

**Internal distribution code:**

- (A) [ ] Publication in OJ  
(B) [ ] To Chairmen and Members  
(C) [X] To Chairmen

**D E C I S I O N**  
**of 27 April 2000**

**Case Number:** T 0855/97 - 3.3.3

**Application Number:** 93912094.5

**Publication Number:** 0642544

**IPC:** C08G 65/40

**Language of the proceedings:** EN

**Title of invention:**

Phase transfer catalyzed preparation of aromatic polyether  
polymers

**Applicant:**

GENERAL ELECTRIC COMPANY

**Opponent:**

-

**Headword:**

-

**Relevant legal provisions:**

EPC Art. 56

**Keyword:**

"Inventive step - obvious combination of known features"

**Decisions cited:**

-

**Catchword:**

-



Case Number: T 0855/97 - 3.3.3

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.3  
of 27 April 2000

**Appellant:** GENERAL ELECTRIC COMPANY  
1 River Road  
Schenectady, NY 12345 (US)

**Representative:** Szary, Anne Catherine, Dr.  
London Patent Operation  
Essex House  
12-13 Essex Street  
London WC2R 3AA (GB)

**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 10 March 1997  
refusing European patent application  
No. 93 912 094.5 pursuant to Article 97(1) EPC.

**Composition of the Board:**

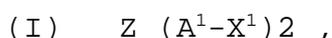
**Chairman:** C. Gérardin  
**Members:** P. Kitzmantel  
W. Moser

## Summary of Facts and Submissions

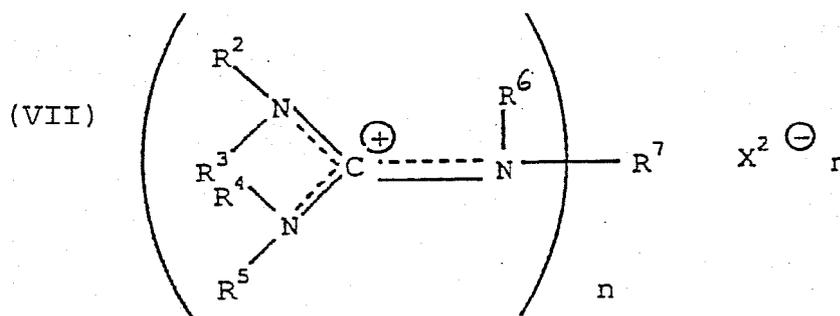
I. This appeal, which was filed on 5 May 1997, lies against the decision of the Examining Division dated 10 March 1997, refusing European patent application No. 93 912 094.5 filed as PCT/US93/02925 on 30 March 1993 in the name of General Electric Company, and published under No. WO 94/22939 (EP publication No. 0 642 544). The appeal fee was paid together with the Notice of Appeal and the Statement of Grounds of Appeal was filed on 11 July 1997.

II. The decision under appeal was based on a set of 13 claims filed with the submission of 11 September 1996, Claim 1 reading as follows:

"1. A method for preparing an aromatic polyether polymer which comprises contacting, in a solvent having a polarity no higher than that of o-dichlorobenzene, 1,2,4-trichlorobenzene and diphenyl sulfone and at a temperature in the range of about 125-250°C, substantially equimolar amounts of at least one alkali metal salt of a dihydroxy-substituted aromatic hydrocarbon and at least one substituted aromatic compound of the formula



wherein Z is an activating radical, A<sup>1</sup> is an aromatic radical and X<sup>1</sup> is fluoro, chloro, bromo or nitro, in the presence of from 0.5 to 10 mole percent based on the bisphenol salt of a phase transfer catalyst selected from an alkylaminopyridinium salt or a guanidinium salt of the formula



wherein:

each of R<sup>2</sup>, R<sup>3</sup>, R<sup>4</sup>, R<sup>5</sup> and R<sup>6</sup> is a primary alkyl radical and R<sup>7</sup> is a primary alkyl or bis(primary alkylene) radical, or at least one of the R<sup>2</sup>-R<sup>3</sup>, R<sup>4</sup>-R<sup>5</sup> and R<sup>6</sup>-R<sup>7</sup> combinations with the connecting nitrogen atoms forms a heterocyclic radical;

R<sup>2</sup> [this is an obvious error and should read X<sup>2</sup>] is an anion; and

n is 1 or 2."

The further claims 2 to 13 were dependent on Claim 1.

III. The decision under appeal held that it was obvious to replace the ammonium and phosphonium phase transfer catalysts used according to document

D1: US-A-4 310 654

by the phase transfer catalysts employed according to documents

D2: US-A-5 081 298 or

D3: EP-A-0 126 950,

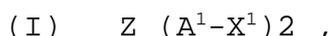
because the person skilled in the art was aware from

these documents that the latter catalysts would be able to enhance the molecular weight of the polymers. Claim 1 did not, therefore, relate to inventive subject-matter, nor did it meet the requirement of unity of invention.

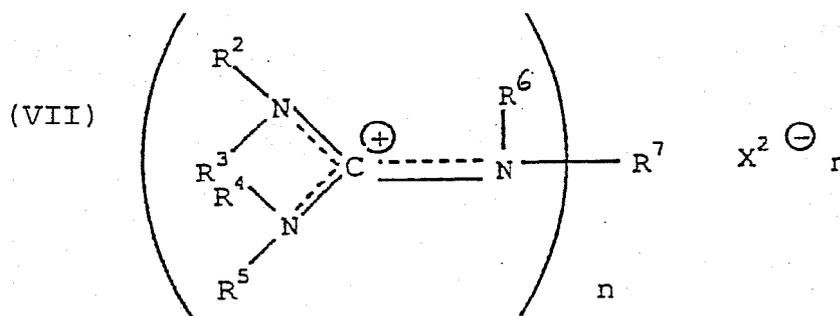
Claim 1, furthermore, contravened Article 123(2) EPC, because the definition of the solvent was not originally disclosed; the latter objection, however, was not a ground for the refusal.

IV. During the oral proceedings held on 27 April 2000, in which the Board emphasized the preliminary objections under Article 123(2) and 56 EPC expressed in the Rapporteur's communication of 16 November 1999, the Appellant abandoned its previous request and submitted an amended set of claims, Claim 1 reading as follows:

"1. A method for preparing an aromatic polyether polymer which comprises contacting, in a solvent selected from o-dichlorobenzene, 1,2,4-trichlorobenzene, dichlorotoluene and diphenyl sulfone and at a temperature in the range of about 125-250°C, substantially equimolar amounts of at least one alkali metal salt of a dihydroxy-substituted aromatic hydrocarbon and at least one substituted aromatic compound of the formula



wherein Z is an activating radical, A<sup>1</sup> is an aromatic radical and X<sup>1</sup> is fluoro, chloro, bromo or nitro, in the presence of from 0.5 to 10 mole percent based on the bisphenol salt of a phase transfer catalyst guanidinium salt of the formula



wherein:

each of  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$  and  $R^6$  is a primary alkyl radical and  $R^7$  is a primary alkyl or bis(primary alkylene) radical, or at least one of the  $R^2$ - $R^3$ ,  $R^4$ - $R^5$  and  $R^6$ - $R^7$  combinations with the connecting nitrogen atoms forms a heterocyclic radical;

$X^2$  is an anion; and

$n$  is 1 or 2."

The further claims 2 to 12 are identical to claims 2 to 10, 12 and 13 of the version according to the decision under appeal.

V. In the Statement of Grounds of Appeal, the written submission dated 27 March 2000 and during oral proceedings the Appellant essentially argued that the inventivity of the subject-matter of Claim 1 could not be denied on the basis of a combination of documents D1, D2 and/or D3, because the conditions of preparation of the **non-polymeric** products according to these documents could not be transferred to the different conditions of the preparation of **polymeric** products as according to the present invention.

D1 merely disclosed the preparation - in the presence of different phase transfer catalysts, namely

quaternary ammonium or phosphonium salts - of bisphenoxide terminated diarylsulfone **oligomers** as starting material for subsequent polymer formation by chain extension reaction with a methylene halide; D2 and D3, although using phase transfer catalysts according to present Claim 1, only disclosed the preparation of **monomeric** reaction products of e.g. alkali metal bisphenoxides and nitro- or halo-substituted activated aromatic compounds.

It was, thus, not possible, in the Appellant's opinion, to predict the molecular weight enhancing ability of the "inventive" phase transfer catalysts.

This conclusion was all the more true as the expert was aware from document

D4: Journal of Polymer Science, Polymer Chemistry Edition, volume 19, 1635 to 1658 (1981),

that the reaction of bis-nitrophthalimides with bisphenols would lead but to low molecular weight products.

- VI. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of the claims submitted during oral proceedings.

### **Reasons for the Decision**

1. The appeal is admissible.
2. *Amendments*

Claim 1 is based on its original version, on page 7, lines 14 to 16 (with respect to the solvent definition), on page 13, lines 7 to 10 (with respect to the amounts of phase transfer catalyst) and on original Claim 3 (with respect to the characterisation of the phase transfer catalyst).

Claims 2 to 5 are based on original Claims 2 and 4 to 6; Claim 6 is based on original Claims 7 and 8; Claims 7 to 10 are based on original Claims 9 to 11 and 13; Claim 11 is based on original Claim 17 and Claim 12 is based on original Claims 18 to 20.

The requirement of Article 123(2) EPC is therefore complied with by all claims.

The present wording of the claims also overcomes the objection of lack of clarity raised initially by the Examining Division (cf. communication of 15 April 1996, point 1) and emphasized by the Board during the oral proceedings arising from the expression "solvent of low polarity" (Article 54 EPC).

3. *Prior art*

3.1 Document D1

This document relates to a method for making polyethersulfoneformals by (A) preparing a bisphenoxide terminated diarylsulfone bisphenol oligomer by reacting a dihalodiarylsulfone  $XRSO_2RX$  ( $X$  = halogen,  $R$  = divalent aromatic radical, e.g. phenylene) and a bisphenoxide salt  $MOZOM$  ( $M$  = alkali metal,  $Z$  = aromatic radical, e.g. bisphenol A), in the presence of a dipolar aprotic solvent or a nonpolar solvent and an effective amount

of a phase transfer catalyst and (B) reacting said oligomer with a methylene halide (Claims 1 and 8; column 2, lines 42 to 43; column 3, lines 8 to 10).

According to formula (4) in column 2, lines 29 to 41 the resulting polyethersulfoneformals may comprise  $n = 1$  to 400 units of the diarylsulfone bisphenol oligomer structure (-ORSO<sub>2</sub>RO-Z-).

Dichlorobenzene is mentioned as nonpolar organic solvent and quaternary ammonium and phosphonium salts as phase transfer catalysts (column 4, lines 46 to 61).

### 3.2 Document D2

This document relates to the preparation of  $\alpha,\omega$ -bis(pentaalkylguanidinium) alkane salts, such as 1,6-bis (penta-n-butylguanidinium) hexane dibromide, and to their use as phase transfer catalysts, particularly in reactions between an alkali metal salt of a hydroxyaromatic compound, e.g. bisphenol A disodium salt, and an activated halo- or nitro-substituted aromatic compound, e.g. a halo-N-alkylphthalimide (Abstract; column 4, lines 1 to 14; column 6, Example 4).

### 3.3 Document D4

This document relates to the preparation of polyetherimides by displacement of nitro groups from disubstituted bis-imides by the dianion of bisphenols (page 1635, Synopsis).

The polymerisation is reported to proceed rapidly under relatively mild conditions in dipolar aprotic solvents

(e.g. DMF) or mixtures thereof with toluene (e.g. DMSO/toluene) to provide high molecular weight products (page 1639, last paragraph).

Depending on the starting materials and on the reaction conditions a wide range of weight average molecular weights  $M_w$  is achieved, the highest values being 123000 (Table III, polymer (18jp)), 99000 (Table II, polymer (18fp)) and 83900 (Table I, polymer (18ap)).

4. *Novelty*

4.1 None of the documents on file discloses the preparation of aromatic polyether polymers from alkali metal salts of dihydroxy-substituted aromatic hydrocarbons and halo- or nitro- substituted activated aromatic compounds in the presence of phase transfer catalysts as specified in present Claim 1.

4.2 Concerning the disclosure of document D1, it is noted that neither the fact that step (A) of Claim 1 only yields an intermediary product, nor the further fact that this intermediary product is designated as "oligomer" qualify as distinguishing features. On the one hand the preparation of these "oligomers" represents state of the art that is clearly made available to the public as required by Article 54(2) EPC, and on the other hand the number of repeating units "n" comprised by the "oligomers" may go up to 400 (column 2, lines 29 to 41, formula (4)), making the "oligomers" undistinguishable from "polymers" as prepared according to present Claim 1. This is also conspicuous from a comparison of the molecular weights exemplified in the Table on page 15 of the application in suit for the polymers prepared according to present Claim 1, which range between 16,200 (Example 15, 2 hrs.) and 226,200 (Example 21, Time 1 hr.) and the maximum molecular weight that can be calculated for an "oligomer" comprising 400 repeating units comprised by subunits from bisphenol A (subunit molecular weight 226) and diphenylsulfone (subunit molecular weight 216), which is 176800.

Moreover, one skilled in the art is aware from common general knowledge that for the achievement of higher

molecular weights of the "oligomer" it is necessary to operate at molar ratios of dihaloarylsulfone and bisphenoxide salt close to 1:1; the fact that according to Claim 1, step (A) of D1 this molar ratio may range from 1.0001 to 2 does not, therefore, detract from the feasibility of the disclosure of D1 for the preparation of high molecular weight "oligomers".

4.3 The subject-matter of Claim 1 is, thus, novel over the citations.

4.4 The same conclusion applies *a fortiori* to the subject-matter of the dependent claims 2 to 12.

5. *Problem and solution*

5.1 The closest prior art is represented by document D1. With regard thereto the problem underlying the subject-matter of present Claim 1 is the provision of an improved method for the preparation of aromatic polyether polymers in nonpolar solvents and in the presence of phase transfer catalysts, which method is able to yield polymers having higher molecular weights.

5.2 According to Claim 1 this problem is to be solved by the use of certain guanidinium salts as phase transfer catalysts in combination with specific solvents.

5.3 From the results comprised by the Table on page 15 (in conjunction with page 13, lines 20 to 30) of the application it can be concluded that this object has been achieved. This is particularly conspicuous from a comparison of the weight average molecular weights  $M_w$  in the following Examples:

Example No.	Subst. aromatic compound	phase transfer catalyst	Time (hrs)	M <sub>w</sub>
18*	1,3-Bis[N-(4-chloro-phthalimido)] toluene	tetra-n-butyl-phosphonium bromide*	1	6,700
20	1,3-Bis[N-(4-chloro-phthalimido)] toluene	hexaethyl-guanidinium bromide	1	153,600
21	1,3-Bis[N-(4-chloro-phthalimido)] toluene	1,6-bis(N,N',N', N'',N''-penta-n-butylguanidinium) hexane dibromide	1	226,200
22	1,3-Bis[N-(4-chloro-phthalimido)] toluene	tris(piperidino) guanidinium bromide	1	136,500

\* comparative

## 6. Obviousness

6.1 The only differences between the subject-matter of present Claim 1 and the disclosure of D1 reside (i) in the use of different phase transfer catalysts (cf. point 4.2 supra) and (ii) in a broader list of suitable solvents.

6.2 Document D2 (cf. point 3.2 supra) relates to the preparation of certain guanidinium salts, which are within the definition of present Claim 1, and to their use as phase transfer catalysts *inter alia*. "in reactions between at least one alkali metal salt of a

hydroxyaromatic compound ... and at least one activated halo- or nitro-substituted aromatic compound" (cf. column 4, lines 8 to 14). Most often the hydroxyaromatic compound is a dihydroxyfunctional compound, e.g. bisphenol A (column 4, lines 31 to 44, especially lines 36 to 38). Because the "activated halo- or nitro-substituted aromatic compounds" are monofunctional (cf. column 5, lines 14 to 43), this process cannot lead to polymeric structures. However, the chemical reactions which occur in the formation of monomeric compounds of the structure A-B-A (A = residue of **monofunctional** activated aromatic compound; B = residue of e.g. bisphenol) and in the formation of polymeric structures (A-B)<sub>n</sub> (A = residue of **difunctional** activated aromatic compound, otherwise corresponding to A) are the same, if the same functional groups are involved.

The skilled person will, therefore, undoubtedly consider the guanidinium based phase transfer catalysts used according to D2 for the preparation of compounds of the structure A-B-A as possible substitutes for the quaternary ammonium and phosphonium phase transfer catalysts used according to D1 for the preparation of polymers of the structure (A-B)<sub>n</sub>.

6.3 Although the present wording of Claim 1 requires the polycondensation reaction to be carried out in a solvent selected from four specific aromatic solvents, this feature cannot contribute to the inventivity of the process, since dichlorobenzene is mentioned in D1 as one of the suitable nonpolar solvents (column 4, lines 58 to 60).

6.4 The issue of inventive step turns on the question

whether, in view of the problem to be solved as set out in point 5.1 supra, it was obvious to replace the quaternary ammonium or phosphonium phase transfer catalysts used according to D1 by the phase transfer catalysts prepared and employed according to D2; or in other terms, whether the person skilled in the art, seeking to enhance the molecular weight of the "oligomers" disclosed in D1, would have expected that this object can be attained by using the guanidinium salt phase transfer catalysts according to D2.

6.5 The reply to the above question must be in the affirmative, because document D2 teaches that the guanidinium phase transfer catalysts provide reaction rates and yields which are superior to those of previously known phase transfer catalysts, including the quaternary ammonium and phosphonium compounds used according to D1. Therefore, the skilled person could legitimately expect that their use would solve the existing technical problem, i.e. the provision of polymers of higher molecular weight.

6.6 This conclusion is based

(i) on the statement on column 2, lines 16 to 30 of D2 setting out with regard to the copending application Ser. No. 07/626,150 that various guanidinium salts, when used as phase transfer catalysts for reactions between polar and non-polar compounds, have been known to increase reaction rate and yield as compared to previously used phase transfer catalysts, among which quaternary ammonium and phosphonium salts (cf. column 1, lines 40 to 51), and

- (ii) on the information contained in Example 4 of D2 (column 6, lines 30 to 45), according to which the use of the "inventive" phase transfer catalyst 1,6-bis(penta-n-butylguanidinium)hexane dibromide led to a yield of 93% after 30 minutes, whereas the use of the prior art catalyst 1,6-bis(tri-n-butylammonium)hexane dibromide in twice the molar amount of the guanidinium catalyst gave a yield of only 20% after 3 hours.
  
- (iii) In the case of difunctional reaction partners necessary for the preparation of polymers, the higher reaction rates and the higher yields evidenced in D2 must both enhance the chain propagation and cause a more efficient formation of bonds -A-B-. In other words, the superiority of the guanidinium phase transfer catalysts with respect to reaction rate and yield over the known ammonium and phosphonium catalysts must, in the Board's judgment, inevitably lead to an increase of the molecular weight, if the same guanidinium catalysts are used under the conditions of polymer formation according to D1.

6.7 The above conclusion is not invalidated by the Appellant's reference to document D4, which discusses the preparation of polymers (18) by reaction of bis-imides (16) with bisphenols (17) in dipolar aprotic solvents **in the absence** of phase transfer catalysts (cf. page 1639, last two paragraphs). Neither is this document relevant to the performance of different phase transfer catalysts, nor is it able to prove, as alleged in the Appellant's submission dated 27 March 2000, that the nitro-group displacement polymerisation discussed

in D4 (cf page 1635, first paragraph of "Introduction"), a reaction-scheme within the scope of present Claim 1, is *per se* unable to give rise to high molecular weight polymers. This can *inter alia* be inferred from the high molecular weight  $M_w$  of 123000 of the polymer (18jp) disclosed in Table III (page 1646). It is without consequence for the obviousness issue under discussion that this polymer (18jp) exhibits a rather high  $M_w/M_n$  ratio, because, being mentioned nowhere in the application in suit, this ratio has no relevance for the issue under discussion.

- 6.8 The subject-matter of Claim 1, thus, does not comply with the requirements of Article 56 EPC.
- 6.9 By virtue of their appendance to Claim 1 the same conclusion applies to Claims 2 to 12.

## **Order**

### **For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:

E. Görgmaier

C. Gérardin