of parameter E (sic), and (e) the SCBi was only a calculated point.
The Examining Division came therefore to the conclusion that a person skilled in the art could not carry out the invention without knowing how to determine all the parameters of Claim 1 as he did not know which copolymers were inside the scope of this claim and which were outside the scope of this claim.

IV. A Notice of Appeal against the decision was lodged on 19 February 1998. The prescribed fee was paid on the 20 February 1998. The Statement of Grounds of Appeal was filed on 22 April 1998.

The arguments presented by the Appellant in the Statement of Grounds of Appeal may be summarized as follows:

(i) Concerning parameter (B):

(i.1) It was indicated in the application that the MFR was determined according to the method prescribed in the standard JIS K6760. According to this standard, the melt flow rate should be carried out in accordance with the standard JIS K 7210. English translations of both standards were annexed to the Statement.

(i.2) As indicated in JIS K 7210, the test conditions for MFR, should be, as a rule, Condition 4, i.e temperature 190°C and load 2.16 kgf.

(ii) Concerning parameter (C):

(ii.1) The references made in the decision under appeal to the formulae (15) and (17) were obscure. It was suspected that the Examining Division intended to refer to formulae (3) and (5).
(ii.2) $H_i$ represented an individual concentration, whose numerical value was given by formula (3). $R_{H_i}$ was the relative concentration.

(ii.3) The values of $SC_{Bi}$ were in practice only dependent on $T_i$ and independent of the nature of the copolymer and were calculated from formula (5).

(ii.4) It was believed that the decision under appeal intended to refer to the document "Lecture Bull. Inst. Chem. Res. Kyoto Univ. Vol.69. No.2. 1991, pages 177 to 183" (referred below as D1) and not to document (iii) as mentioned in paragraph III of this decision.

(ii.5) The equation between the SCB and the elution temperature disclosed in D1 was substantially identical to the equation (5) of the application in suit. The SCB was plotted against cumulative weight values in D1 and against relative weight values in the application in suit to obtain a chemical distribution curve. The $SC_{ave}$ and the standard deviation $\sigma$ were determined from this curve.

(ii.6) Thus, the method used in the application was self-consistent and reasonably based on the published literature.

(iii) It was thus concluded that the application in suit met the requirements of Article 83 EPC.

V. Following a communication by the Board dated 3 September 2001 and a letter of the Appellant dated 1 March 2002, the Appellant was informed in an annex to oral proceedings issued on 23 April 2002 about a number of essential questions to be discussed at the oral
proceedings scheduled for 31 July 2002 and concerning the parameters (B) and (C). Concerning the parameter (B), it was stressed that, although, the application in suit referred to the Japanese standard JIS K 6760, the former, however, related to ethylene copolymers which were outside the scope of the JIS K 6760. Concerning the parameter (C), it was, in particular, pointed out, that the calculations of the first $H_i$ with formula (3) and of the standard deviation $\sigma$ needed to be clarified, that the formula (5) would lead to negative values of the short chain branching for the last two elution temperatures, and that the correction proposed by the Appellant in formula (6) would be checked in the light of the principles set out in decision G 03/89 (OJ EPO 1993, 117).

VI. With its letter dated 1 July 2002, the Appellant submitted an amended version of Claims 1 to 6 (part) of its main request and amended description pages 3, 6, 7, 8, 26, 29 and 30, as well as eight new documents. Its arguments may be summarized as follows:

(i) Concerning the parameter B:

(i.1) Whether or not the polymers according to the application in suit fell within the ambit of the JIS K 6760, the specification taught to use the conditions prescribed in that document.

(i.2) Thus, it was clear which conditions should be used for determining the MFR.

(ii) Concerning the parameter (C):

(ii.1) $C(x)$ was simply a measure of both the degree and the breadth of distribution of the short chain branching of the polymer. Various techniques were known to determine the SCB as shown by
document D1. It was also evident that the wordings "chemical composition" and "short chain branching" used in the application referred to the SCB. Thus, the replacement of "chemical composition" by "short chain branching" did not offend Article 123(2) EPC.

(ii.2) The first temperature corresponded to $i = 0$ and was selected such that essentially no polymer was eluted. Thus, the first reading could be used uncorrected (i.e. $H_0 = S_0$) and was zero anyway. Thus, whether the reading at $T_0$ was used in the determination of the $SCB_{ave}$ or the standard deviation was not an issue.

(ii.3) The formula given for calculating the SCB did not apply to all temperatures but only at the temperatures where polymer would in fact be eluted from the column. The two last readings would not in any case contribute to the values of $SCB_{ave}$ and the standard deviation.

(ii.4) The correction proposed in the formula for calculating the $SCB_{ave}$ corresponded to the normal meaning of the term "average". The skilled person would arrive at this formula from common general knowledge. In that respect, the Appellant referred to the document "N. A. Weiss et al. 'Introductory Statistics', Third Edition, Addison-Wesley Publishing Company (1991), pages 107, 128 to 130, 233 to 245" (referred to below as D2), submitted with its letter of 1 July 2002.
(ii.5) In the present case, the SCBi values did not have equal weight. Thus, the standard deviation, which applied, was the one for such variables, i.e., which took into consideration the specific probability of each value. Reference was also made to D2.

VII. On 30 July 2002, an interview took place with the Appellant. At the interview, the Appellant submitted a new set of Claims 1 to 8 as sole request.

Claim 1 of this request reads as follows:

"An ethylene-α-olefin copolymer in which
(A) a density is 0.870 to 0.945 g/cm³,
(B) a relation between an activation energy of flow Ea (J/mole K) obtained by measurement of viscoelasticity at at least three temperatures in the molten state and a melt flow rate MFR (g/10 min) measured according to the method prescribed in JIS K 6760 satisfies the following equation (1):

$$\log E_a \geq 4.6 - 0.04 \times \log MFR$$ (1)

(C) a coefficient Cx of variation of short chain branching distribution represented by the following equation (2) is 0.40 to 0.80:

$$Cx = \frac{\sigma}{SCB_{ave}}$$ (2)

wherein $\sigma$ is a standard deviation of short chain branching distribution (1/1,000 C) and SCB_{ave} is the average of short chain branchings per 1,000 C (1/1,000 C), and

(D) a ratio of a weight average molecular weight Mw to a number average molecular weight Mn (Mw/Mn) is 3 to 20."
Claims 2 to 7 were dependent on Claim 1. Independent Claim 8 relates to a molded article formed from the ethylene-alpha-olefin copolymer according to Claim 1 or 2.

Following the interview, the Board decided to cancel the oral proceedings scheduled for 31 July 2002.

VIII. The Appellant requested that the decision of the Examining Division be set aside, and the case be remitted to the Examining Division for further prosecution on the basis of the sole request filed during the interview of 30 July 2002.

Reasons for the Decision

1. The appeal is admissible.

2. Amendments

2.1 Claim 1 differs from Claim 1 as originally filed by (i) the indication that the MFR is "measured according to the method prescribed in JIS K 6760" and the replacement of the expression "chemical composition" by the expression "short chain branching".

2.2 Amendment (i) is supported by lines 25 to 26 on page 5 of the application as originally filed.

2.3 It is also evident from the quotations between parentheses of the term SCB on line 22 on page 29 and on line 10 on page 30 of the application as originally filed, and from the expression "SCB1 was plotted against RH1 to obtain a chemical composition distribution curve" (lines 27 and 28 on page 29) that the expression "chemical composition" was indeed intended to refer to
the short chain branching. Thus, the amendment (ii) is also supported by the application documents as originally filed.

2.4 Claims 2 to 8 find their support in original Claims 2 to 8, respectively.

2.5 It thus follows that the requirements of Article 123(2) EPC are complied with by all the claims.

2.6 The amendment carried out by the Appellant in the formula (4) on page 29 clearly represents an obvious correction of an error resulting from an obviously erroneous calculation. It is therefore allowable under Rule 88 EPC.

2.7 The correction made by the Appellant in formula (6) on page 30 establishes, in the Board view, what the skilled person, using general knowledge (cf. D2, page 129, formula 3.4), would already have derived from the description of the application in suit on the date of filing, in order to calculate the mean value (i.e. average) of short chain branching of the whole copolymer from the short chain branching of copolymer fractions (i.e. SCBi) each having a specific relative concentration (i.e. RHj). This correction is therefore allowable under Rule 88 EPC.

3. Sufficiency

3.1 According to the decision under appeal, the parameter (B) and the parameter (C) in Claim 1 then on file cannot be determined by a person skilled in the art using only the disclosure of the application and his general knowledge. Thus, it concludes that a person skilled in the art cannot carry out the invention
without knowing how to determine all the parameters of Claim 1 as he does not know which copolymers are inside the scope of this claim and which are outside the scope of this claim.

3.2 Concerning the parameter (B) it was held in the decision under appeal that the MFR needed an indication of the measuring temperature and load.

3.3 As indicated in paragraph 2.1 above Claim 1 now refers to the method prescribed in JIS K 6760 for the determination of the MFR of the claimed copolymers.

3.4 The JIS K 6760 is a Japanese standard which defines the testing methods for polyethylenes. In that respect, it states that the measurement of melt flow rate shall be carried out in accordance with JIS K 7210, and that the test conditions shall be, as a rule, Condition 4 specified in table I of JIS 7210, i.e. testing temperature 190°C and testing load 2.16 kg.

3.5 While it is true that Claim 1 relates to ethylene copolymers which might be outside the scope of the JIS K 6760, in terms of amount of comonomers and density, the Board accepts the submissions of the Appellant that the application in suit (cf. page 5, lines 25 to 26) teaches the skilled artisan to use the prescribed conditions thereof whether or not the copolymer falls within the scope of JIS K 6760.

3.6 It thus follows that the skilled would know how to determine the MFR and by consequence the parameter (B) since the application in suit further teaches how to measure the activation energy Ea (cf. page 25, line 20 to page 26, line 24 of the application as originally filed) which is also part of the calculation of parameter (B).
The parameter (C) is defined by the equation (2), as the ratio (Cx) of the standard deviation of short chain branching σ to the average of short chain branching \( SCB_{ave} \). Thus, the determination of the parameter (C) presupposes that the skilled person would know how to determine the values of \( SCB_{ave} \) and σ.

As indicated above, the mathematical formula for calculating the \( SCB_{ave} \) is known to the skilled person. The same is true for the mathematical formula for calculating the standard deviation σ of short chain branching from the short chain branching of the copolymer fragments having each a specific relative concentration (cf. D2, page 129, formula 3.5). These formulae read as follows:

\[
SCB_{ave} = \frac{\Sigma (SCB_i \times RH_i)}{\Sigma RH_i}, \quad \text{and} \\
\sigma = \left[ \frac{\Sigma ((SCB_i - SCB_{ave})^2 \times RH_i)}{100} \right]^{1/2}
\]

Consequently, the question as to whether the skilled person would know how to determine the parameter (C) boils down as to whether he would know how to determine both of the terms \( SCB_i \) and \( RH_i \) used in the above formulae.

According to the description of the application in suit, the \( SCB_i \) is calculated by the equation (5) and is only dependent on the elution temperature \( T_i \) at which the "i" copolymer fraction elutes from the column. It is also evident that the eluting temperatures are known (cf. page 27, lines 15 to 27).

While it is true, as mentioned by the Examining Division in its decision, that the short chain branching of the claimed copolymer would appear to be independent of the copolymer in question, the Board is not aware of a document showing that this formula (5)
does not work for ethylene-\(\alpha\)-olefin copolymers such as those according to the application in suit. On the contrary, the document D1 shows that there is indeed the same linear relation between short chain branching of ethylene-\(\alpha\)-olefin copolymers and elution temperature (cf. D1, page 182, formula (1)).

3.12 It is also true that the formula (5) would theoretically lead to "negative" SCB values when the elution temperature is as high as 101°C and 105°C. In view of D1 (cf. in particular Figure 7) which shows that the value of SCB is regarded as being zero when the elution temperature is greater than 100°C, the Board accepts the submissions of the Appellant that, as in D1, the two last SCB_i values corresponding to the temperatures 101 and 105°C would be considered by the skilled person as being equal to zero.

3.13 It thus follows that the skilled person, using the formula (5) would know how to obtain all the SCB_i values corresponding to the different elution temperatures mentioned on page 27 of the description of the application in suit.

3.14 Concerning the term RH_i, while the application in suit gives a detailed description of the method of how to determine this term (cf. passage from line 25 on page 27 to line 28 on page 29, some unclarities might have, \textit{prima facie}, arisen from the equation (3) and, in particular the way of determining the first term H_i corresponding to the first elution temperature i.e. -10°C.

3.15 It should, however, be taken into account that the method described in the application in suit, is based on the temperature rising elution fractionation technique which is well known to the skilled person in the polymer field (cf. D1).
3.16 Thus, the skilled person, reading the description of the method disclosed in the application in suit would try, with synthetical propensity, i.e. building up rather than tearing down, to arrive at an interpretation of the formula (3) which is technically sensible in respect to the temperature rising elution fractionation technique.

3.17 In that respect, formula (3) can only have a practical technical meaning, provided it relies on well defined temperature intervals. This requires that the term \( T_i - T_{i-1} \) must be known. In the practice of the temperature rising elution technique, there will be inevitably no \( "T_{i-1}" \) corresponding to the first elution temperature (i.e. in the present case, \(-10^\circ C\)) for defining the temperature interval necessary for the calculation of the \( H_i \) corresponding to the first elution temperature (i.e. \( H_0 \)). This will de facto imply that the formula (3) can only be applied starting from the first temperature interval known i.e. between \(-10^\circ C\) and \(0^\circ C\). By way of consequence, this would also imply that, in order to get a technically conclusive analysis on the whole copolymer from the fragments thereof eluted from the column, practically, all the copolymer should elute in the temperature range selected, i.e. the cumulative weight of the copolymer fragments eluted in the range from \(-10^\circ C\) to \(105^\circ C\) should, in practice correspond to total weight of copolymer put into the column.

3.18 In that context, it is credible to the Board, as submitted by the Appellant, that the skilled person would understand that the first elution temperature would be selected in such a way that practically no copolymer would elute below it from the column and that, consequently, \( S_0 \) and \( H_0 \) would be equal to zero.
Thus, it follows from the above that the skilled person would know how to determine the terms $H_i$ and $R_{H_i}$ and therefore the terms $\sigma$ and SCB used in the calculation of the parameter (C).

It is also evident that the skilled person would know how to measure the further parameters (A) (i.e. density) and (D) (i.e. polydispersity) mentioned in Claim 1, since the application in suit explicitly discloses the methods for their determination (cf. page 25, lines 1 to 3; page 24, lines 2 to 20).

In addition to this, the application in suit provides sufficient information regarding the processing conditions concerning the copolymerisation process (cf. page 9, lines 8 to 14; page 19, line 21 to page 21, line 2; Examples 1 and 2), so that there can be no doubt that the skilled person would know how to obtain a copolymer within the terms of Claim 1.

Thus, it follows from the above that the requirements of Article 83 EPC must be regarded as met.

As indicated above, the application in suit discloses the methods as how to determine the parameters (A), (B), (C) and (D) used for defining the claimed copolymers. These parameters are therefore appropriately defined. Thus, the Board takes the view that the requirements of Article 84 EPC are met.

In view of the above findings, all the objections specifically mentioned in the decision under appeal have been met. It is therefore necessary for the decision under appeal to be set aside.
6. Furthermore, it is clear from the wording of the decision under appeal itself (Reasons for the decision, point 5) that the examination of the application with respect to the requirements of Article 54 and 56 EPC has not yet taken place. Thus, in order to avoid the loss of instance, the Board makes use of its powers under Article 111(1) EPC to refer the case back to the first instance.

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 for further prosecution on the basis of Claims 1 to 8 submitted at the interview of 30 July 2002.

The Registrar: The Chairman:

E. Görgmaier R. Young