## Appeal decision

Appeal No. 2018-2480

# Tokyo, Japan Appellant

### MITSUBISHI GAS CHEMICAL COMPANY, INC.

Patent Attorney TSUKUNI & ASSOCIATES

The case of appeal against the examiner's decision of refusal for Japanese Patent application No. 2014-69513, titled "AROMATIC POLYCARBONATE RESIN COMPOSITION" [published on November 2, 2015, Japanese Unexamined Patent Application Publication No. 2015-189906] has resulted in the following appeal decision.

Conclusion

The appeal of the case was groundless.

#### Reason

No. 1 History of the procedures

The present application is an application filed on March 28, 2014. The history of the procedures is set forth as below:

October 11, 2017:	Notice of reason for refusal
December 14 of the same year:	Submission of written opinion and written
	amendment
December 20 of the same year:	Decision of Rejection
February 21, 2018:	Submission of notice of appeal and written
	amendment
April 9 of the same year:	Notice of reason for refusal (by the examiner
	prior to appeal proceedings)
June 13 of the same year:	Submission of written opinion and written
	amendment
January 22, 2019:	Notice of reason for refusal (by the body in the
	appeal proceeding)
March 27 of the same year:	Submission of written opinion and written

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#### amendment

# No. 2 The Invention

The inventions according to Claims 1 to 6 of the present application should be specified in the matters recited in Claims 1 to 6 of the Claims amended according to the written amendment submitted on March 27, 2019. The invention according to Claim 1 (hereinafter referred to as "Invention 1") is recited as below:

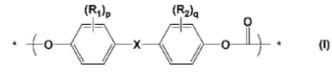
## "[Claim 1]

An aromatic polycarbonate resin composition comprising an aromatic polycarbonate resin having a structural unit represented by the following general formula (I), and a weight average molecular weight of 40000 to 100000, an inorganic filler, and a cyclic carbonate represented by the following general formula (IIa),

wherein said aromatic polycarbonate resin has a terminal hydroxyl group content of 1000 ppm or less, and said aromatic polycarbonate resin comprises a structural unit represented by the following general formula (III), and the content is less than 2000 ppm in an aromatic polycarbonate resin having a structural unit represented by said general formula (I),

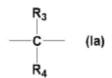
wherein said inorganic filler is at least one kind selected from the group consisting of glass fiber, glass beads, and glass flake, and the content of said inorganic filler is 3 to 200 mass parts with respect to 100 mass parts of aromatic polycarbonate resin having a structural unit represented by the following general formula (I), and the content of cyclic carbonate represented by the following general formula (IIa) is 0.1 ppm or more to 3000 ppm or less with respect to an aromatic polycarbonate resin having a structural unit represented by the following general formula (I).

[Chemical Formula 1]

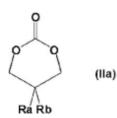


(where p and q represent 0, and X represents a group selected from the group of the following (Ia))

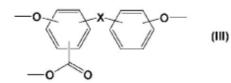
[Chemical Formula 2]



(where each of R3 and R4 represents a methyl group) [Chemical Formula 3]



(where Ra represents an ethyl group and Rb represents a butyl group) [Chemical Formula 5]



(where X has the same meaning as X in general formula (I))."

No. 3 Summary of reasons for refusal notified by the body

The reasons for refusal notified by the body on January 22, 2019 include the following:

The inventions according to Claims 1 to 10 of the present application were easily conceivable by a person skilled in the art who had ordinary knowledge in the technical field to which the invention pertained on the basis of the invention described in Cited Document 1 that had been distributed in Japan or foreign country before the filing and the matter described in Cited Document 2 and on the basis of the invention described in Cited Document 1 and the matters described in Cited Documents 2 to 4. Thus the inventions cannot be granted a patent under the provision of Article 29(2) of the Patent Act.

Cited Document 1: International Publication No. WO 2012/157766 Cited Document 2: Japanese Unexamined Patent Application Publication No. 2001-329161

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Cited Document 3: Japanese Unexamined Patent Application Publication No. 2013-1801

Cited Document 4: Japanese Unexamined Patent Application Publication No. 2014-55255

Note that Invention 1 is an invention that further limits "X" of general formula (I) of a part of the invention according to Claim 4 depending from Claim 2 of the scope of claims to which an amendment was made by the written amendment on June 13, 2018

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and limits the alternatives of R3 and R4 to a methyl group, and the alternative of Ra in the general formula (IIa) to ethyl and the alternative of Rb to a butyl group.

No. 4 Description in Cited Documents and Cited Inventions

1 Description of Cited Document 1

Cited Document 1 describes the following matters:

# "[Claim 16]

A process for producing a highly polymerized aromatic polycarbonate resin which comprises a highly-polymerizing process wherein an aromatic polycarbonate is reacted with an aliphatic diol compound represented by the following general formula (g1) in the presence of a transesterification catalyst to increase the molecular weight: [Chemical Formula 9]

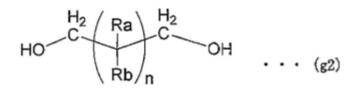
(In general formula (g1), each of Ra and Rb represents, independently, a hydrogen atom, or a linear or branched alkyl group or phenyl group with a carbon number of 1 to 12, and m is an integer of 1 to 30.)

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# [Claim 18]

The production method of Claim 16, wherein said aliphatic diol compound represented by said general formula (g1) is a compound represented by the following general formula (g2).

[Chemical Formula 10]



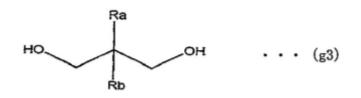
(In general formula (g2), each of Ra and Rb represents, independently, a hydrogen atom, or a linear or branched alkyl group or phenyl group with a carbon number of 1 to 12, and n is an integer of 1 to 28.)

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[Claim 20]

The production method of Claim 18, wherein said aliphatic diol compound represented by said general formula (g2) is a compound represented by the following general formula (g3).

[Chemical Formula 11]



(In general formula (g3), each of Ra and Rb represents, independently, a hydrogen atom, or a linear or branched alkyl group or phenyl group with a carbon number of 1 to 12.)

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[Claim 22]

The production method of Claim 20, wherein, in general formula (g3), each of Ra and Rb represents, independently, a linear or branched alkyl group with a carbon number of 1 to 4.

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[Claim 23]

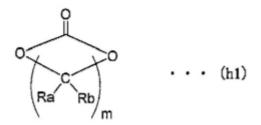
The production method of a highly polymerized aromatic polycarbonate resin of Claim 22, wherein said aliphatic diol compound is selected from the group consisting of 2-butyl-2-ethylpropane-1,3-diol, 2,2-diisobutylpropane-1,3-diol, 2-ethyl-2-methylpropane-1,3-diol, 2,2-diethylpropane-1,3-diol, and 2-methyl-2-propylpropane-1,3-diol.

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[Claim 26]

The process for producing a highly polymerized aromatic polycarbonate resin according to Claim 16 or 24, which comprises a highly-polymerizing process wherein an aromatic polycarbonate is reacted with said aliphatic diol compound, and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highly-polymerizing process is removed from the reaction system. [Claim 27]

The production method of Claim 26, wherein said cyclic carbonate is a compound represented by the following general formula (h1). [Chemical Formula 14]



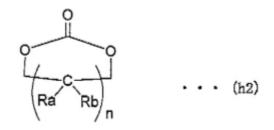
(In general formula (h1), each of Ra and Rb represents, independently, a hydrogen atom, or a linear or branched alkyl group or phenyl group with a carbon number of 1 to 12, and m is an integer of 1 to 30.)

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[Claim 29]

The production method of Claim 27, wherein said cyclic carbonate represented by said general formula (h1) is a compound represented by the following general formula (h2).

[Chemical Formula 15]



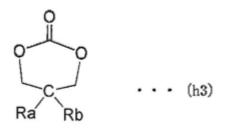
(In general formula (h2), each of Ra and Rb represents, independently, a hydrogen atom, or a linear or branched alkyl group or phenyl group with a carbon number of 1 to 12, and n is an integer of 1 to 28.)

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[Claim 31]

The production method of Claim 29, wherein said cyclic carbonate represented by said general formula (h2) is a compound represented by the following general formula (h3).

[Chemical Formula 16]

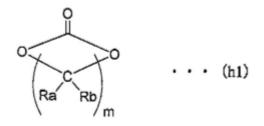


(In general formula (h3), each of Ra and Rb represents, independently, a hydrogen atom, or a linear or branched alkyl group or phenyl group with a carbon number of 1 to 12.)

... [Claim 39]

A polycarbonate resin composition comprising a highly polymerized aromatic polycarbonate resin obtained by the process according to Claims 16 or 24 as a main component and a cyclic polycarbonate represented by the following general formula (h1) in an amount of not more than 3000 ppm:

[Chemical Formula 17]



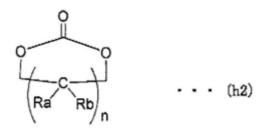
(In general formula (h1), each of Ra and Rb represents, independently, a hydrogen atom, or a linear or branched alkyl group or phenyl group with a carbon number of 1 to 12, and m is an integer of 1 to 30.)

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[Claim 41]

The polycarbonate resin composition of Claim 39, wherein said cyclic carbonate represented by said general formula (h1) is a compound represented by the following general formula (h2).

[Chemical Formula 18]



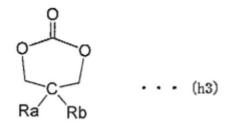
(In general formula (h2), each of Ra and Rb represents, independently, a hydrogen atom, or a linear or branched alkyl group or phenyl group with a carbon number of 1 to 12, and n is an integer of 1 to 28.)

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[Claim 43]

The polycarbonate resin composition of Claim 41, wherein said cyclic carbonate represented by said general formula (h2) is a compound represented by the following general formula (h3).

[Chemical Formula 19]



(In general formula (h3), each of Ra and Rb represents, independently, a hydrogen atom, or a linear or branched alkyl group or phenyl group with a carbon number of 1 to 12.) [Claim 44]

The polycarbonate resin composition of Claim 43, wherein, in said general formula (h3), each of Ra and Rb represents, independently, a hydrogen atom, or a linear alkyl group with a or carbon number of 1 to 5.

[Claim 45]

The polycarbonate resin composition according to Claim 39, wherein said highly polymerized aromatic polycarbonate resin has a structural viscosity index (N-value) represented by the following mathematical formula (1) of 1.25 or less: [Mathematical Formula 4]

N value =  $(\log(Q160 \text{ value}) - \log(Q10 \text{ value}))/(\log 160 - \log 10)...(1)"$ "[0004]

[0004]

Since polycarbonate is excellent in heat resistance, impact resistance, and transparency, it has been widely used in many fields in recent years. ..."

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"[0020]
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Examples of methods for improving fluidity include decrease in weight average molecular weight of a polycarbonate. However, the polycarbonate thus obtained has defects of a significant decrease in impact resistance and stress cracking-resistance, and also in solvent resistance. It is proposed to improve fluidity by widening a molecular weight distribution by blending polycarbonates differing in molecular weight (Patent Document 14; U.S. Patent No. 3,166,606, Patent Document 15; Japanese Unexamined Patent Application Publication No. S56-45945)."

While the above-mentioned methods may enable achievement of high fluidity, they have defects such as deterioration of properties that a polycarbonate originally has, complexity of the production process by adding a mixing and kneading operation or the like, deterioration of moldability such as releasability other than fluidity, restriction of intended use, and possibility of having high toxicity. Therefore, it had not been easy to obtain a high-fluidity polycarbonate resin while keeping good physical properties such as impact resistance or heat resistance which are useful properties of an aromatic polycarbonate resin."

# "[0033]

The problem to be solved by the present invention is to provide a novel polycarbonate copolymer having high fluidity in spite of having high molecular weight, while keeping good quality that a polycarbonate originally has without combining other resins or additives.

Another problem to be solved by the present invention is to provide an improved method for producing a highly polymerized aromatic polycarbonate resin which enables an increase in molecular weight of the aromatic polycarbonate resin satisfactorily while keeping good quality of the resin.

Another problem to be solved by the present invention is to provide an aromatic polycarbonate compound which is a prepolymer suitable for producing a highly polymerized polycarbonate using an aliphatic diol compound."

# "[0120]

The polycarbonate copolymer according to the present invention has achieved high molecular weight and high fluidity without using additives while keeping useful properties that a polycarbonate resin originally has such as physical strength such as impact resistance, abrasion resistance, and stress-cracking resistance, a satisfactory hue, optical properties, low equilibrium water absorption, heat resistance, dimensional stability, clarity, weather resistance, hydrolysis resistance, and flame retardance. In addition, the polycarbonate copolymer of the present invention has not only high molecular weight and high fluidity but also has a low N-value, which means that it scarcely has branch structures and/or different kinds of structures.

# [0121]

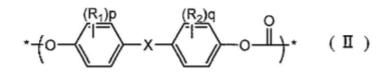
According to the novel process for producing a highly polymerized aromatic polycarbonate resin, an aromatic polycarbonate (= prepolymer) is reacted with an aliphatic diol compound having a specific structure to become highly polymerized, and at the same time, said aliphatic diol compound is removed away from the reaction system as the form of cyclic carbonate. As a result, the aliphatic diol compound is scarcely introduced into the main chain of the highly polymerized aromatic polycarbonate resin thus obtained. Therefore, since the highly polymerized aromatic polycarbonate resin thus obtained scarcely has a linked part remaining in the main chain, its structure is almost the same as that of a polycarbonate obtained by the conventional

interfacial polymerization or melt polymerization. For example, when an aromatic polycarbonate prepolymer obtained by using bisphenol A (BPA) as an aromatic dihydroxy compound is subjected to the production process according to the present invention, a polycarbonate having almost the same chemical structure as an ordinary polycarbonate resin derived from bisphenol A (BPA-PC) is obtained. The polycarbonate resin thus obtained has not only the same properties as polycarbonates obtained by conventional interfacial polymerization, but also has benefits in quality such as the low branching degree and a small amount of different kinds of structures, by virtue of conducting highly polymerizing rapidly using the aliphatic diol compound as a linking agent. In addition, it has significantly improved heat resistance or thermal stability under high temperature, which is caused by not containing a unit derived from the linking agent consisting of the aliphatic diol compound in the main chain."

#### (2) STRUCTURAL UNIT REPRESENTED BY GENERAL FORMULA (II)

The aromatic polycarbonate forming-unit of the polycarbonate copolymer of the present invention is a structural unit represented by general formula (II): [0201]

[Chemical Formula 57]



#### [0202]

In general formula (II), R1and R2 each independently represents a halogen atom, an alkyl group having 1-20 carbon atoms, an alkoxy group having 1-20 carbon atoms, a cycloalkyl group having 6-20 carbon atoms, an aryl group having 6-20 carbon atoms, a cycloalkoxyl group having 6-20 carbon atoms, or an aryloxy group having 6-20 carbon atoms. p and q each independently represents an integer of 0-4. X represents a single bond or an organic group selected from the group consisting of the divalent organic groups represented by the following general formula (II'):

### [0203]

[Chemical Formula 58]

$$R_3$$
  
-C-, -S-, -O-, -SO-, -CO-, -SO<sub>2</sub>- · · · (II')  
 $R_4$ 

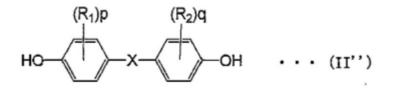
[0204]

In general formula (II'), R3 and R4 each independently represents a hydrogen atom, an alkyl group having 1-10 carbon atoms, or an aryl group having 6-10 carbon atoms. R3 and R4 can be linked with each other to form an aliphatic ring. [0205]

Examples of aromatic dihydroxy compounds which can derive the structural unit represented by the above-mentioned general formula (II) include a compound represented by the following general formula (II"):

[0206]

[Chemical Formula 59]



[0207]

In general formula (II'), R1-R2, p, q, and X are each the same as in the general formula (II).

[0208]

Examples	of	the	aromatic	dihydroxy	compounds	include	bis(4-
hydroxyphenyl)me	thane	,	1,1-bis	(4-hydroxyph	enyl)ethane,	2,	2-bis(4-
hydroxyphenyl)pro	opane	,	2,2-bis	(4-hydroxyph	enyl)butane,	2,	2-bis(4-
hydroxyphenyl)oct	ane,		bis(4-hydi	roxyphenyl)pł	nenylmethane,	1,	1-bis(4-
hydroxyphenyl)-1-	pheny	ylethar	ne, bis(4-)	hydroxypheny	l)diphenylmet	hane, 2,	2-bis(4-
hydroxy-3-methylp	oheny	l)prop	ane, 1,1-bi	s(4-hydroxy-3	3-tert-butylphe	nyl)propar	ne, 2,2-
bis(3,5-dimethyl-4	-hydr	oxyph	enyl)propan	ne, 2,2-bis(4-h	ydroxy-3-pher	ylphenyl)	propane,
2,2-bis(3-cyclohex	yl-4-l	nydrox	yphenyl)pro	opane,	2,2	-bis(4-hyd	lroxy-3-
bromophenyl)prop	ane,	2,2-	-bis(3,5-dib	romo-4-hydro	xyphenyl)prop	ane, 1,	1-bis(4-
hydroxyphenyl)cyd	cloper	ntane,	1,1-bis	(4-hydroxyph	enyl)cyclohexa	ine, 2,	2-bis(4-

hydroxy-3-methoxyphenyl)propane, 4,4'-dihidroxydiphenylether 4,4'-dihydroxy-3,3'dimethylphenylether, 4,4'-dihydroxyphenylsulfide, 4,4'-dihydroxy-3,3'dimethyldiphenylsulfide, 4,4'-dihydroxydiphenylsulfoxide, 4,4'-dihydroxy-3,3'dimethyldiphenylsulfoxide, 4,4'-dihydroxydiphenylsulfone, 4,4'dihydroxydiphenylsulfone, and 4,4'-dihydroxy-3,3'-dimethyldiphenylsulfone. [0209]

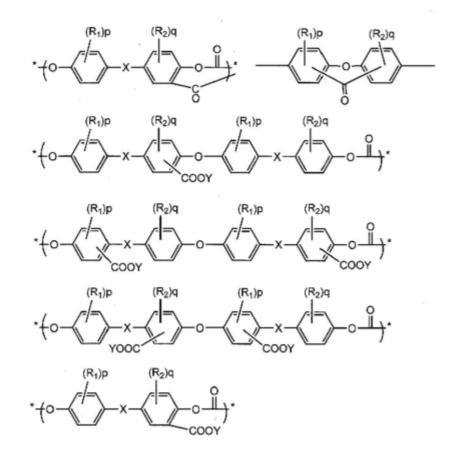
Among them, it is preferable to use 2,2-bis(4-hydroxyphenyl)propane, because of the stability as a monomer and availability of a commercial product having a low impurity content."

# "[0252]

The polycarbonate copolymer obtained by reacting the end-capped aromatic polycarbonate prepolymer with an aliphatic diol compound in the presence of a transesterification catalyst under reduced pressure according to the present invention has a high Q-value while having high molecular weight, and preferably, has a low N-value. In addition, the content of a unit having different kinds of structures which might cause negative effects on the present invention can be reduced to an extremely small amount. A unit having different kinds of structures here means a branching point unit or the like that a polycarbonate obtained by the conventional melt polymerization usually contains at a high proportion. Examples of units having different kinds of structures include compounds having the structures shown below, but are not limited to them. In the structural formulas below, (R1)p, (R2)q, and X are the same as shown in the above formula (II). "Y" represents a hydrogen atom, a phenyl group, a methyl group, the structure of the formula (II), or the like.

[0253]

[Chemical Formula 61]



#### "[0302] (2) Aromatic Polycarbonate

The aromatic polycarbonate to be used for the process for producing a highly polymerized aromatic polycarbonate resin of the present invention is a polycondensation polymer having a structure represented by the above general formula (II) as a main repeating unit which is an aromatic polycarbonate prepolymer. The process for producing of the present invention comprises a process wherein the aromatic polycarbonate prepolymer is reacted with the aliphatic diol compound having a structure represented by any one of the above formulas (g1) - (g4) under reduced pressure. According to the present invention, an aromatic polycarbonate resin having advantages of a linked and highly polymerized polycarbonate using a linking agent which enables high molecular weight and high fluidity and also having remarkably improved heat resistance can be obtained while keeping useful properties such as impact resistance that a common polycarbonate originally has.

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#### [0309]

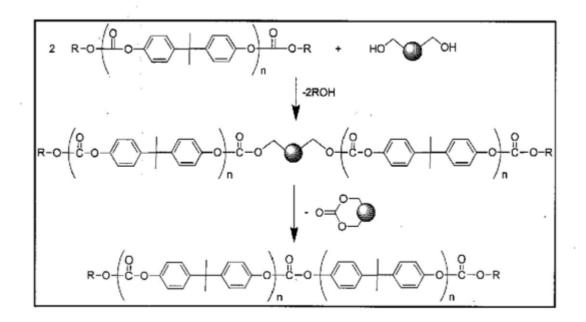
It is also possible to analyze the concentration of terminal hydroxyl groups by

spectrometric measurement using Ti complex. The concentration of terminal hydroxyl groups by this measurement is preferably 1,500 ppm or less, more preferably 1,000 ppm or less. When the concentration of terminal hydroxyl groups is higher than the above range or the concentration of the end-capped terminal groups is lower than the above range, a polymer having sufficiently high molecular weight may fail to be obtained by the transesterification reaction with the aliphatic diol compound."

However, according to the method of the present invention using the aliphatic diol compound having the structure represented by any one of the above formulas (g1) - (g4), a cyclic carbonate having a specific structure is produced as a by-product. After removing the by-product cyclic carbonate from the reaction system, a highly polymerized polycarbonate resin having almost the same skeleton as a polycarbonate homopolymer is obtained. Since the by-product cyclic carbonate has a structure corresponding to the aliphatic diol compound used, it can be assumed to be a ring compound derived from the aliphatic diol compound. However, the reaction mechanism of producing a cyclic carbonate as a by-product simultaneously with the highly polymerizing reaction is not necessarily defined. [0333]

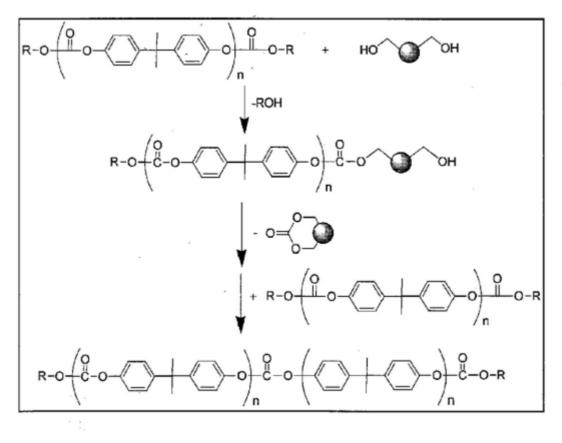
For example, mechanisms shown by the following reaction schemes (1) and (2) are assumed but they are not altogether clarified. The process of the present invention using the aliphatic diol compound having the structures represented by any one of the above formulas (g1) - (g4) is simply characterized in the step of reacting an aromatic polycarbonate prepolymer with an aliphatic diol compound having a specific structure and the step of removing a by-product cyclic carbonate having a structure corresponding to the aliphatic diol compound, and is not necessarily limited to any reaction mechanism. [0334]

Scheme (I) [Chemical Formula 73]





Scheme (II) [Chemical Formula 74]



[0336]

Unlike the case of a highly polymerized polycarbonate copolymer obtained by a linking and highly polymerizing process using an aliphatic diol compound as a linking agent, the highly polymerized polycarbonate resin obtained by the process of the present invention using the aliphatic diol compound having the structure represented by any one of the above formulas (g1) - (g4) contains little copolymerizing components derived from the linking agent, and the skeleton of polymer is almost the same as a polycarbonate homopolymer.

[0337]

Since the highly polymerized aromatic polycarbonate resin obtained by using the above-mentioned aliphatic diol compound substantially does not contain copolymerizing components derived from the aliphatic diol compound as a linking agent or they are extremely few in amount even if contained, it has extremely high heat stability and is excellent in heat resistance, compared to the highly polymerized polycarbonate copolymer obtained by a linking and highly polymerizing process using other aliphatic diol compounds as a linking agent. In addition, while having almost the same skeleton as a polycarbonate homopolymer obtained by a conventional method, the highly polymerized aromatic polycarbonate resin obtained by using the abovementioned aliphatic diol compound has excellent properties such as a low N-value, a low content of units having different kinds of structures, and an excellent color tone, which are the same advantages as those of the highly polymerized polycarbonate resin obtained by a linking and highly polymerizing method using other aliphatic diol compounds. The unit having different kinds of structures here means a branching point unit or the like that a polycarbonate obtained by the conventional melt polymerization usually contains at a high proportion. Examples of the units having different kinds of structures include the units shown in the above description regarding the polycarbonate copolymer but are not particularly limited to them."

"[0355]

# (iii) Cyclic Carbonate-Removing Process

According to the process for producing a highly polymerized aromatic polycarbonate resin of the present invention, an aromatic polycarbonate prepolymer is highly polymerized by the above-mentioned high polymerization reaction, and at the same time, a cyclic carbonate produced by said reaction as a by-product is removed away from the reaction system. By removing the by-product cyclic carbonate from the reaction system, the high polymerization reaction of the aromatic polycarbonate prepolymer is promoted.

[0356]

Examples of methods for removing the cyclic carbonate include distillation wherein the cyclic carbonate is distilled away together with phenol, which is also a by-product, and an unreacted aliphatic diol compound. The temperature of distillation is preferably 260°C to 320°C.

## [0357]

A removal process is carried out for at least a part of the cyclic carbonate. It is most preferable to remove the entirety of the by-product cyclic carbonate. Usually, however, it is not easy to remove completely. In the case where the cyclic carbonate cannot be removed completely, the cyclic carbonate is allowed to remain in the manufactured product of the polycarbonate resin. The preferable remaining amount of cyclic carbonate in the manufactured product is up to 3000 ppm. That is, according to the process of the present invention using the aliphatic diol compound having the structure represented by any one of the above formulas (g1) - (g4), a polycarbonate resin composition containing 3000 ppm or less of cyclic carbonate is obtained, as mentioned later."

# "[0364]

According to the present invention, various additives such as heat stabilizers, antioxidants, pigments, dye enhancing agents, fillers, ultraviolet absorbers, lubricants, mold release agents, crystal nucleating agents, plasticizers, fluidability-improving agents, and antistatic agents can be added to the polymer."

"[0367]

(4) Highly polymerized aromatic polycarbonate resin

The highly polymerized aromatic polycarbonate resin obtained by the process of the present invention using the aliphatic diol compound having the structure represented by any one of the above formulas (g1) - (g4) has a weight average molecular weight (Mw) of 30,000 - 100,000, preferably 30,000 - 80,000, more preferably 35,000 - 75,000. That is, the highly polymerized aromatic polycarbonate resin has high fluidity while being highly polymerized. When the weight average molecular weight of the highly polymerized aromatic polycarbonate resin is too low, blow molding and/or extrusion molding might be difficult because of decline of the melt tension, which might cause a drawdown, to thereby make it difficult to obtain a satisfactory molded product. Injection molding might be difficult because of stringing, which makes it difficult to obtain a satisfactory molded product. Properties such as physical strength and heat resistance of the molded product obtained might be deteriorated. Properties such as one provide a product obtain a satisfactory molded product obtained might be deteriorated. Properties such as nesistance to organic solvents might also be deteriorated because of extension of an oligomer region. When the weight average molecular weight of the highly

polymerized aromatic polycarbonate resin is too high, injection molding of precision components or thin components might be difficult, which may cause an increase of a molding cycle time and a negative influence to the production cost. Therefore, it would be necessary to take countermeasures such as elevating the molding temperature, but problems such as gelation, emergence of different kinds of structures, or an increase in N-value might occur under a high temperature.

[0368]

The highly polymerized aromatic polycarbonate resin obtained by the process of the present invention has a structural viscosity index (N-value) represented by the following mathematical formula (1) of preferably 1.3 or less, more preferably 1.28 or less, most preferably 1.25 or less:

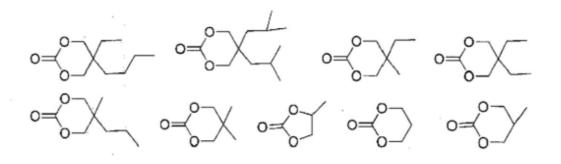
[Mathematical formula 7] N value =  $(\log(Q160 \text{ value}) - \log(Q10 \text{ value}))/(\log 160 - \log 10)...(1)"$ 

# "[0370]

The structural viscosity index (N-value) is an index of a branching degree of an aromatic polycarbonate resin. The highly polymerized aromatic polycarbonate resin obtained by the process of the present invention has a low N-value, which means that the content of a branching structure is low and the content of a linear or straight chain structure is high. In the case of conventional polycarbonate resins having the same Mw, there is a tendency that the fluidity becomes high and the Q-value becomes high when the content of a branching structure or the N-value is increased. In the case of the polycarbonate copolymer of the present invention, on the other hand, high fluidity (high Q-value) can be achieved while keeping the N-value low."

Examples of the above-mentioned cyclic carbonate include the following compounds:

[Chemical Formula 78]



### [0379]

The content of the cyclic carbonate represented by the above formula (h1) in the polycarbonate resin composition of the present invention is 3000 ppm or less, preferably 1000 ppm or less, more preferably 500 ppm or less, most preferably 300 ppm or less. While the lower limit of the content of cyclic carbonate is not particularly limited, it is ideally 0%, usually the detection limit or measurable limit, preferably 0.0005 ppm. When the content of cyclic carbonate is too large, defects such as deterioration of physical strength might occur."

# "[0444]

The present invention will be described in more detail below, referring to examples, which are not intended to limit the scope of the present invention. The measurement values of the examples and comparative examples below were measured by using the following methods and/or devices:

## [0451]

•••

7) Fluidity (Q-Value): Q-value represents an outflow rate of a molten resin (ml/sec), which was evaluated by a molten fluid volume per unit time measured under the conditions of 280°C and 160 kg load after drying at 130°C for 5 hours by using a Koka type flow tester manufactured by Shimadzu Corporation, trade name 'CFT-500D'. [0452]

8) N-value: A sample of aromatic polycarbonate dried at 130°C for 5 hours was subjected to a measurement using a Koka type flow tester manufactured by Shimadzu Corporation, trade name 'CFT-500D'. 'Q160 value' was evaluated with the sample by a molten fluid volume per unit time measured under the conditions of 280°C and 160 kg load, and 'Q10 value' was evaluated in the same manner by a molten fluid volume per unit time measured under the conditions of 280°C and 160 kg load, and 'Q10 value' was evaluated in the same manner by a molten fluid volume per unit time measured under the conditions of 280°C and 10 kg load. N-value was calculated by the following mathematical formula (1) using 'Q160 value' and 'Q10 value':

[0453]

[Mathematical formula 10] N value =  $(\log(Q160 \text{ value}) - \log(Q10 \text{ value}))/(\log 160 - \log 10)...(1)"$ 

"[0488]

<Production example 4 of prepolymer; PP-D>

10,000.6g (43.81 mol) of 2,2-bis(4-hydroxyphenyl)propane, 10,560.0g (49.30 mol) of diphenyl carbonate, and 0.5  $\mu$ mol/mol of cesium carbonate as a catalyst were

charged into a 50-liter SUS reactor equipped with a stirrer and a distiller, and the air in the reactor was substituted with nitrogen. The amount of catalyst was calculated as the number of moles based upon the amount of 2,2-bis(4-hydroxyphenyl)propane. The degree of pressure reduction was adjusted to 27 kPaA (200 torr) and the starting materials were heated and melted at 200°C with stirring for 30 minutes. [0489]

Subsequently, a transesterification reaction was carried out for 4 hours while condensing phenol distilled from the reaction system in a cooling tube and removing, keeping the internal temperature at 260°C and the degree of pressure reduction at 0.13kPaA (1 torr) or less for 1 hour to obtain a polycarbonate prepolymer having the weight average molecular weight (Mw) of 22,000, hereinafter 'PP-D'. The concentration of hydroxyl groups (ppm) and the concentration of terminal phenyl groups (mol %) of the polycarbonate prepolymer thus obtained are shown in Table 5. In Table 5, the concentration of hydroxyl groups means the concentration of all the OH groups contained in the polymer which was calculated by NMR analysis. In Table 5, the concentration of terminal phenyl groups means the concentration of the terminal phenyl groups based on all the phenylene groups and phenyl groups in phenyl terminals, including the phenyl groups substituted by hydroxyl groups. ..."

"[0498] [Table 5]

20	
	PP-D
BPA(g)	10000.6
BPA(moi)	43.81
DPC(g)	10560
DPC(mol)	49.30
DPC/BPA 刊此	1.13
Mw	22000
OH 濃度(ppm)	60
Ph 末端濃度(mol%)	5.0

表5

表 5 Table 5

OH濃度 OH concentration

Ph末端濃度 Ph terminal concentration

"[0503]

<Example 20>

30.13g of the aromatic polycarbonate prepolymer PP-D which was obtained by the above-mentioned Example of Production of Prepolymer 4 was charged into a fourneck flask of 300 cc equipped with a stirrer and a distiller and was heated and melted under normal pressure at 280°C. Subsequently, 0.34g of 2-butyl-2-ethylpropane-1,3diol (BEPD) which was the aliphatic diol compound was charged therein, and the mixture was stirred and kneaded for 3 minutes at a jacket temperature of 280°C under normal pressure.

[0504]

Subsequently, the mixture was stirred and kneaded under reduced pressure of 0.04 kPaA (0.3 torr) for 70 minutes at a jacket temperature of 280°C to carry out a transesterification reaction. Regarding the catalyst, the polymerization catalyst which had been used in the process of preparing the aromatic polycarbonate prepolymer was continuously used directly.

#### [0505]

Phenol, cyclic carbonate (5-butyl-5-ethyl-1,3-dioxane-2-on), and unreacted 2butyl-2-ethylpropane-1,3-diol (BEPD) distilled from the reaction system were condensed in a cooling tube for removal to obtain a polycarbonate resin having a weight average molecular weight (Mw) of 56,400, an N-value of 1.19, and a content of cyclic carbonate (5-butyl-5-ethyl-1,3-dioxane-2-on) of 154 ppm. [0506]

1g of the resin thus obtained was charged into a test tube and was dried for 2 hours by a block heater at 120°C in a nitrogen-substituted glove box with an oxygen concentration of 0.0%. Subsequently, a heat retention test was conducted for 50 minutes in said glove box by a block heater at 360°C. As a result, the molecular weight (Mw) retention rate (%) before and after the heat retention test was 98% and the amount of change in YI-value was +5.0.

# [0507]

A 1H-NMR chart of the mixture at the time that adding BEPD and stirring was completed is shown in Fig. 5. A 1H-NMR chart of the polycarbonate resin finally obtained is shown in Fig. 6. In Fig. 5, the peak of the structure derived from BEPD reacted with the aromatic polycarbonate prepolymer is recognized as a different peak from that of the unreacted BEPD monomer. In Fig. 6, on the other hand, the peak of

the structure derived from BEPD reacted with the aromatic polycarbonate prepolymer and the peak of the unreacted BEPD monomer disappeared. [0508]

From this result, the aromatic polycarbonate resin thus obtained was recognized as a polycarbonate homopolymer which does not have a structure derived from the aliphatic diol compound. It can be assumed that the aromatic diol compound added was once reacted with the aromatic polycarbonate prepolymer and then was removed as a cyclic carbonate.

#### [0509]

#### <Examples 21 to 26>

The experiments were carried out in the same manner as in Example 20 except for using the aromatic polycarbonate prepolymers, the aliphatic diol compounds, and the amounts used as shown in Table 7 to obtain polycarbonate resins. The properties of the polycarbonate resins thus obtained are shown in Table 7.

# [0510]

# <Comparative Example 6>

The experiments were carried out in the same manner as in Example 20; that is, the reaction was carried out in a short time in the same manner as in Example 20, except that the aliphatic diol compound was not added. Mw of the polycarbonate resin thus obtained was 22,000, which was not increased by the reaction.

# [0511]

[Table 7]

				表7				
	実施例20	実施例21	実施例22	実施例23	実施例24	実施例25	実施例26	比較例6
使用 した PP	PP-D	PP-D						
使用したPP(量)	30.13	30.02	29.98	30.05	30.13	30.00	30.06	30.00
脂肪族ジオール種	BEPD	DIBPD	EMPD	DEPD	MPPD	NPG-DI	1,2-PD	無添加
脂肪族ジオールの沸点 (℃)	271	280	226	250	230	-	188	-
脂肪族ジオールの式量	160	188	118	132	132	234	76	-
脂肪族ジオール量 (g)	0.34	0.36	0.19	0.21	0.27	0.52	0.53	
脂肪族ジオール量 (モル)	0.00211	0.00189	0.00163	0.00161	0.00205	0.00220	0.00693	-
PPの全末端量1モルに対するモル数(モル)	0.33	0.29	0.25	0.25	0.32	. 0.34	0.35	-
常圧混練終了時 脂肪族ジオール化合物由来の構造単位(一 般式(1)で表される構造単位)の割合(モ ル%)	1.21	1.14	0.71	0.79	0.93	1.02	0.13	-
得られた樹脂のMw	56400	41700	44000	42700	37000	38600	36100	22000
得られた樹脂のN値	1.21	1.21	1.20	1.19	1.21	1.19	1.20	
得られた樹脂のTg(℃)	149	149	149	149	148	148	148	
得られた樹龍中の 脂肪族ジオール化合物由来の構造単位(一 般式(I)で表される構造単位)の割合(モ ル%)	0.00	0.07	0.09	0.03	0.00	0.34	0.00	
環状カーボネート含量(ppm)	154.211	674.570	23.876	49.286	23,247	4.931	0.042	
360°C50分滞留試験前 Mw	56400	41700	44000	42700	37000	38600	36100	
860℃50分滞留試験後 Mw	55000	38300	33200	40000	31000	30000	22000	
860°C50分滞留試験後 Mw保持率(%)	98	92	75	94	84	78	61	
860°C50分滞留試験前 YI值	0.5	0.9	1,1	0,9	1.2	1.2	0.8	
860°C50分滞留試験後 YI值	6.0	24.5	17.3	16.3	25,1	29.4	18.8	
860°C50分滞留試験前後 YI 值变化量	5.0	23.6	16.2	15.4	23.9	28.2	18.0	

表7 Table 7

実施例 Example

比較例 Comparative Example

無添加 No Additive

使用した P P used

使用した P P (量) PP used (amount)

脂肪族ジオール種 Aliphatic diol species

脂肪族ジオールの沸点(℃) Boiling point of aliphatic diol(C)

脂肪族ジオールの式量 Formula weight of aliphatic diol (g)

脂肪族ジオール量(g) Amount of aliphatic diol (g)

脂肪族ジオール量(モル) Amount of aliphatic diol (mol)

PPの全末端量1モルに対するモル数(モル) Mol number per 1 mol of total amounts of terminals of PP (mol)

常圧混練終了時脂肪族ジオール化合物由来の構造単位(一般式(I)で表され る構造単位)の割合(モル%) A proportion of structural unit (structural unit represented by general formula (I)) derived from aliphatic diol compound after the completion of ambient kneading (mol%)

得られた樹脂のMw Mw of obtained resin

得られた樹脂のN値 N value of obtained resin

得られた樹脂のTg(℃) Tg of obtained resin(C)

得られた樹脂中の脂肪族ジオール化合物由来の構造単位(一般式(I)で表される構造単位)の割合(モル%) A proportion of structural unit (structural unit represented by general formula (I)) derived from aliphatic diol compound in the resultant resin (mol%)

環状カーポネート含量 (ppm) Cyclic carbonate content (ppm)

360℃50分滞留試験前 Mw Mw before the retention test at 360℃ for 50 minutes

360℃50分滞留試験後 Mw Mw after the retention test at 360°C for 50 minutes

360℃50分滞留試験後 Mw保持率(%) Mw retention rate (%) after the retention rate at 360℃ for 50 minutes

3 6 0 ℃ 5 0 分滞留試験前 Y I 値 YI value before the retention test at 360 ℃ for 50 minutes 3 6 0 ℃ 5 0 分滞留試験後 Y I 値 YI value after the retention test at 360 ℃ for 50 minutes

360℃50分滞留試験後 YI值変化量 Variations of YI value after the

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retention test at 360°C for 50 minutes [0512]

As shown in Examples 20-26, a proportion (mol number) of a structure derived from an aliphatic diol compound in a resin finally obtained was remarkably reduced compared to the proportion (mol number) at the time when the addition and kneading of the aliphatic diol compound were completed. According to the process for producing of the present invention using the aliphatic diol compound having the structure represented by any of the above formulas (g1) - (g4), the proportion (mol number) of the structure derived from the aliphatic diol compound in the polymer finally obtained was 50% or less, preferably 40% or less, more preferably 30% or less, further preferably 20% or less, most preferably 10% or less based upon the proportion (mol number) of the structure derived from the aliphatic diol compound in the resin finally obtained at the time that adding the aliphatic diol compound and kneading were completed. [0513]

From the results shown in Examples 20-26, it is clear that the highly polymerized polycarbonate resin obtained by the process of the present invention using the aliphatic diol compound having the structure represented by any one of the above formulas (g1) to (g4) which has a structure similar to a polycarbonate homopolymer is highly excellent in heat stability. Moreover, the high molecular weight (Mw) retention rate (%) is high and the YI-value variation is low before and after the heat retention test of extremely severe conditions of 360°C-50 min."

## 2 The invention described in Cited Document 1

# (1) Cited Invention 1a

Cited Document 1 respectively describes the invention of "high fluidity polycarbonate copolymer" as recited in Claims 1 to 15 of the scope of claims, "a method of producing a highly polymerized aromatic polycarbonate resin" as recited in Claims 16 to 45, and "aromatic polycarbonate compound" as recited in Claims 46 to 51. Further, with regard to "a process for producing a highly polymerized aromatic polycarbonate resin", Claim 16 describes "a method of producing a highly polymerized aromatic polycarbonate resin, comprising the steps of reacting the aliphatic diol compound represented by general formula (g1) with aromatic polycarbonate in the presence of a transesterification catalyst to highly polymerize." Claim 23 depending from Claim 16 and Claims 18, 20, and 22 in this order describes a production method in which "2-butyl-2-ethylpropane-1,3-diol" described as one alternative of "said aliphatic diol compound". Further, paragraph [0367] has a general description of "highly polymerized aromatic polycarbonate resin obtained by a production process using aliphatic diol compound represented by" "general formula (g1)". Paragraphs [0503] to [0508] describe in Example 20 "a highly polymerized aromatic polycarbonate resin" obtained by reacting 2-butyl-2-ethylpropane-1,3-diol as the "aliphatic diol compound" with an aromatic polycarbonate prepolymer produced by production example 4 as the "aromatic polycarbonate" as a specific example of the "highly polymerized aromatic polycarbonate".

Consequently, it is recognized that Cited Document 1 describes in Example 20 using Production Example 4 an invention of:

"10,000.6g (43.81 mol) of 2,2-bis(4-hydroxyphenyl)propane, 10,560.0g (49.30 mol) of diphenyl carbonate, and 0.5 µmol/mol of cesium carbonate as a catalyst were charged into a 50-liter SUS reactor equipped with a stirrer and a distiller, and the air in the reactor was substituted with nitrogen. The amount of catalyst was calculated as the number of moles based upon the amount of 2,2-bis(4-hydroxyphenyl)propane. The degree of pressure reduction was adjusted to 27 kPaA (200 torr) and the starting materials were heated and melted at 200°C with stirring for 30 minutes. Subsequently, a transesterification reaction was carried out for 4 hours while condensing phenol distilled from the reaction system in a cooling tube and removing, keeping the internal temperature at 260°C and the degree of pressure reduction at 0.13kPaA (1 torr) or less for 1 hour, to obtain a polycarbonate prepolymer having a weight average molecular weight (Mw) of 22,000 and an OH concentration (ppm) of 60 ppm (hereinafter sometimes referred to as 'PP-D'). The resultant aromatic polycarbonate prepolymer 'PP-D' in an amount of 30.13 g was fed to a 300 cc four-neck flask equipped with a stirrer and a distiller and was heated and melted under normal pressure at 280°C, and subsequently, 0.34g of 2-butyl-2-ethylpropane-1,3-diol (BEPD) which was the aliphatic diol compound was charged therein, and the mixture was stirred and kneaded for 3 minutes at a jacket temperature of 280°C under normal pressure. Then, the mixture was stirred and kneaded under reduced pressure of 0.04 kPaA (0.3 torr) for 70 minutes at a jacket temperature of 280°C to carry out a transesterification reaction. Regarding the catalyst, the polymerization catalyst which had been used in the process of preparing the aromatic polycarbonate prepolymer was used directly. Phenol, cyclic carbonate (5butyl-5-ethyl-1,3-dioxane-2-on), and unreacted 2-butyl-2-ethylpropane-1,3-diol (BEPD) were distilled from the reaction system and condensed in a cooling tube for removal to

obtain a polycarbonate resin having a weight average molecular weight (Mw) of 56,400, an N-value of 1.19, and a content of cyclic carbonate (5-butyl-5-ethyl-1,3-dioxane-2-on) of 154 ppm" (hereinafter referred to as "Cited invention 1a").

#### (2) Cited Invention 1b

Claim 16 of the scope of claims of Cited Document 1 describes "a process for producing a highly polymerized aromatic polycarbonate resin which comprises the step of reacting an aromatic polycarbonate with an aliphatic diol compound represented by general formula (g1) in the presence of a transesterification catalyst to increase the molecular weight." Claim 26 depending from this Claim 16 describes the production process "comprising a highly-polymerizing process wherein an aromatic polycarbonate is reacted with said aliphatic diol compound in the presence of a transesterification catalyst for high polymerization and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highly-polymerizing process is removed from the reaction system". Claim 27 depending from this Claim 26 describes the production process in which "said cyclic carbonate" is "a compound represented by general formula (h1)".

Further, Claim 39 depending from this Claim 16 describes "a polycarbonate resin composition comprising a highly polymerized aromatic polycarbonate resin obtained by the production process as a main component" and "cyclic polycarbonate represented by the following general formula (h1) in an amount of not more than 3000 ppm"; and Claim 45 depending from this Claim 39 and Claims 41 and 43 in this order describes "the polycarbonate resin composition, wherein said highly polymerized aromatic polycarbonate resin has a structural viscosity index (N-value) represented by the following mathematical formula (1) of 1.25 or less:

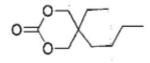
[Mathematical formula 4]

N value =  $(\log(Q160 \text{ value}) - \log(Q10 \text{ value}))/(\log 160 - \log 10)...(1)"$ .

Here, paragraph [0332] to [0336] describe a reaction mechanism regarding "a production method of highly polymerized aromatic polycarbonate resin" as shown in the above item 1. It is recognized from this reaction mechanism that "a by-product cyclic carbonate" is derived from "aliphatic diol compound", and according to the description of [0355] to [0357], the removal of the cyclic carbonate results in a "polycarbonate resin composition" having a content of the cyclic carbonate of a preferable upper limit of 3000 ppm. Thus it is recognized that "cyclic carbonate co-produced" when reacting "aliphatic diol compound (g1)" in Claim 26 depending

from Claim 16 is "a compound represented by general formula (h1)" of Claim 27, and its removal results in "polycarbonate resin composition" "comprising 3000 ppm or less cyclic carbonate represented by general formula (h1)" of Claim 39.

Further, Claim 23 depending from Claim 16 and Claims 18, 20, and 22 in this order describes "2-butyl-2-ethylpropane-1,3-diol" as one alternative of "said aliphatic diol compound", which is allegedly a precursor of by-product cyclic carbonate. As a "by-product cyclic carbonate" of a "compound represented by general formula (h1)" of Claim 27, paragraphs [0378] to [0379] describe "



" (5-butyl-5-ethyl-1,3-dioxan-2-on) as one specific example.

Further, paragraphs [0503] to [0508] describe in Example 20 that as a specific example of the "highly-polymerized aromatic polycarbonate resin obtained by the production method" there is described a "highly-polymerized aromatic polycarbonate resin" obtained by using an aromatic polycarbonate prepolymer produced in production example 4 as the "aromatic polycarbonate", and reacting this with the "aliphatic diol compound" of 2-butyl-2-ethylpropane-1,3-diol, and removing a by-product cyclic carbonate of "5-butyl-5-ethyl-1,3-dioxan-2-on". What was obtained in this Example 20 is one comprising 154 ppm cyclic carbonate of "5-butyl-5-ethyl-1,3-dioxan-2-on", which corresponds to 3000 ppm or less, with a "highly-polymerized aromatic polycarbonate resin" as "a main component". Thus it is recognized that Example 20 describes the "polycarbonate resin composition" in which the aliphatic diol compound is 2-butyl-2-ethylpropane-1,3-diol and the cyclic carbonate is 5-butyl-5-ethyl-1,3-dioxan-2-on.

Consequently, Cited Document 1 discloses the invention of "a polycarbonate resin composition comprising 3000 ppm or less of 5-butyl-5-ethyl-1,3-dioxan-2-on with a highly-polymerized aromatic polycarbonate resin obtained by a method of producing a highly-polymerized aromatic polycarbonate resin comprising a highly-polymerizing process of reacting an aromatic polycarbonate with 2-butyl-2-ethylpropane-1,3-diol in the presence of a transesterification catalyst for highly-polymerization and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highly-polymerizing process is removed from the reaction system as a main component, wherein said highly polymerized aromatic polycarbonate resin has a structural viscosity index (N-value) represented by the following mathematical formula (1) of 1.25 or less:

[Mathematical formula 4] N value =  $(\log(Q160 \text{ value}) - \log(Q10 \text{ value}))/(\log 160 - \log 10)...(1)$ " (hereinafter referred to as "cited invention 1b").

3 Description of Cited Document 2

Cited Document 2 describes the following matters:

# "[Claim 1]

An aromatic polycarbonate resin composition comprising 100 weight parts of aromatic polycarbonate and 1 to 300 weight parts of inorganic filler, wherein said aromatic polycarbonate has a content of a cyclic oligomer represented by formula (I) of 1000 ppm or less and the proportion of the total amount of oligomers represented by formulae (I), (II), and (III) satisfies the relationship (1), wherein said polycarbonate is produced by the reaction of an aromatic dihydroxy compound with a compound capable of introducing a carbonate bond and having a viscosity average molecular weight of 12000 to 40000. ... (Omitted)...

#### [Claim 2]

The aromatic polycarbonate resin composition of Claim 1, wherein said inorganic filler is at least one kind or two or more kinds selected from glass or fibrous, powder or flake inorganic fillers of carbon.

[Claim 3]

The aromatic polycarbonate resin composition of Claim 1 or 2, wherein said aromatic polycarbonate is produced by transesterification reaction of the aromatic dihydroxy compound and diester carbonate.

# [Claim 4]

The aromatic polycarbonate resin composition of any one of Claims 1 to 3, wherein said aromatic polycarbonate has a content of terminal hydroxyl group of 100 to 1000 ppm."

#### "[0002]

### [Conventional Art]

Polycarbonate resin is widely used in various fields as a resin excellent in impact resistance and transparency. As a mold of a housing and cover for mobile phones becomes thinner, a material further excellent in strength and rigidity is required. In general, the requirement for strength and rigidity corresponds to the mixture of inorganic filler such as glass fiber and carbon fiber that performs a reinforcing effect of resin. However, the mixture of these inorganic fillers generally reduces fluidity of polycarbonate resin composition. As a result, a polycarbonate resin composition must be kneaded and molded at a temperature as high as 290 to 380°C, and has a problem of being easily colored in kneading and molding, and further has a problem of being easily colored in a high-temperature use condition. For these problems, in a polycarbonate produced by an interface method of reacting an aromatic dihydroxy compound and phosgene for polymerization, there are attempts to improve heat resistance by the reduction of amount of methylene chloride and the reduction of chlorine component derived from phosgene. Further, in a transesterification method causing an aromatic dihydroxy compound and carbonate diester compound to be reacted under reduced pressure, there has also been an attempt to improve heat resistance by the reduction of the content of a raw material of carbonate diester, etc. and the deactivation of catalyst (Japanese Unexamined Patent Application Publication No. H7-126374); however, sufficient heat stability has not yet been achieved. On the other hand, an attempt has already been made to add a phosphorous-based antioxidant such as phosphite ester and an antioxidant such as hindered phenol to polycarbonate for imparting heat resistance, but the coloring at a high temperature cannot be sufficiently suppressed. Further, there are attempts to reduce molecular weight of polycarbonate based resin, to mix a fluidity modifier such as carbonate oligomer, and to modify fluidity by alloying with another resin such as ABS. These methods have had a problem such as sacrifice of the impact strength and heat resistance of an original polycarbonate.

# [0003]

[Problem to be solved by the Invention]

The present invention provides a polycarbonate composition excellent in color tone and heat resistance, and in particular provides an aromatic polycarbonate resin composition excellent in color tone, heat resistance, etc. reinforced by an inorganic filler."

# "[0028]

An inorganic filler used in the present invention may include, for example, glass fiber, glass milled fiber, glass flake, glass beads, carbon fiber, silica, alumina, titanium oxide, calcium sulfate powder, calcium sulfate, calcium sulfate whisker, barium sulfate, talc, mica, calcium silicate, carbon black, graphite, iron powder, copper powder, molybdenum disulfide, silicon carbide, silicon carbide fiber, silicon nitride, silicon nitride fiber, brass fiber, stainless fiber, potassium titanate fiber or whisker, aromatic polyamide fiber, preferably glass, or fibrous, powder, or flake filler of carbon, more preferably glass fiber, carbon fiber, glass milled fiber, glass flake, etc. A mixing amount of inorganic filler is from 1 to 300 weight parts with respect to 100 weight parts

aromatic polycarbonate of the present invention. If the mixing amount of inorganic filler is less than 1 weight part, reinforcing effects are poor. If it exceeds 300 weight parts, appearance tends to be poor. A preferable mixing amount is 3 to 50 weight parts. Glass fiber and glass milled fiber used in the present invention may be either of thermoplastic resin, and an alkali-free glass (E glass) is preferable. A diameter of glass fiber is preferably 6 to 20  $\mu$ m, more preferably 9 to 14  $\mu$ m. If a fiber diameter is less than 6  $\mu$ m, the reinforcing effects tend to be insufficient. If the fiber diameter exceeds 20  $\mu$ m, it tends to cause an adverse effect on product appearance. Glass fiber may preferably include a chopped strand cut into a length of 1 to 6 mm, and glass milled fiber may preferably include commercially available ones ground into a length of 0.01 to 0.5 mm, or they may be mixed for use. Glass fiber used in the present invention may be subjected to a surface treatment by a silane coupling agent such as amino silane and epoxy silane for the purpose of the improving adhesiveness with a resin, or subjected to a sizing treatment by an acrylic resin or urethane resin for the purpose of improving handleability for use.

#### [0029]

Glass beads used in the present invention may include any thermoplastic resin, and an alkali-free glass (E glass) is preferable. Glass bead has a spherical shape and preferably has a particle diameter of 10 to 50 µm. Glass flake used in the present invention may include scale-like glass flake. Generally, a maximum diameter after mixing resin is 1000  $\mu$ m or less, preferably 1 to 500  $\mu$ m, and an aspect ratio (a ratio of maximum diameter to thickness) is 5 or more, preferably 10 or more, further preferably 30 or more. Carbon fiber used in the present invention is generally produced by sintering raw materials such as acrylic fiber, petroleum or carbon-based special pitch, cellulose fiber, and lignin, including various types such as flameproof, carbonaceous, or plumbaginous, but no particular limitation is imposed on a substrate. An average of aspect ratio (fiber length/fiber diameter) of carbon fiber is preferably 10 or more. If an average of aspect ratio is less than 10, electroconductivity, strength, and rigidity are reduced. An average of aspect ratio (fiber length/fiber diameter) of carbon fiber is more preferably 50 or more. In general, a diameter of carbon fiber is 3 to 15  $\mu$ m. To adjust to such an aspect ratio, any shape such as chopped strand, robing strand, and milled fiber is used, and one kind or two or more kinds may be mixed for use."

# 4 Description of Cited Document 3

Cited Document 3 describes the following matters:

# "[Examples]

[0091]

The present invention is explained hereinafter in further detail with reference to examples, but the present invention is not restricted by these examples. Further, an evaluation was implemented for the following items.

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•••
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[0093]

(ii) Impact resistance (Measurement of Charpy impact strength)

The evaluation was implemented by preparing an impact test piece by injection molding and measuring notched Charpy impact strength in compliance with ISO179.

•••

[0096]

As a raw material, the following is used.

(Component A)

A-1: Linear aromatic polycarbonate resin powder synthesized by an interface polycondensation method from bisphenol A and an end stopping agent of p-tertbutylphenol and phosgene (manufactured by Teijin Chemical, product name: Panlite L-1225WS, viscosity average molecular weight of 20,900)

•••

[0099]

(Component D)

D-1: Glass fiber (diameter: 13  $\mu$ m, cut length: 3mm) [ECS-03T-511 (product name) manufactured by Nippon Electric Glass Company, Limited]

D-2: Glass milled fiber (diameter: 9 µm, numerical average fiber length: 30 µm) [PEF-301S (product name) manufactured by Nitto Boseki Co., Ltd.]

D-3: Talc (plate-like, average particle size 2  $\mu$ m) [manufactured by Hayashi Kasei Co., LTD., Upn HS-T0.8 (product name)]

D-4: Flat Cross section Glass fiber (long diameter: 28  $\mu$ m, short length: 7  $\mu$ m, cut length: 3mm) [CSG 3PA-830 (product name) manufactured by Nitto Boseki Co., Ltd.]

•••

[0102]

[Examples 1 to 55, Comparative Examples 1 to 10]

(Production of polycarbonate resin composition)

To a polycarbonate resin powder produced from bisphenol A and phosgene by an interface polycondensation method there were added various additives described in

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Table 1 to Table 6 in the respective mixing amounts, and after mixing with a blender, the mixture was melted and kneaded by use of a vent-type biaxial extruder (manufactured by THE JAPAN STEEL WORKS, LTD., TEX30 $\alpha$  (complete engagement, rotation in same direction, double-threaded screw)) to obtain a pellet. A preliminary mixture of additives and polycarbonate resin powder was prepared at a concentration ten times the mixing amount by a Henschel mixer and the whole was mixed by a blender. The extrusion conditions are an ejection amount of 20 kg/h, a screw rotation number of 150 rpm, and a degree of vacuum of vent of 3 kPa, and the extrusion temperature was set to 260°C from a first supply port to a die part.

Subsequently, the resultant pellet was dried by a hot air circulation drier for 5 hours at 100°C, and after drying, sample pieces for various evaluation were molded at a cylinder temperature of 280°C and a die temperature of 70°C by an injection molding machine (Sumitomo Heavy Industries, Ltd., SG-150U). Each property was measured for these molded products. The results are shown in Table 1 to Table 6."

# "[0105]

[Table 3]

			単位	実施例19	実施例20	実施例21	実施例22	実施例23	実施例24	実施例25	実施例26	実施例27	実施例28	実施例29	実施例30
	A成分	A-1		97	91	91	84	97	91	91	84	97	84	97	84
	B成分	B-1	3	3	9	9	16	3	9	9	16	3	16	3	16
		B-2													~
	C成分	C-1											0.00		
		C-2		1996											
		C-3			· · · · ·			0				3			
		C-4		0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
	(合計)			100	100	100	100	100	100	100	100	100	100	100	100
40 -	D成分	D-1		10	10		10	20	20		20	5	5	10	10
組成		D-2	重量部									5	5	20	20
		D-3													
		D-4			2	10				20					
	その他	EW		0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
		TMP		0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
	耐熱性			135	120	120	105	138	122	122	107	125	95	127	100
	耐衝撃性	kJ/m <sup>3</sup>		5	3	3	2	6	1	4	3	7	7 ·	5	5
	耐加木分解件	ΔMv		2,100	3,900	3,900	7,500	2,200	4,100	4,100	7,700	2,300	9,500	3,100	10,200
	難然性	2. Ommt		V O	V O	V-0	V-0	V-0	V-0	V-0	V-0	∨–0	V-0	V-0	V-0

単位 Unit

- 実施例 Example
- A成分 Component A
- B成分 Component B
- C成分 Component C
- D成分 Component D
- 組成 Composition
- 合計 Total
- その他 Other
- 耐熱性 Heat resistance

耐衝擊性 Impact resistance

耐加水分解性 Hydrolysis resistance

難燃性 Retardancy

重量部 Weight part

# [0106]

[Table 4]

			単位	実施例31	実施例32	実施例33	実施例34	実施例35	実施例36	実施例37	実施例38	実施例39
	A成分	A-1	0	97	84	84	97	91	91	84	91	84
	B成分	B-1		3	16	16	3	9	9	16	9	16
		B-2										
	C成分	C-1										
	Dev See	C-2										
		C-3										
		C-4		0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
	合計			100	100	100	100	100	100	100	100	100
組成	D成分	D-1	重量部	20	20		30	30		30		
和日月发		D-2	王重即	10	10	10					1000	
		D-3	0			2 200	anteres en		100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100			
1		D-4	i.			20	ez.		30		50	50
	その他	EW		0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
		TMP		0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
1 1	耐熱的			130	102	102	139	123	123	108	125	110
	耐衝撃性	kJ/m <sup>3</sup>		6	6	6	7	5	5	4	10	6
	耐加水分解性	ΔMv		2,800	9,200	9,800	2,400	4,300	4,300	8,000	4,600	8,400
	難燃性	2. Ommt		V-0	V-0	V-0	V-0	V-0	V~0	V-D	V-0	V-0

単位 Unit

実施例 Example

A成分 Component A

B成分 Component B

C成分 Component C

D成分 Component D

組成 Composition

合計 Total

その他 Other

耐熱性 Heat resistance

耐衝擊性 Impact resistance

耐加水分解性 Hydrolysis resistance

難燃性 Retardancy

重量部 Weight part

"

5 Description of Cited Document 4

Cited Document 4 discloses the following matters:

"[0158] [Impact resistance]

An ISO multipurpose sample piece obtained by the aforementioned method (3 mmt) was used in a notched Charpy test in compliance with ISO179 (unit:  $kJ/m^2$ ) and was evaluated. Note that the results are represented as "impact resistance" in Tables 3 to 4."

"[0159]

[Table 2]

	略号	材 料
		界面重合法で製造されたビスフェノールAを出発原料とする
ポリカーボ		芳香族ポリカーボネート樹脂、粘度平均分子量:19000
ネート樹脂 (A)	A	三菱エンジニアリングプラスチックス社製、
		商品名:ユービロンH3000
		長径28μm、短径7μm、
	B-1-1	長径と短径の比:4
	B-1-1	日東紡績社製、商品名:CSG 3PA-830
扁平断面 ガラス繊維		ウレタン/エポキシ系収束剤
(B-1)		長径20μm、短径10μm、
	B-1-2	長径と短径の比:2
	B-1-2	日東紡績社製、商品名:CSG 3PL-822
		ウレタン系収束剤
	B-2-1	平均厚み5μm、平均粒径600μmのガラスフレータ
ガラス フレーク	D-2-1	日本板硝子社製、商品名:フレカREFG101
(B-2)	B-2-2	平均厚み0.7μm、平均粒径160μmのガラスフレーク
	D-2-2	日本板硝子社製、商品名:フレカMEG160FYX
ホスファ	с	フェノキシホスファゼン化合物
ゼン(C)		伏見製薬社製、商品名:ラビトルFP-110
フルオロ	D	フィブリル形成能を有するフルオロポリマー
ポリマー(D)	2	ダイキン工業社製、商品名:FA-500H
スチレン系樹	Е	AS樹脂
脂(E)	L	テクノポリマー社製、商品名:SAN290FF
		熱安定剤
	F 1	トリス (2, 4ージーtert-ブチルフェニル) ホスファイト
		ADEKA社製 商品名:アデカスタブ2112
		熱安定剤
その他の 成分	F 2	オクタデシルー3-(3,5-ジ-tert-プチル
		- 4-ヒドロキシフェニル) プロピオネート
		BASF社製、商品名:イルガノックス1076
		滑剤
		ポリエチレンワックス
I		クライアント社製、商品名:リコワックスPE520
リン酸	C	レゾルシノールピス-2,6-キシレニルホスフェート、
エステル	G	大八化学工業社製、商品名: PX-200
円形断面		直径10μm、カット長3mmのチョップドガラス繊維、
门形时间	H	and the second s

略号 Abbreviated symbol

材料 Materials

ポリカーボネート樹脂 Polycarbonate resin 扁平断面ガラス繊維 Flat cross sectional glass fiber ガラスフレーク Glass flake ホスファンゼン (C) Phosphanzen (C) フルオロポリマー (D) Fluoropolymer (D) スチレン系樹脂(E) Styrene resin (E) その他の成分 Other components リン酸エステル Phosphate ester 円形断面ガラス繊維 Round-shaped cross sectional glass fiber 界面重合法で製造されたビスフェノールAを出発原料とする Bisphenol А produced by an interface polymerization method is a starting material 芳香族ポリカーボネート樹脂、粘度平均分子量:19000 Aromatic polycarbonate resin, viscosity average molecular weight: 19000 三菱エンジニアリングプラスチックス社製、 Manufactured by Mitsubishi **Engineering Plastics**, 商品名:ユーピロンH3000 Product name: Euperon H3000 長径28µm、短径7µm、 Long diameter 28  $\mu$ m, Short diameter 7  $\mu$ m, 長径と短径の比:4 Ratio of long diameter to short diameter: 4 日東紡績社製、商品名:CSG 3 P A - 8 3 0 Manufactured Nitto by Boseki Co., Ltd., Product name: CSG 3PA-830 ウレタン/エポキシ系収束剤 Urethane/epoxy sizing agent 長径20µm、短径10µm、 Long diameter 20 µm, Short diameter 10 µm, 長径と短径の比:2 Ratio of long diameter to short diameter: 2 日東紡績社製、商品名:CSG 3 P L - 8 2 2 Manufactured by Nitto Boseki Co., Ltd., Product name: CSG 3PL-822 ウレタン系収束剤 Urethane-based sizing agent 平均厚み5µm、平均粒径600µmのガラスフレーク Glass flake with an average thickness of 5  $\mu$ m and an average particle diameter of 600  $\mu$ m 日本板硝子社製、商品名:フレカREFG101 Manufactured by Nippon Sheet Glass Company, Limited, Product name: Fleka REFG-101 平均厚み0.7µm、平均粒径160µmのガラスフレーク Glass flake with an average thickness of 0.7  $\mu$ m and an average particle diameter of 160  $\mu$ m 日本板硝子社製、商品名:フレカREFG101 Manufactured by Nippon Sheet Glass Company, Limited, Product name: Fleka MEG160FYX フェノキシホスファゼン化合物 Phenoxyphosphazene compound 伏見製薬社製、商品名: ラビトルFP-110 Manufactured by FUSHIMI Pharmaceutical Co., Ltd., Product name: Rabitle FP-110 フィルブリル形性能を有するフルオロポリマー Fluoropolymer having fibril-type performance ダイキン工業社製、商品名:FA-500H Product name: FA-500H AS樹脂 **AS** Resin テクノポリマー社製、商品名: SAN290FF Manufactured by Technopolymer, Product name: SAN290FF 熱安定剤 Heat Stabilizer トリス(2, 4-ジーtert-ブチルフェニル)ホスファイト Tris (2,4-di-tert-butylphenyl)phospite ADEKA社製、商品名:アデカスタブ2112 Manufactured by ADEKA, Product name: ADK STAB 2112 熱安定剤 Heat Stabilizer オクタデシル-3-(3, 5-ジーtert-ブチル-4-ヒドロキシフェニ ル) プロピオネート Octadecyl-3-(3,5-di-tert-butyl-4hydroxyphenyl)propionate BASF社製、商品名:イルガノックス1076 Manufactured by BASF, Product name: Irganox 1076 滑剤 Lubricants ポリエチレンワックス Polyethylene wax クライアント社製、商品名:リコワックスPE520 Manufactured by Client, Product name: LICOWAX PE520 レゾルシノールビス-2、6-キシレニルホスフェート、 Resorcinol bis-2,6-xylenylphosphate, 大八化学工業社製、商品名: PX-200 Manufactured by DAIHACHI CHEMICAL INDUSTRY CO., LTD., Product name: PX-200 直径10µm、カット長3mmのチョップドガラス繊維、 Chopped glass fiber having a diameter of 10  $\mu$ m and a cut length of 3 mm, 日本板硝子社製、商品名: ECS03T-187H Manufactured by Nippon Sheet Glass Company, Limited, Product name: ECS03T-187H

[0160] [Table 3]

	略号	単位	実施例					
	醉方		1	2	3	4	5	6
樹	А	質	51.02	51.02	41.02	52.02	51.02	51.02
	B-1-1		30	30	35	20		
	B-1-2						30	20
	B-1-3							
	B-2-1		10		15	20	10	20
	B-2-2			10				
脂	С	量	8	8	8	11	8	8
組成	D	部	0.4	0.4	0.4	0.4	0.4	0.4
	Е						5	5
	F 1		0.03	0.03	0.03	0.03	0.03	0.03
	F 2		0.05	0.05	0.05	0.05	0.05	0.05
	F 3		0.5	0.5	0.5	0.5	0.5	0.5
	G							
	Н							
UL難燃性			V-0	V-0	V~0	V-0	V-0	V-0
弾性率		GРа	12.2	12.4	13.5	11.2	11.6	10.5
低ソリ性			0.75	0.74	0.79	0.81	0.73	0.8
耐衝	撃性	$k J/m^2$	15	16	16	13	14	12
耐熱性		ĉ	113	113	113	107	111	111

略号 Abbreviated symbol

単位 Unit

実施例 Example

樹脂組成 Resin composition

質量部 Mass part

UL 難燃性 UL retardancy

弹性率 Elastic modulus

低ソリ性 Low warpage

耐衝擊性 Impact resistance

## 耐熱性 Heat resistance

# 6 Description of Cited Document 5

Cited Document 5, which is newly cited by the body as a document showing common general knowledge as of the filing of the present application, describes the following matters:

Cited Document 5: Seiichi HOMMA, Handbook of polycarbonate resin, Japan, 1992, pp. 82, 106-108

# (1)

## "4. 5. 1 Glass fiber reinforcing grade

For the purpose of improving strength, rigidity, and dimensional stability, a grade filled with 10 to 30% glass fiber is commonly used." (page 82, lines 12 to 14)

# (2)

# "5. 2 Composite reinforcing grade

Polycarbonate resin is widely used, taking advantage of its transparency and impact resistance. On the other hand, in a case where further high mechanical strength or heat resistance and dimensional stability are required, it becomes popular to make a composite with a reinforcing material. In particular, for the use of precision components that requires dimensional stability, a noncrystalline resin of polycarbonate has a smaller mold shrinkage factor compared to a crystalline resin such as polyamide (PA) and polyoxymethylene (POM), and is thus advantageous. Utilizing this feature, there are many commercially available grades in which polycarbonate is mixed with an inorganic material such as glass fiber and carbon fiber in order to obtain a material having rigidity similar to metals and a smaller linear expansion coefficient. Further, many attempts have been made to alloy with the other resins and further mix a reinforcing material with this alloy for improvement of properties such as chemical resistance, low-temperature impact resistance, and fluidity, which are defects of polycarbonate." (page 106, lines 8 to 20)

"5. 2. 1 Purpose of making a composite and reinforcing materials

The objective of making a composite is set forth as below:

(a) Reinforcing effects; Aim to improve mechanical properties and heat resistance

(b) Bulking effects; Aim to reduce cost by the addition of reinforcing material when using an expensive base material

(c) Dimensional stability; Improve dimensional stability of materials due to decrease in linear expansion coefficient and water absorbability by the addition of reinforcing material

(d) Impart special function; Impart special function such as electroconductivity, magnetic property, and flame retardancy that a base polymer does not originally have in addition to the reinforcing effects of (a)

(e) Others; Improve shielding effects, drip prevention, vibration suppression, weather proofing, printing performance, and adhesiveness

For these purposes, a reinforcing material shown in Table 5.4 is used. In a case of polycarbonate, when a reinforcing material is basic or strongly acidic, a polymer decomposes in melt kneading, thereby causing a decrease in molecular weight, and therefore it is necessary to formulate very carefully for the use of the polymer.

Most representative reinforcing materials may include glass fiber (hereinafter GF) and carbon fiber (hereinafter CF), and a mixture with a reinforcing material such as talc, mica, clay, potassium titanate whisker, and zinc oxide whisker is currently under development.

機能	特 性	複合素材(例)			
	引張り特性	CF, GF, 有機質繊維(合成繊維)			
力学的機能	圧 縮 特 性	マイカ、ガラスフレーク			
	衝 撃 特 性	GF, 繊維質材料			
	摩擦・摩耗特性	CF, 黒鉛, 硫化モリブデン, PTFE			
_	絶縁特性	マイカ、クレー			
電気的機能	尊 電 性	CF. CB, 金属粉および繊維 (Au. Ag. Ni. Cu Al), 金属コーティングフィラー			
	耐熱性	CF. GF. タルク, マイカ			
熱的機能	耐燃性(難燃性)	水酸化アルミニウム,水酸化マグネシウム,三酸化アンチモン,三酸化モリブデン,含水無機 材料			
磁気的機能	磁気特性	磁性材料(フェライト、希土コバルトなど)			
防音機能	遮 音 性	鉛,鉄粉,酸化鉄,砂鉄			

**夷5.4** プラスチックの複合化により付与される機能と複合素材(充塡材)

表 5.4 プラスチックの複合化により付与される機能と複合素材(充填材) Table 5.4 Function imparted by composite plastic and composite material (filler) 機能 Function 特性 Characteristics 複合素材 (例) Composite material (example) 力学的機能 **Kinetic Function** 電気的機能 Electric function 熱的機能 **Heating Function** 磁気的機能 **Magnetic Function** 防音機能 Noise insulation Function 引張り特性 Tensile characteristics 圧縮特性 **Compression characteristics** 衝擊特性 Impact characteristics 摩擦・摩耗特性 Abrasion characteristics 絶縁特性 Insulation properties 導電性 Electroconductivity 耐熱性 Heat resistance 耐燃性 (難燃性) Retardancy 磁気特性 Magnetic properties 遮音性 Noise shielding performance 有機質繊維(合成繊維) Organic fiber (synthetic fiber) マイカ Mica ガラスフレーク Glass flake 繊維質材料 **Fibrous Materials** 黒鉛、硫化モリブデン Graphite, molybdenum sulfide クレー Clay 金属粉および繊維 Metal powder and fiber 金属コーティングフィラー Metal coating filler タルク Talc 水酸化アルミニウム、水酸化マグネシウム、三酸化アンチモン、三酸化モリブ デン、含水無機材料 Ammonium hydroxide, magnesium hydroxide, antimony trioxide, molybdenum trioxide, aqueous inorganic materials 磁性材料(フェライト、希土コバルトなど) Magnetic material (ferrite, rare earth cobalt, etc.) 鉛、鉄粉、酸化鉄、砂鉄 Lead, iron powder, ferrous oxide, iron sand ガラス繊維 Glass fiber 炭素繊維 Carbon fiber カーボンブラック Carbon black

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" (page 106, line 2 from the bottom to page 108, line 1)

## (4)

"The shape of fiber may include a roving bundled by a sizing agent and wound by a drum, a chopped strand (hereinafter CS) cut into several mm length, and powder-milled fiber (hereinafter MF) that grinds a glass fiber to have a length of several 10  $\mu$ m as well as glass flake and spherical beads." (page 108, lines 5 to 8)

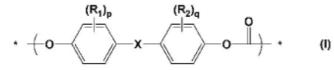
No. 5 Comparison between Invention 1 and Cited Invention 1a and the determination

## 1 Comparison

Invention 1 and Cited Invention 1a are compared.

"Aromatic polycarbonate resin" of Cited Invention 1a is obtained by subjecting "30.13 g 'aromatic polycarbonate prepolymer 'PP-D' obtained" from "10,000.6 g 2,2-bis(4-hydroxyphenyl)propane (43.81 mol) and 10,560.0 g diphenylcarbonate (49.30 mol)" and "0.34 g of 2-butyl-2-ethylpropane-1,3-diol (BEPD) as an aliphatic diol compound" to a transesterification reaction" and it has "a weight average molecular weight (Mw) of 56,400", and thus it corresponds to "an aromatic polycarbonate resin having a structural unit represented by the following general formula (I) and a weight average molecular weight of 40,000 to 100,000" of Invention 1

"[Chemical Formula 1]



(where p and q represent 0, and X represents a group selected from the group of the following (Ia))

[Chemical Formula 2]

(where R3 and R4 each represent a methyl group)".

Further, "cyclic carbonate (5-butyl-5-ethyl-1,3-dioxan-2-on)" of Cited Invention

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1a is a "cyclic carbonate represented by the following general formula (IIa)" of Present Invention 1 corresponds to

"[Chemical Formula 3]

(where Ra represents an ethyl group, and Rb represents a butyl group)." Further, "aromatic polycarbonate resin" of Cited Invention 1a "comprises 154 ppm" "cyclic carbonate (5-butyl-5-ethyl-1,3-dioxan-2-on)", and thus contains "cyclic carbonate represented by general formula (IIa)" of Present Invention 1, and the content overlaps "0.1 ppm or more to 3000 ppm or less" of Present Invention 1.

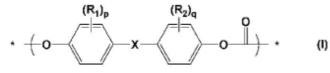
Further, "aromatic polycarbonate resin" of Cited Invention 1a and "aromatic polycarbonate resin composition" of Invention 1 have in common that they are "aromatic polycarbonate resins".

Consequently, these inventions have the following Corresponding Feature:

# <Corresponding Feature>

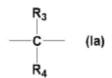
"An aromatic polycarbonate resin composition comprising an aromatic polycarbonate resin having a structural unit represented by the following general formula (I), and a weight average molecular weight of 40000 to 100000 and a cyclic carbonate represented by the following general formula (IIa), wherein the content of cyclic carbonate represented by the following general formula (IIa) is 0.1 ppm or more to 3000 ppm or less on a basis of the aromatic polycarbonate resin having a structural unit represented by the following general formula (I):

[Chemical Formula 1]

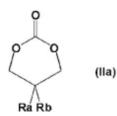


(where p and q represent 0. X represents a group selected from the group of the following (Ia))

[Chemical Formula 2]



(where R3 and R4 each represent a methyl group) [Chemical Formula 3]



(where Ra represents an ethyl group, and Rb represents a butyl group)"

Further, these inventions differ from each other in the following Different Features:

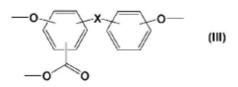
<Different Feature 1>

Invention 1 specifies that "said aromatic polycarbonate resin has a terminal hydroxyl group concentration of 1000 ppm or less", whereas Cited Document 1 is silent about the terminal hydroxyl group concentration of aromatic polycarbonate resin in Cited Invention 1a.

<Different Feature 2>

Invention 1 specifies that "said aromatic polycarbonate resin comprises a structural unit represented by the following general formula (III), and the content is less than 2000 ppm in an aromatic polycarbonate resin having a structural unit represented by said general formula (I)"

"[Chemical Formula 5]



(where X has the same meaning as X in general formula (I)).", whereas Cited Invention 1a does not specify as such.

#### <Different Feature 3>

Invention 1 is an aromatic polycarbonate "resin composition" comprising an "inorganic filler", and "said inorganic filler is at least one kind selected from the group consisting of glass fiber, glass beads, and glass flake, and the content of said inorganic filler is 3 to 200 mass parts on 100 mass parts of aromatic polycarbonate resin having a structural unit represented by the following general formula (I), whereas Cited Document 1 fails to specify the inclusion of inorganic filler, and Cited Invention 1a is an aromatic polycarbonate "resin".

# 2 Judgment

## (1) Regarding Different Feature 1

The aromatic polycarbonate resin of Cited Invention 1a is obtained by subjecting "30.13 g aromatic polycarbonate prepolymer 'PP-D'" and "0.34 g of 2-butyl-2ethylpropane-1,3-diol (BEPD) as an aliphatic diol compound" to a "transesterification reaction" with a polymerization catalyst which had been used in the process of preparing the aromatic polycarbonate prepolymer being used directly, and "removing from the reaction system" phenol, cyclic carbonate (5-butyl-5-ethyl-1,3-dioxane-2-on), and unreacted 2-butyl-2-ethylpropane-1,3-diol (BEPD)", wherein an aromatic polycarbonate resin has an OH concentration of aromatic polycarbonate prepolymer of 60 ppm. Here, Cited Document 1 describes in paragraphs [0332] to [0336] a reaction mechanism of "a method of producing a highly-polymerized aromatic polycarbonate resin" as shown in the above No. 4, 2(1), and the resultant aromatic polycarbonate resin has a drastically less structural unit derived from aliphatic diol compound as a linking agent, and the skeleton of the resin is almost the same as homopolycarbonate resin. It can be seen from this reaction mechanism that "aromatic polycarbonate resin" obtained by "a method of producing aromatic polycarbonate resin" in which an aromatic polycarbonate prepolymer is highly-polymerized by an aliphatic diol compound has a lower terminal hydroxyl group concentration compared to a terminal hydroxyl group concentration of the raw material of aromatic polycarbonate prepolymer. Further, Cited Document 1 describes in [Table 7] that "a proportion of a structural unit derived from an aliphatic diol compound in a resultant resin" of Example 20 is 0.00%. In view of this, the reaction in Example 20 is in compliant with the above reaction mechanism. Thus it is obvious that an aromatic polycarbonate resin of Cited Invention 1a has a lower terminal hydroxyl group concentration than 60 ppm of prepolymer "PP-D".

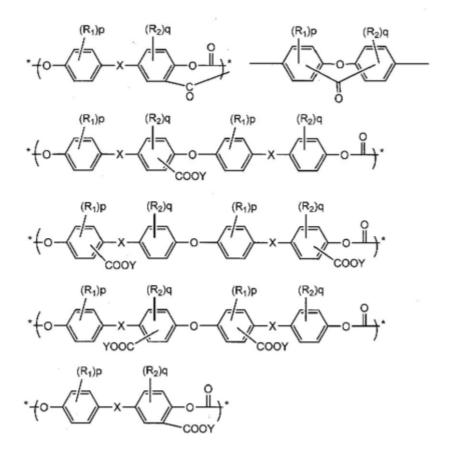
Therefore, the aromatic polycarbonate resin of Cited Invention 1a has a terminal

hydroxyl group concentration of 1000 ppm or less.

Therefore, Different Feature 1 is not a substantial difference.

# (2) Regarding Different Feature 2

Cited Invention 1a is an aromatic polycarbonate resin having a N value of 1.19. Regarding this N value, Cited Document 1 describes in paragraph [0370] that "the structural viscosity index (N-value) is an index of a branching degree of an aromatic polycarbonate resin, and a polycarbonate copolymer (body's note: a typo of "aromatic polycarbonate resin") of the Invention has a low N-value, which means that the content of a branching structure is low and the content of a linear or straight chain structure is high". Here, regarding the description of lower content of this branched structure, paragraph [0337] describes that a highly-polymerized aromatic polycarbonate resin using a linking agent has an advantage of a low N value and a lower proportion of a unit having a different kind of structure, and this unit having a different kind of structure is neans a branched unit, and specific examples of units having different kinds of structures included in the highly-polymerized aromatic polycarbonate resin may include the units shown in the above description regarding the polycarbonate copolymer". Further, for these "different kinds of structures making a reference to polycarbonate copolymer", paragraphs [0252] to [0253] provide examples "

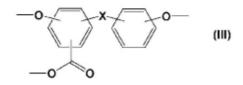


The exemplified structure includes a structure represented by general formula (III) of Invention 1.

".

Consequently, an aromatic polycarbonate resin of Cited Invention 1a is construed as including a unit having a different kind of structure exemplified as above, and the unit corresponds to "include a structural unit represented by the following general formula (III)" of Invention 1:

"[Chemical Formula 5]



(where X has the same meaning as X in general formula (I))." Further the "unit having a different kind of structure" is a "branched structure". As aforementioned, Cited Document 1 discloses a lower proportion of branched structure. Thus the aromatic polycarbonate resin of Cited Invention 1a has lower content of the "unit having a different kind of structure".

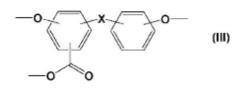
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Here, paragraph [0054] of the specification of the present application describes that "an aromatic polycarbonate resin having a lower proportion of different kinds of structure may be produced by a method comprising, for example, a highly-polymerizing process in which an aromatic polycarbonate prepolymer is highly polymerized by use of a linking agent comprising a diol compound having the below-mentioned specific structure", paragraph [0111] describes that "a preferable production method of aromatic polycarbonate resin is a production method comprising a highly-polymerizing process wherein an aromatic polycarbonate prepolymer is reacted with a diol compound represented by the following general formula (IV) in the presence of a transesterification catalyst and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highly-polymerizing process is removed from the reaction system.", and regarding the diol compound, paragraph [0121] describes that "particularly preferable is a diol compound selected from the 2-butyl-2-ethylpropane-1,3-diol ...". group consisting of ... Further, paragraph [0063] describes that "the structural viscosity index (N-value) is an index of a branching degree of an aromatic polycarbonate resin. The aromatic polycarbonate resin preferably has a low N value, and a lower content of branched structure and high proportion of linear chain structure." The same [Table 1] describes that an aromatic polycarbonate resin composition "PC-1" has an N value of 1.19 and a content of a different kind of structure of 500 ppm, and the composition "PC-2" has an N value of 1.29 and a content of a different kind of structure of 2000 ppm. It can be seen from this description that a lower N value has a less amount of a different kind of structure, and at least produced by a method "which comprises a highly-polymerizing process wherein an aromatic polycarbonate prepolymer is reacted with a specific diol compound in the presence of a transesterification catalyst and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highlypolymerizing process is removed from the reaction system", and the content of different kinds of structure of "aromatic polycarbonate resin" having an N value smaller than 1.29 is less than 2000 ppm.

Further, a method of subjecting an "aromatic polycarbonate prepolymer 'PP-D'" of Cited Invention 1a and an aliphatic diol compound of 2-butyl-2-ethylpropane-1,3-diol (BEPD)" to a "transesterification reaction with a polymerization catalyst which had been used in the process of preparing the aromatic polycarbonate prepolymer being used directly" and "removing from the reaction system" phenol, cyclic carbonate (5-butyl-5-ethyl-1,3-dioxane-2-on), and unreacted BEPD distilled from the reaction system" corresponds to the above method "comprising a highly-polymerizing process wherein

an aromatic polycarbonate prepolymer is reacted with a specific diol compound in the presence of a transesterification catalyst and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highly-polymerizing process is removed from the reaction system", and Cited Invention 1a has an N value of 1.19, and thus the content of the above "unit having a different kind of structure" is less than 2000 ppm.

Consequently, it can be said that said aromatic polycarbonate resin of Cited Invention 1a "comprises a structural unit represented by the following general formula (III), and the content is less than 2000 ppm in an aromatic polycarbonate resin having a structural unit represented by said general formula (I) [Chemical Formula 5]



(where X has the same meaning as X in general formula (I))."

Therefore, Different Feature 2 is not a substantial difference.

Further, Cited Document 1 describes in [Table 7] that "aromatic polycarbonate resin" obtained in Example 20 has an N value of 1.21. Even if Cited Invention 1a should have an N value of 1.21, the concentration of the above "unit having a different kind of structure" would be less than 2000 ppm, and thus the above Different Feature 2 would not be a substantial difference.

#### (3) Regarding Different Feature 3

Cited Document 1 describes in paragraph [0121] that with regard to "a production method of highly polymerized aromatic polycarbonate resin" as shown in the above No. 4, 2(1), an aromatic polycarbonate resin "with drastically improved heat stability (heat resistance) at a high temperature" may be obtained. Paragraph [0302] describes that "an aromatic polycarbonate resin having advantages of a linked and highly polymerized polycarbonate using a linking agent which enables high molecular weight and high fluidity, and also having remarkably improved heat resistance can be obtained while keeping useful properties such as impact resistance that a common polycarbonate originally has". [Table 7] demonstrates Mw retention rate and YI value change before and after the retention test at a high temperature together with specific

data with regard to a resin obtained in Example 20. Thus an aromatic polycarbonate resin of Cited Invention 1a obtained by Example 20 has excellent heat stability (heat resistance) at a high temperature and improved fluidity, while maintaining impact Further, as described in paragraphs [0020] and [0025], the impact resistance. resistance generally deteriorates when trying to improve fluidity in the technical field of polycarbonate resin. In other words, it can be said that the fluidity and impact resistance are in a so-called trade-off relationship. On the other hand, paragraph [0004] describes that polycarbonate resin has excellent heat resistance and impact resistance. Further, as described in point 6(2) of Cited Document 5, which was cited as a document showing the common general knowledge as of the filing date of the present application, it was a matter of common general knowledge that polycarbonate resin has an advantage of impact resistance and many attempts had been made for making a composition of reinforcing materials in the case of requiring further higher mechanical strength and heat resistance. Consequently, it can be said that a person skilled in the art would recognize the problem to reinforce impact resistance that is reduced according to the improvement on fluidity of polycarbonate resin and to further improve heat resistance and mechanical strength in Cited Invention 1a.

Here, with regard to "a production method of highly polymerized aromatic polycarbonate resin" as shown in the above No. 4, 2(1), Cited Document 1 describes in paragraph [0364] that

"further, according to the present invention, various additives such as heat stabilizers, antioxidants, pigments, dye enhancing agents, fillers, ultraviolet absorbers, lubricants, mold release agents, crystal nucleating agents, plasticizers, fluidability-improving agents, and antistatic agents can be added to the polymer." It can be said from this description that Cited Document 1 suggests mixing a reinforcing agent and a filler with Cited Invention 1a obtained in Example 20.

Further, as described in points 6(1) to (4) of Cited Document 5, it is a matter of common general knowledge for a person skilled in the art to mix glass fiber, glass beads, glass flake, etc. at a mixing ratio of about 10 to 30% for the purpose of imparting a reinforcing effect to a polycarbonate resin and improving mechanical properties. Further, Cited Document 5 describes the use of glass fiber for imparting tensile characteristics, impact characteristics, and heat resistance in Table 5.4 (point 6(3)). As described in paragraph [0002] of Cited Document 2, it is a matter of common general knowledge for a person skilled in the art to mix an inorganic filler such as glass fiber that performs reinforcing effects on a resin for improved strength. The document describes in Claims 1 and 2 and paragraphs [0028] and [0029] glass fiber, glass beads,

glass flake, etc. as an inorganic filler that performs reinforcing effects, and the mixing amount is 1 to 300 weight parts of inorganic filler on a basis of 100 weight parts aromatic polycarbonate. It can be said from these descriptions that it was a well-known technical matter as of the filing of the present application to include an inorganic filler such as glass fiber, glass beads, glass flake, etc. into a polycarbonate resin and adjust the content of inorganic filler to 1 to 300 weight parts with respect to 100 weight parts of polycarbonate for the improved tensile properties, impact properties, heat resistance, etc. in the technical field of polycarbonate resin.

Consequently, it is mere employment of well-known technique to make a resin "composition" by adding 3 to 200 weight parts with respect to 100 weight parts of aromatic polycarbonate resin of inorganic fillers such as glass fiber, glass beads, glass flake, etc. to the aromatic polycarbonate resin of Cited Invention 1, as suggested in Cited Document 2, for the sake of improvement in impact characteristics, heat resistance, tensile characteristics, etc. And it was easily conceivable by a person skilled in the art on the basis of Cited Document 5 and the matters described in Cited Document 2.

#### (4) Effect of Invention 1

First, a consideration is given as to whether or not Invention 1 causes an advantageous effect compared to Cited Invention 1a. Comparing Invention 1 with Cited Invention 1a, as discussed in the above 1, 2(1) and (2), they are substantially different from each other in terms of whether or not to include a specific amount of "inorganic filler", which is "at least one kind selected from the group consisting of glass fiber, glass beads, and glass flake". Thus a consideration is given to the advantageous effect due to this difference.

Paragraph [0009] of the specification of the present application describes that "a polycarbonate resin composition having practically sufficient Charpy impact resistance and resistance to moist heat may be obtained by mixing a polycarbonate resin comprising a certain amount or less of a specific cyclic carbonate with various inorganic fillers, and has reached the Invention." Paragraph [0029] describes that "according to the Invention, an aromatic polycarbonate resin composition having excellent Charpy impact strength and resistance to moist heat may be provided." Thus the effect of Invention 1 is "the excellent Charpy impact strength and resistance to moist heat specification of the present application show data in the test of Charpy impact strength and humidity resistance of Examples 1 to 2 and Comparative Examples 1 to 2.

On the other hand, Cited Document 1 describes that polycarbonate resin has excellent heat resistance and impact characteristics, and Cited Documents 2 and 5 describe the use of inorganic filler such as glass fiber for imparting tensile characteristics, impact characteristics, and heat resistance. In view of these, one could have a certain expectation that the mixture of glass fiber, etc. with polycarbonate resin would improve tensile characteristics, impact characteristics, impact characteristics, heat resistance, etc.

Here, further consideration is given about the effects on the basis of Examples 1 to 2 and Comparative Examples 1 to 2 of the specification of the present application.

Examples 1 and 2 use PC-1 and Comparative Examples 1 to 2 use PC-2 as an aromatic polycarbonate resin. With reference to the description of paragraphs [0179] to [0215] of the specification of the present application and Table 1, PC-1 is an aromatic polycarbonate resin that has undergone a highly-polymerizing process wherein an aromatic polycarbonate prepolymer is reacted with an aliphatic diol in the presence of a transesterification catalyst and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highly-polymerizing process is removed from the reaction system, and the resin has a weight average molecular weight of 47200, Mw/Mn of 2.3, a terminal hydroxyl group concentration of 400 ppm, an amount of cyclic carbonate of 7 ppm, an amount of different kind of structure of 500 ppm, and an N value of 1.19, whereas PC-2 is an aromatic polycarbonate resin that has been simply subjected to high polymerization of an aromatic polycarbonate prepolymer having a weight average molecular weight of 48000, Mw/Mn of 2.6, a terminal hydroxyl group concentration of 1100 ppm, being free of cyclic carbonate, an amount of a different kind of structure of 2000 ppm, and an N value of 1.29. Specifically, it can be recognized that PC-2 is an aromatic polycarbonate resin that does not satisfy Invention 1 in terms of terminal hydroxyl group concentration, an amount of cyclic carbonate, and an amount of a different kind of structure.

Further, the assessment result of a resin composition prepared by use of these resins are set forth below: Regarding Charpy impact test that assesses mechanical strength (4 mm, notched) (kJ/m<sup>2</sup>), Example 1 including PC-1 is 15 and Example 2 is 10, Comparative Example 1 including PC-2 is 9, and Comparative Example 2 is 5. Similarly, regarding Charpy impact test that assesses mechanical strength (4 mm, notched) (kJ/m<sup>2</sup>), Example 1 including PC-1 is 62 and Example 2 is 50, Comparative Example 1 including PC-2 is 55, and Comparative Example 2 is 30. Regarding  $\Delta$ Q of moist heat test that assesses moist heat resistance, Example 1 including PC-1 is 3 and Example 2 is 10, Comparative Example 1 including PC-2 is 5, and Comparative Example 1 including PC-1 is 3 and Example 2 is 10, Comparative Example 1 including PC-2 is 5, and Comparative Example 1 including PC-1 is 3.

Further, comparing Example 1 with Comparative Example 1 and Example 2 with Comparative Example 2 having the same formulation other than polycarbonate resin, Example 1 including PC-1 has a larger value of Charpy impact test and a smaller  $\Delta Q$  value compared to Comparative Example 1 including PC-2, and further Example 2 including PC-1 has a larger value of Charpy impact test and a smaller  $\Delta Q$  value compared to Comparative Example 2 including PC-2. It can be said from these facts that it is demonstrated with specific data that Examples 1 and 2 including PC-1 cause improved Charpy impact strength and resistance to moist heat compared to Comparative Examples 1 and 2 including PC-2. A difference in formulation between Examples 1 and 2 and Comparative Examples 1 and 2 is only an aromatic polycarbonate resin. It is thus recognized that the effect is caused by a composition using a specific aromatic polycarbonate resin obtained by a specific production method. It cannot be recognized, however, that the effect is caused by mixing a specific inorganic filler.

Consequently, it cannot be said that the effect caused by mixing a specific amount of "inorganic filler", which is "at least one kind selected from the group consisting of glass fiber, glass beads, and glass flake" in Cited Invention 1a is particularly significant.

Subsequently, if a consideration is given to the effects of the whole aromatic polycarbonate resin composition, as described in Table 5.4 (point 6(3)) of Cited Document 5, it is a well-known technical matter for a person skilled in the art that mixing a glass fiber with a polycarbonate resin results in improved impact characteristics, and it is well known that the Charpy impact test is conducted for the measurement of mechanical strength of a resin composition in which an inorganic filler such as glass fiber is mixed with an aromatic polycarbonate resin (in this regard, see, if necessary, paragraphs [0091] and [0106], Tables 3 and 4 of Cited Document 3, paragraph [0158], Table 3 of Cited Document 4.) Therefore, it cannot be said that the mechanical strength of aromatic polycarbonate resin composition of Invention 1 in compliance with the Charpy impact test is particularly significant to the extent that cannot be expected by a person skilled in the art. Furthermore, as described in paragraph [0120] of Cited Document 1, it is a well-known technical matter for a person skilled in the art that an aromatic polycarbonate resin has excellent physical properties such as low equilibrium water absorption, heat resistance, and resistance to hydrolysis. It can be seen that physical properties such as low equilibrium water absorption, heat resistance, and resistance to hydrolysis are relevant to resistance under a high temperature and high humidity environment. As discussed in the above (3),

polycarbonate resin of Cited Invention 1a has excellent heat resistance. As described in Table 5.4 (point 6(3)) of Cited Document 5, it is a well-known technical matter for a person skilled in the art that mixing a glass fiber with a polycarbonate resin results in improved heat resistance. Thus it cannot be said that the effects of Invention 1 having excellent resistance to moist heat are particularly significant to the extent that cannot be expected by a person skilled in the art.

# (5) Summary

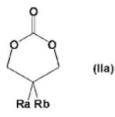
As described above, Invention 1 was easily conceivable by a person skilled in the art on the basis of Cited Invention 1a, well-known technique, and the matters described in Cited Documents 2 and 5, Cited Invention 1a, well-known technique, and the matters described in Cited Documents 2 to 5.

No. 6 Comparison between Invention 1 and Cited Invention 1b and judgment

1 Comparison

Invention 1 and Cited Invention 1b are compared.

"5-butyl-5-ethyl-1,3-dioxan-2-on" of Cited Invention 1b corresponds to "cyclic carbonate represented by the following general formula (IIa)" of Invention 1 "[Chemical Formula 3]



(where Ra represents an ethyl group, and Rb represents a butyl group)".

"Including 3000 ppm or less" of Cited Invention 1b overlaps "from 0.1 ppm or more to 3000 ppm or less" of Invention 1.

"Polycarbonate resin composition" of Cited Invention 1b corresponds to "aromatic polycarbonate resin composition" of Invention 1.

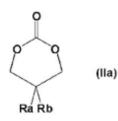
Consequently, these inventions have the following Corresponding Feature:

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# <Corresponding Feature>

"An aromatic polycarbonate resin composition comprising an aromatic polycarbonate resin and a cyclic carbonate represented by the following general formula (IIa), and the cyclic carbonate represented by the following general formula (IIa) is 0.1 ppm or more to 3000 ppm or less on an aromatic polycarbonate resin basis.

[Chemical Formula 3]



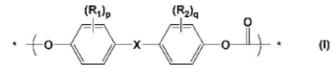
(where Ra represents and ethyl group, and Rb represents a butyl group)."

Further, there are the following Different Features 4 to 7 between them:

<Different Feature 4>

Regarding aromatic polycarbonate resin, Invention 1 specifies that "having a structural unit represented by the following general formula (I), and a weight average molecular weight of 40000 to 100000", and

"[Chemical Formula 1]



(where p and q represent 0. X represents a group selected from the group of the following (Ia))

[Chemical Formula 2]

(where R3 and R4 each represent a methyl group)", whereas Cited Invention 1b does not specify structural unit and weight average molecular weight.

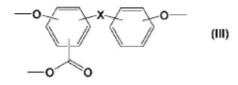
#### <Different Feature 5>

Invention 1 specifies that "aromatic polycarbonate resin has a terminal hydroxyl group concentration of 1000 ppm or less", whereas Cited Document 1 is silent about the terminal hydroxyl group concentration of aromatic polycarbonate resin of the Cited Invention 1b.

#### <Different Feature 6>

Invention 1 specifies that "said aromatic polycarbonate resin comprises a structural unit represented by the following general formula (III), and the content is lower than 2000 ppm in an aromatic polycarbonate resin having a structural unit represented by said general formula (I)"

"[Chemical Formula 5]



(where X has the same meaning as X in general formula (I)).", whereas Cited Invention 1b does not specify as such.

## <Different Feature 7>

Invention 1 specifies that "said inorganic filler is at least one kind selected from the group consisting of glass fiber, glass beads, and glass flake, and the content of said inorganic filler is 3 to 200 mass parts with respect to 100 mass parts of aromatic polycarbonate resin having a structural unit represented by the following general formula (I)", whereas Cited Invention 1b does not specify the inclusion of inorganic filler.

#### 2 Judgment

#### (1) Regarding Different Feature 4

Cited Document 1 describes in paragraph [0302] that "the aromatic polycarbonate to be used for the process for producing a highly polymerized aromatic polycarbonate resin" shown in the above No. 4, 2(1) is "a polycondensation polymer (aromatic polycarbonate prepolymer) having a structure represented by the above general formula (II) as a main repeating unit, similarly for the use in producing the

above polycarbonate copolymer." Paragraphs [0200] to [0204] describe general formula (II), and paragraphs [0205] to [0209] describe 2,2-bis(4-hydroxyphenyl)propane "as an aromatic dihydroxy compound that derives a structural unit represented by the above general formula (II)". Further, 2,2-bis(4-hydroxyphenyl)propane is also used in Example 20 of Cited Document 1.

Here, a structure shown in the general formula (II) derived from 2,2-bis(4-hydroxyphenyl)propane is a structure in which "p and q" in the formula are 0, and "X" is (where R3 and R4 represent methyl group).

Consequently, the use of a polycondensed polymer (aromatic polycarbonate prepolymer) having a main repeating unit represented by general formula (II) derived from 2,2-bis(4-hydroxyphenyl)propane as an "aromatic polycarbonate" was easily conceivable by a person skilled in the art in Cited Invention 1b, and a highly polymerized one has "a structure represented by general formula (II)". Thus the resultant "highly polymerized aromatic polycarbonate resin" comprises "a structural unit represented by the following general formula (I)" and

"[Chemical Formula 1]

$$+ \left( \mathbf{O} - \left( \begin{array}{c} (\mathbf{R}_{1})_{\mathbf{p}} & (\mathbf{R}_{2})_{\mathbf{q}} \\ -\mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} (\mathbf{R}_{2})_{\mathbf{q}} & \mathbf{O} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \\ -\mathbf{A} \end{array} \right) - \mathbf{A} - \left( \begin{array}{c} -\mathbf{A} \\ -\mathbf{A$$

(where p and q represent 0. X represents a group selected from the group of the following (Ia))

[Chemical Formula 2]

(where R3 and R4 each represent a methyl group)" of Invention 1.

Further, Cited Document 1 describes in paragraph [0367] that an aromatic polycarbonate resin obtained by "a method of producing a highly-polymerized aromatic polycarbonate resin" as shown in the above No. 4, 2(1) has a weight average molecular weight (Mw) of 30,000 to 100,000.

Consequently, "aromatic polycarbonate resin" of Cited Invention 1b has a weight average molecular weight (Mw) of 30,000 to 100,000, which overlaps "weight average molecular weight falls within a range of 40,000 to 100,000" of Invention 1.

## (3) Regarding Different Feature 5

Cited Document 1 describes in paragraph [0309] that an aromatic polycarbonate prepolymer using "a method of producing a highly-polymerized aromatic polycarbonate resin" as shown in the above No. 4, 2(1) has a terminal hydroxyl group concentration of "preferably 1500 ppm or less, further preferably 1000 ppm or less", and "if hydroxyl group terminal exceeds this range", "a polymer having sufficiently high molecular weight might fail to be obtained by the transesterification reaction with the aliphatic diol Consequently, "aromatic polycarbonate" of Cited Invention 1b is a compound". prepolymer using the above "a method of producing a highly-polymerized aromatic polycarbonate resin", and thus the terminal hydroxyl group concentration is 1000 ppm or less. Further, as discussed in the above No. 5, 2(1), it is obvious that "aromatic polycarbonate resin" obtained by "a method of producing aromatic polycarbonate resin" in which an aromatic polycarbonate prepolymer is highly polymerized by an aliphatic diol compound has a lower terminal hydroxyl group concentration compared to a terminal hydroxyl group concentration of the raw material of aromatic polycarbonate prepolymer. Therefore, "aromatic polycarbonate resin" of Cited Invention 1b has a terminal hydroxyl group concentration of 1000 ppm or less.

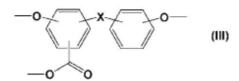
Therefore, the above Different Feature 5 is not a substantial difference.

Even if it were substantial, a person skilled in the art could have easily conceived of reducing an amount of terminal hydroxyl group of an aromatic polycarbonate prepolymer before the reaction and adjusting a "terminal hydroxyl group concentration" of "aromatic polycarbonate resin" of Cited Invention 1b to 1000 ppm or less for sufficiently highly polymerizing by a transesterification reaction with an aliphatic diol compound.

# (2) Regarding Different Feature 6

Similarly to the discussion of the above No. 5, 2(2), an aromatic polycarbonate resin of Cited Invention 1b is construed as including a unit having a different kind of structure exemplified in the paragraphs [0252] to [0253] of Cited Document 1, and the unit corresponds to "include a structural unit represented by the following general formula (III)" of Invention 1:

"[Chemical Formula 5]



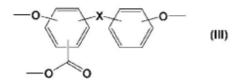
(where X has the same meaning as X in general formula (I))." Further, similarly to the discussion of the above No. 5, 2(2), it can be said that an aromatic polycarbonate resin of Cited Invention 1b has a lower content of the "unit having a different kind of structure".

Further, as discussed in the above No. 5, 2(2), it can be seen from the description of paragraphs [0054], [0063], [0111], [0121], and [Table 1] of the specification that a lower N value has a lower amount of a different kind of structure, and at least is produced by a method "which comprises a highly-polymerizing process wherein an aromatic polycarbonate prepolymer is reacted with a specific diol compound in the presence of a transesterification catalyst and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highly-polymerizing process is removed from the reaction system", if an N value is lower than 1.29, a content of different kinds of structure of the obtained "aromatic polycarbonate resin" is less than 2000 ppm.

Consequently, "a method of producing a highly-polymerized aromatic polycarbonate resin comprising a highly-polymerizing process of reacting an aromatic polycarbonate with 2-butyl-2-ethylpropane-1,3-diol the presence of in а transesterification catalyst for highly-polymerization and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highly-polymerizing process is removed from the reaction system" of Cited Invention 1b corresponds to the above method "comprising a highly-polymerizing process wherein an aromatic polycarbonate prepolymer is reacted with a specific diol compound in the presence of a transesterification catalyst and a cyclic carbonate-removing process wherein at least a part of cyclic carbonate produced as a by-product in the highlypolymerizing process is removed from the reaction system", Cited Invention 1b has an N value of 1.25 or less, and thus the content of the above "unit having a different kind of structure" is less than 2000 ppm.

Consequently, said aromatic polycarbonate resin of Cited Invention 1b "comprises a structural unit represented by the following general formula (III), and the content is less than 2000 ppm in an aromatic polycarbonate resin having a structural unit represented by said general formula (I)

[Chemical Formula 5]



(where X has the same meaning as X in general formula (I))."

Therefore, the above Different Feature 6 is not a substantial difference.

## (4) Regarding Different Feature 7

Similarly to the above No. 5, 2(3), it is simply an addition of well-known technique to include an inorganic filler such as glass fiber, glass beads, glass flake, etc. into an aromatic polycarbonate resin composition of Cited Invention 1b as a filler that was suggested for mixing and to adjust the content of inorganic filler to 3 to 200 weight parts on 100 weight parts of aromatic polycarbonate resin for improved impact characteristics, heat resistance, tensile characteristics, etc., and it was easily conceivable by a person skilled in the art on the basis of Cited Document 5 and the matters described in Cited Document 2.

# (5) The effects of Invention 1

The determination in the above No. 5, 2(4) shall apply similarly.

#### (6) Summary

Therefore, Invention 1 was easily conceivable by a person skilled in the art on the basis of Cited Invention 1b, well-known technique, and the matters described in Cited Documents 2 and 5, or on the basis of Cited Invention 1b, well-known technique, and the matters described in Cited Documents 2 to 5.

#### No.7 Appellant's allegation

1 The appellant's allegations

In the written argument dated March 27, 2019, the appellant alleges the following:

<Allegation 1> "An aromatic polycarbonate resin composition of Cited Document 2 is disclosed as having excellent anti-heat aging and retention stability (see paragraphs

[0037] to [0040]). It is completely silent about the effect of the Invention; i.e., the improved impact resistance and resistance to moist heat due to the addition of inorganic filler. Specifically, one could not be motivated from Cited Document 2 to improve impact resistance and resistance to moist heat of aromatic polycarbonate resin by an inorganic filler." It cannot thus be said that one could easily conceive of the invention according to an aromatic polycarbonate resin composition comprising a specific inorganic filler by considering the description of Cited Document 2 in Cited Inventions 1a and 1b.

<Allegation 2> "It can be seen from the comparison between Examples 1 and 2 and comparison between Comparative Examples 1 and 2 that the addition of additives such as flame retardant and mold release agent may result in a decrease in Charpy impact strength and resistance to moist heat. It can be seen that the degree of the decrease in a resin composition comprising an aromatic polycarbonate resin (PC-1) having a structural unit derived from bisphenol A including a specific cyclic carbonate (5-butyl-5-ethyl-1,3-dioxan-2-on) and an inorganic filler is suppressed compared to a resin composition comprising an aromatic polycarbonate resin (PC-2) having a structural unit derived from bisphenol A including a specific cyclic carbonate (5-butyl-1,3-dioxan-2-on) and an inorganic filler is suppressed compared to a resin composition comprising an aromatic polycarbonate resin (PC-2) having a structural unit derived from bisphenol A including a specific cyclic carbonate (5-butyl-1,3-dioxan-2-on) and an inorganic filler.

Such an effect of a resin composition of the present invention is a superior effect unexpected from the invention described in Cited Document 1 that does not refer to these additives or Cited Document 2 in which the improvement of impact resistance and resistance to moist heat is not at all supported."

#### 2 Examination on Appellant's allegations

#### (1) Allegation 1

Cited Document 2 does not explicitly describe the improved impact resistance and resistance to moist heat of aromatic polycarbonate resin by the mixture of inorganic filler; however, as is discussed in the above No. 5, 2(3) and No. 6, 2(4), it was a wellknown technical matter for a person skilled in the art as of the filing date of the present application to mix inorganic filler such as glass fiber, glass flake, and glass beads having a reinforcing effect on a resin for the improvement on mechanical characteristics of impact resistance and heat resistance in the technical field of polycarbonate resin. Therefore, there is a motivation for a person skilled in the art to mix the inorganic filler in Cited Invention 1a or 1b, and it is easily conceivable by a person skilled in the art. Further, as discussed in the above No. 5, 2(4), the effects of improved impact resistance and resistance to moist heat of Invention 1 cannot be said to be the effects caused by mixing inorganic filler with an aromatic polycarbonate resin.

Therefore, the above appellant's allegation 1 is not acceptable.

## (2) Allegation 2

The appellant alleges the effect of Invention 1 for the reason that a resin composition comprising an aromatic polycarbonate resin (PC-1) including a specific cyclic carbonate (5-butyl-5-ethyl-1,3-dioxan-2-on) of Invention 1 and an inorganic filler has suppressed the decrease in impact resistance and resistance to moist heat when additives such as flame retardant and mold releasing agent are added compared to a resin composition comprising an aromatic polycarbonate resin (PC-2) free of a specific cyclic carbonate and an inorganic filler from the comparison between Examples 1 and 2 and the comparison between Comparative Examples 1 and 2. This effect of "suppressing the decrease in impact resistance and resistance to moist heat" is an effect caused by a resin composition in which additives such as flame retardant and mold releasing agent are mixed in addition to a specific aromatic polycarbonate resin shown in the above Examples 1 and 2 and Comparative Examples 1 and 2 (PC-1) and a specific inorganic filler. It is not an effect caused by a resin composition in which only a specific aromatic polycarbonate resin (PC-1) and a specific inorganic filler are mixed. Consequently, it cannot be recognized that the effect of "suppressing the decrease in impact resistance and resistance to moist heat" is an effect caused by Invention 1.

Therefore, as discussed in the above No. 5, 2(4) and No. 6, 2(5), the effect of mixing inorganic filler cannot be said to be particularly significant in Cited Inventions 1a and 1b.

Further, as aforementioned, it cannot be said that the appellant's allegation discusses the effect of Invention 1. Just to be safe, even when a consideration is given to the appellant's allegation, it cannot be said that the effect is significant as set forth below.

Regarding the above allegation 2, the effect of suppressing the decrease in impact resistance and resistance to moist heat is shown by comparing a difference between Examples 1 and 2 and a difference between Comparative Examples 1 and 2. A difference in formulations between Examples 1 and 2 and Comparative Examples 1 and 2 is only an aromatic polycarbonate resin. It is thus recognized that the effect is caused by a composition using a specific aromatic polycarbonate resin.

Here, Cited Document 1 discloses in paragraph [0302] that "aromatic polycarbonate resin" of the Cited Inventions 1a and 1b is "the original characteristics of polycarbonate resin such as impact resistance are maintained". As discussed in the above No. 5, (4), Cited Document 1 also suggests that an aromatic polycarbonate resin has resistance under high temperature and high humidity environment (the paragraph [0120]). Thus "aromatic polycarbonate resin" of Cited Inventions 1a and 1b is a resin excellent in impact resistance and resistance to moist heat.

Further, as discussed in the above No. 5, (4), it is a well-known technical matter for a person skilled in the art that mixing a glass fiber with a polycarbonate resin results in improved impact characteristics and heat resistance. In a composition comprising a polycarbonate resin and a glass fiber, they are simply mixed together and it cannot be recognized that they interact with each other. Thus it cannot be recognized that the mixture of them would cause unexpected synergistic effects.

Consequently, the effect of suppressing the decrease in impact resistance and resistance to moist heat by use of a composition with a specific aromatic polycarbonate resin is an effect included in the fact that an aromatic polycarbonate resin itself of Cited Invention 1a or 1b has excellent impact resistance and resistance to moist heat. Thus it cannot be said that it is such a significant effect that cannot be expected from the description of Cited Document 1.

As described above, the above Appellant's allegation 2 is not acceptable.

## No. 8 Closing

As described above, Invention 1 was easily conceivable by a person skilled in the art who had ordinary knowledge in the technical field to which the invention pertained on the basis of Cited Invention 1a, well-known technique, and the matters described in Cited Documents 2 and 5, Cited Invention 1a, well-known technique, and the matters described in Cited Documents 2 to 5; Cited Invention 1b, well-known technique, and the matters described in Cited Documents 2 and 5; or Cited Invention 1b, well-known technique, and the matters described in Cited Documents 2 to 5. Thus this invention cannot be granted a patent under the provision of Article 29(2) of the Patent Act.

Therefore, without considering the inventions according to the remaining claims, the present application should be rejected.

Therefore, the appeal decision shall be made as described in the conclusion.

June 11, 2019

Chief administrative judge:KONNO MitsutomoAdministrative judge:SATO TakefumiAdministrative judge:TAKESADA Ayumi