

Appeal Decision

Appeal No. 2020-2773

Appellant ARLANXEO Singapore Pte. Ltd.

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The case of appeal against the examiner's decision of refusal of Japanese Patent Application No. 2016-565318, entitled "Diluent for the Production of Butyl Rubber" (International publication No. WO2015/164972 published on November 5, 2015, and National Publication of International Patent Application No. 2017-516891 published on June 22, 2017) 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 (hereinafter, referred to as "the Application") is a patent application (Japanese Patent Application No. 2016-565318) filed on April 30, 2013 as an international filing date (priority claim under the Paris Convention: received on April 30, 2014 by the foreign receiving office, European Patent Office (EP)), and the history of the further procedures is as follows:

As of March 20, 2019	Notification of reasons for refusal
June 25, 2019	Written opinion and Written amendment
As of October 23, 2019	Decision of refusal
As of February 28, 2020	Written request for appeal

No. 2 Regarding the Invention

Inventions according to Claims 1 to 24 of the Application are specified by the

matters recited in Claims 1 to 24 of the scope of claims in the written amendment dated June 25, 2019, and among them, Claim 1 is as follows (hereinafter, referred to as "the Invention," and the specification of the Application is referred to as "the Specification"):

"A process for the preparation of elastomers, the process comprising at least the steps of:
a) providing a reaction medium comprising an organic diluent and at least two monomers, wherein at least one monomer is an isobutene and at least one monomer is an isoprene;
and

b) polymerizing the monomers within the reaction medium in the presence of an initiator system to form an organic medium comprising the copolymer and the organic diluent, wherein

the organic diluent comprises

48 to 52 vol.% of methyl chloride, and

48 to 52 vol.% of 1,1,1,2-tetrafluoroethane, and

the methyl chloride and the 1,1,1,2-tetrafluoroethane add up to 90 to 100 vol.% of the total volume of the organic diluent."

No. 3 Outline of reasons for refusal stated in the examiner's decision

The outline of reasons for refusal stated in the examiner's decision includes a reason that the Invention could have been easily invented by a person ordinarily skilled in the art of the Invention before the priority date, based on the invention described in Cited Document 1 below, which had been distributed in Japan or a foreign country or available to the public through electric telecommunication lines before the priority date of the Application, and therefore should not be granted a patent under the provisions of Article 29(2) of the Patent Act.

1. National Publication of International Patent Application No. 2010-522262

No 4 Matters described in the Specification and Cited Documents

1 Matters described in the Specification

The Specification describes the following matters:

(S-a) [Technical Field]

[0001]

The Invention relates to a method to efficiently produce butyl rubber via a slurry process in a novel diluent.

[Background Art]

[0002]

Rubbers, in particular those containing repeating units derived from isoolefins, are industrially prepared by carbocationic polymerization processes. Of particular importance is butyl rubber, which is an elastomer of isobutylene and a smaller amount of a multiolefin such as isoprene.

...

[Problem to be solved by the invention]

[0012]

However, at the low temperatures applied, the rate of polymerization is typically low, making it desirable to provide a process allowing a higher throughput compared to prior art processes.

(S-b) "[0016]

(Monomers)

In this embodiment, in step a) a reaction medium comprising an organic diluent, and at least two monomers is provided whereby at least one monomer is an isoolefin and at least one monomer is a multiolefin.

...

[0018]

Examples of suitable isoolefins include ... isobutene, ... preferred isoolefin is isobutene.

As used herein the term multiolefin denotes compounds comprising more than one carbon-carbon-double-bond, either conjugated or non-conjugated.

[0019]

Examples of suitable multiolefins include isoprene,

Preferred multiolefins are isoprene and butadiene. Isoprene is particularly preferred.

...

[0022]

In another embodiment, the monomer mixture comprises 90 mass% to 95 mass% of at least one isoolefin monomer and 5 mass% to 10 mass% by weight of a multiolefin monomer based on the weight sum of all monomers employed. ... The isoolefin is preferably isobutene and the multiolefin is preferably isoprene."

(S-c) "[0026]

(Diluent)

In one embodiment the diluent comprises

- 42 to 58 vol.% of methyl chloride and
- 42 to 58 vol.% 1,1,1,2-tetrafluoroethane

whereby the two components add up to 90 to 100 vol.%, preferably 95 to 100 vol.%, more preferably 98 to 100 vol.% and yet even more preferably 99 to 100 vol.% of the total volume of the diluent.

[0027]

In another embodiment the diluent comprises

- 45 to 55 vol.% of methyl chloride and
- 45 to 55 vol.% 1,1,1,2-tetrafluoroethane

whereby the two components add up to 90 to 100 vol.%, preferably 95 to 100 vol.%, more preferably 98 to 100 vol.% and yet even more preferably 99 to 100 vol.% of the total volume of the organic diluent.

[0028]

In yet another embodiment the diluent comprises

- 48 to 52 vol.% of methyl chloride and
- 48 to 52 vol.% 1,1,1,2-tetrafluoroethane

whereby the two components add up to 96 to 100 vol.%, preferably 98 to 100 vol.%, more preferably 99 to 100 vol.% and yet even more preferably 99.5 to 100 vol.% of the total weight of diluent.

[0029]

The remainder to 100 vol.% if existent may comprise diluents other than methyl chloride and 1,1,1,2-tetrafluoroethane such as other fluorinated or chlorinated hydrocarbons or aliphatic hydrocarbons.

Examples of other chlorinated hydrocarbons include methylene chloride and ethyl chloride."

(S-d) "[0034]

(Initiator Systems)

In step b) the monomers within the reaction medium are polymerized in the presence of an initiator system to form a medium comprising the elastomer, the organic diluent, and optionally residual monomers.

[0035]

Initiator systems, in particular those for elastomers obtained by cationic polymerizations, typically comprise at least one Lewis acid and at least one initiator.

[0036]

(Lewis Acids)

...

[0037]

Further suitable Lewis acids include compounds represented by formula $MR(m)X(3-m)$, where M is a group 13 element, X is a halogen, R is a monovalent hydrocarbon radical selected from the group consisting of C1-C12 alkyl, ... ; and m is one or two.

[0038]

Examples for such compounds include ... ethyl aluminum dibromide, ... and any mixture thereof. Preferred are ... ethyl aluminum dichloride ($EtAlCl_2$ or EADC), ... and any mixture thereof.

...

[0061]

[Initiators]

Initiators useful in this invention are those initiators which are capable of being complexed with the chosen Lewis acid to yield a complex which reacts with the monomers to thereby form a propagating polymer chain.

[0062]

In a preferred embodiment the initiator comprises at least one compound selected from the groups consisting of water, hydrogen halides, carboxylic acids, ... alcohols, ..., tertiary alkyl esters, tertiary aralkyl esters,

[0063]

Preferred hydrogen halide initiators include hydrogen chloride, hydrogen bromide, and hydrogen iodide. A particularly preferred hydrogen halide is hydrogen chloride.

...

[0069]

Alcohols useful in this invention include methanol,

...

[0078]

In another particularly preferred initiator system, the Lewis acid is ethyl aluminum dichloride, preferably in a diluent.

...

[0081]

In another embodiment the amount of hydrogen chloride is in the range of 0.10 to 1 moles of hydrogen chloride per mole of aluminum of the alkyl aluminum halides,

preferably in the range of 0.2 to 0.5 moles of hydrogen chloride per mole of aluminum of the alkyl aluminum halides."

(S-e) "[0082]

(Polymerization Conditions)

In one embodiment, the organic diluent and the monomers employed are substantially free of water. As used herein, substantially free of water is defined as less than 50 ppm based upon total weight of the reaction medium, preferably less than 30 ppm, more preferably less than 20 ppm, even more preferably less than 10 ppm, yet even more preferably less than 5 ppm.

[0083]

A person skilled in the art is aware that the water content in the organic diluent and the monomers needs to be low to ensure that the initiator system is not affected by additional amounts of water which are not added by purpose, e.g. to serve as an initiator.

[0084]

Steps a) and/or b) may be carried out in continuous or batch processes, whereby continuous processes are preferred.

...

[0093]

The process may be carried out batchwise or continuously. Where a continuous reaction is performed the reaction time given above represents the average residence time.

...

[0095]

In another embodiment the reaction is quenched by the contact with the aqueous medium in step c). In one embodiment, the aqueous medium may have a pH value of 5 to 10, preferably 6 to 9, and more preferably 7 to 9 measured at 20°C and 1013 hPa.

...

[0099]

Typically in a step c) residual monomers of the monomer mixture and preferably additionally the diluent from the reaction medium are at least partially removed to obtain the elastomer, preferably by distillation."

(S-f) "[Examples]

[0107]

Experiment:

(General Procedure for Polymerization)

All polymerizations were performed in a dried, inert atmosphere. Batch reactions were run in 600 mL stainless steel reaction vessels, equipped with an overhead 4-blade stainless steel impeller driven by an external electrically driven stirrer. Reaction temperature was measured via a thermocouple. The reactor was cooled to the desired reaction temperature, listed in the Tables, by immersing the assembled reactor into a pentane cooling bath. The temperature of the stirred hydrocarbon bath was controlled to $\pm 2^\circ\text{C}$. All apparatus in liquid contact with the reaction medium were dried at 150°C for at least 6 hours and cooled in a vacuum-nitrogen atmosphere alternating chamber before use.

[0108]

High purity isobutene and methyl chloride were received from a manufacturing facility and used as is. The hydrofluorocarbon 1,1,1,2-tetrafluoroethane (>99.9% purity) (HFC-134a, Genetron@ 134a) was used as received. All were condensed and collected as liquids in the dry box.

Isoprene (Sigma-Aldrich, >99.5% purity) was dried over activated 3A molecular sieves for several days and distilled under nitrogen. A 1.0 M solution of ethylaluminum dichloride in hexanes (Sigma-Aldrich) was used as received.

A solution of $\text{HCl}/\text{CH}_2\text{Cl}_2$ was prepared by bubbling anhydrous HCl gas (Sigma-Aldrich, 99% purity) through a pre-dried Sure/SealTM bottle containing anhydrous CH_2Cl_2 (VWR). The $\text{HCl}/\text{CH}_2\text{Cl}_2$ solution was then titrated using 0.1 N NaOH (VWR) standard solution to determine its concentration.

[0109]

The slurry polymerizations were performed by charging isobutene, isoprene, and the diluent (specified in each of the examples) into a chilled 600 mL stainless steel reaction vessel at polymerization temperature and stirred at a predetermined stirring speed of 500 to 900 rpm.

[0110]

The initiator systems were prepared in methyl chloride. The initiator systems were prepared under the same temperature conditions as the reaction vessel by diluting the $\text{HCl}/\text{CH}_2\text{Cl}_2$ solution into an aliquot of methyl chloride and adding the 1.0 M solution of the ethylaluminum dichloride to a 1:4 molar ratio of HCl : EADC, followed by gentle swirling. The initiator/coinitiator solution was used immediately, and added to the polymerization using a chilled glass Pasteur pipette. The reaction was allowed to run for 5 minutes and stopped by the addition of 2 mL of a 1% sodium hydroxide in ethanol solution. Conversion is reported as weight percent of monomers converted to polymer during polymerization after isolation and vacuum drying of the polymer.

[0111]

(Raman Spectroscopy)

...

[0112]

(Characterization): ...

[0113]

The molecular weight of the polymers was determined by GPC (gel permeation chromatography) using a Waters 2690/5 Separations Module and a Waters 2414 Refractive Index Detector. Tetrahydrofuran was used as an eluent (0.8 mL/min, 35°C) with a series of three Agilent PL gel 10 μ m Mixes-B LS 300 \times 5.7 mm columns.

[0114]

(Examples 1 to 5)

A series of polymerizations were performed in pure methylchloride, pure 1,1,1,2-tetrafluoroethane, and various blend ratios of 1,1,1,2-tetrafluoroethane with methylchloride at -95°C. All polymerizations were performed consistently as described above. Polymerizations were run with 180 mL diluent, 20 mL isobutene, and 0.6 mL (2.3 mol.-%) isoprene. The initiator system solution was prepared in 80 mL methylchloride by adding 11 mL of a 0.18 M HCl/CH₂Cl₂ solution and 8 mL of a 1.0 M hexane solution of ethylaluminum dichloride (EADC). 5 ml of said initiator system solution was used for all polymerizations according to Examples 1 to 5.

[0115]

A Raman probe was placed directly into the reaction medium in order to monitor the progress of the reaction. The results are summarized in Table 1:

[0116]

[Table 1]

表 1:

実施例	希釈剤	体積 (%)	反応 デルタ T (°C)	転化 (質量%)	全不飽和 ¹⁾ (mol%)	Mw x10 ³	Mw/Mn
1*	CH ₃ Cl	100	7.8	78	1.49	468	4.7
2*	CH ₃ Cl/ HFC-134a	75/25	9.8	81	1.62	322	3.6
3	CH ₃ Cl/ HFC-134a	50/50	10.9	89	1.82	261	3.3
4*	CH ₃ Cl/ HFC-134a	25/75	9.6	88	2.03	241	3.6
5*	HFC-134a	100	8.8	72	1.87	291	4.9

*: 比較のため

表 1 :
 実施例
 希釈剤
 体積
 反応デルタ T
 転化 (質量%)
 全不飽和
 比較のため

Table 1:
 Example
 Diluent
 Volume
 Reaction Delta T
 Conversion (mass%)
 Total Unsaturation
 For comparison

[0117]

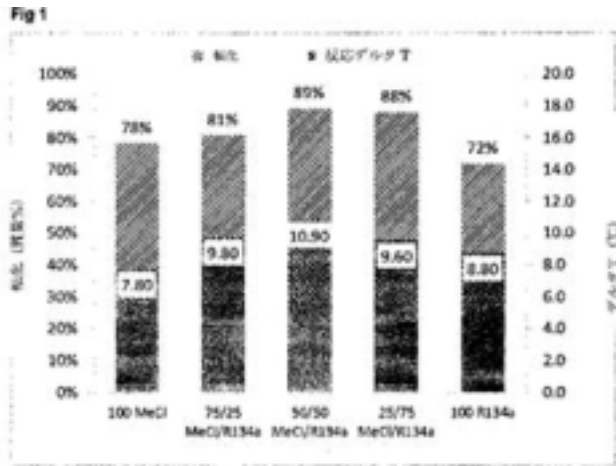
Conversion and Reaction delta T of Examples 1 to 5 are depicted in FIG. 1 and FIG. 2. It is apparent and surprising that for the 50/50 v/v ratio of methyl chloride and 1,1,1,2-tetrafluoroethane, the highest temperature rise and thus the highest rate of polymerization is observed during the reaction and the yield of butyl rubber produced is at a maximum at this diluent ratio. The observed temperature rise and conversion rates decrease as the ratio of either methyl chloride or HFC-134A is increased to 100% as illustrated in FIG. 1. This is an important advantage for a continuous slurry production process in order to maximize the throughput of the polymerization reactors.

[0118]

In addition, butyl rubber produced by slurry polymerization reactions performed in various ratios of methyl chloride and HFC-134A displayed a minimum in the polydispersity (PDI) for a 50/50 ratio of methyl chloride and 1,1,1,2-tetrafluoroethane.

As illustrated in FIG. 3, an increase in either methyl chloride or HFC-134A ratio results in products with higher polydispersity values. A narrow molecular weight distribution can lead to materials with improved processibility characteristics, which is advantageous for mixing operations to produce tire inner liner compounds."

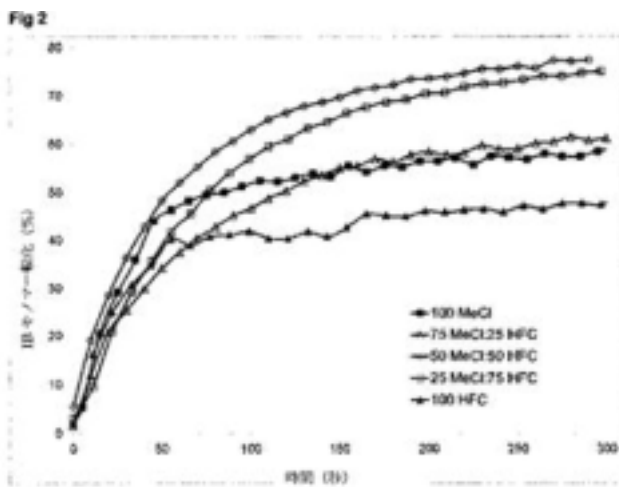
(S-g) "[FIG. 1]



転化
 反応デルタ T
 転化 (質量%)
 デルタ T

Conversion
 Reaction Delta T
 Conversion (mass%)
 Delta T

[FIG. 2]

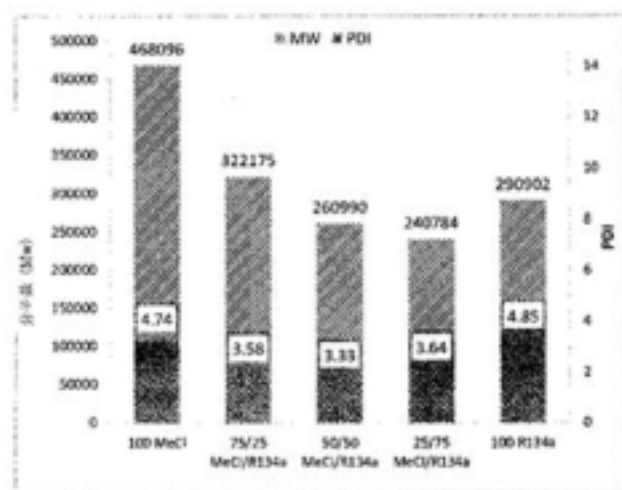


モノマー転化
 時間 (秒)

Monomer Conversion
 Time (s)

[FIG. 3]

Fig 3



分子量

Molecular Weight

"

2 Matters described in Cited Document

The following matters are described in National Publication of International Patent Application No. 2010-522262, which is a publication distributed on July 1, 2010, which is before the priority date of the Application (the same as the above "Cited Document 1" and is hereinafter also referred to as "Cited Document 1").

(C1-a) "[Scope of Claims]

[Claim 1]

A method to polymerize one or more monomer(s) to form isoolefin polymers and copolymers, comprising:

polymerizing the one or more monomer(s) in a polymerization medium comprising the one or more monomer(s), a diluent, and a catalyst system; wherein

the diluent comprises one or more halogenated hydrocarbon(s); wherein

the catalyst system comprises one or more Lewis acid(s) and a plurality of modifiers comprising one or more initiator(s) and one or more polymorphogenate(s); and

adjusting the concentration of said one or more polymorphogenate(s) in said polymerization medium so that said isoolefin polymers and copolymers have a controlled MWD greater than 2.0.

...

[Clam 18]

The method of any one of claims 1 to 17, wherein the diluent is selected from the group consisting of ethyl chloride, methyl chloride, methylene chloride, CHCl_3 , CCl_4 , n-butyl chloride, chlorobenzene, and mixtures thereof.

[Clam 19]

The method of any one of claims 1 to 18, wherein the diluent further comprises a hydrocarbon or a non-reactive olefin and/or an inert gas.

[Clam 20]

The method of any one of claims 1 to 19, wherein the diluent comprises one or more hydrofluorocarbon(s) (HFC(s)).

[Clam 21]

The method of claim 20, wherein the diluent comprises from 15 to 100 volume % HFC based upon the total volume of the diluent.

..."

(C1-b) "[Technical Field]

[0001]

The disclosure relates to new polymerization methods and polymerization systems using polymorphogenates in a catalyst system to control polydispersity or molecular weight distribution (MWD).

[Background Art]

[0002]

Isoolefin polymers are prepared in carbocationic polymerization processes. Of special importance is butyl rubber, which is a copolymer of isobutylene with a small amount of isoprene. Butyl rubber is made by low temperature cationic polymerization that generally requires that the isobutylene have a purity of >99.5 wt% and the isoprene have a purity of >98.0 wt% to prepare high molecular weight butyl rubber.

...

[Problem to be solved by the invention]

[0014]

This invention provides new polymerization systems and processes using a new class of catalyst system modifiers, referred to herein as polymorphogenates. A polymorphogenate modifies the catalyst system to promote or mimics the formation of different active catalyst complex species having different polymerization rates; i.e. different propagation, chain transfer, or termination reactions, as observed by different

polydispersities resulting from the presence of relatively different proportions of the polymorphogenate.

The polymorphogenate is selected from molecular oxygen and chalcogenates, for example, organic oxygenates and thio compounds corresponding to the organic oxygenate.

In the polymerization of isoolefin monomers with a Lewis acid and initiator in diluent, the polymorphogenate is used as a control mechanism to modulate the molecular weight characteristics of the polymers produced, including MWD."

(C1-c) "[0043]

<Monomers and Polymers>

Monomers which may be polymerized by this system include any hydrocarbon monomer that is polymerizable using this disclosure.

Preferred monomers include one or more of olefins, alpha-olefins, disubstituted olefins, isoolefins, conjugated dienes, non-conjugated dienes, styrenics, and/or substituted styrenics and vinyl ethers.

...

[0046]

Preferred polymers include homopolymers of any of the monomers disclosed herein.

Examples of homopolymers include Preferred polymers also include copolymers of 1) ...; and 2) isobutylene and isoprene."

(C1-d) "[0049]

<Lewis acid>

In a preferred embodiment the Lewis acid (also referred to as the co-initiator or catalyst) may be any Lewis acid based on metals from Group 4, 5, 13, 14, and 15 of the Periodic Table of the Elements, including boron, aluminum, gallium, indium, titanium, zirconium, tin, vanadium, arsenic, antimony, and bismuth.

A person skilled in the art will recognize that some elements are better suited in the practice of the disclosure. In one embodiment, the metals are aluminum, boron, and titanium, with aluminum being desirable.

...

[0058]

Particularly preferred Lewis acids may be any of those useful in cationic polymerization of isobutylene copolymers, including: ... ethylaluminum sesquichloride, ...

and ethylaluminum sesquichloride being preferred.

(C1-e) "[0060]

<Polymorphogenates>

Polymorphogenates in the present disclosure include any element or chemical compound that can modify or partially modify catalyst complexes in a carbocationic polymerization catalyst system comprising a Lewis acid and initiator to promote or mimic the formation of different active catalyst complex species having different polymerization rates.

...

[0061]

The polymorphogenate is preferably selected from molecular oxygen and chalcogenates, including organic oxygenates and thio compounds corresponding to the oxygenates.

...

[0062]

Oxygenates used in the present invention can include substituted or unsubstituted ethers, ketones, aldehydes, alcohols, esters, carboxylic acids, and the like. The oxygenate can have from 1 to 12 carbon atoms or more, preferably from 1 to 8 carbon atoms, more preferably from 2 to 5 carbon atoms.

...

[0068]

In an embodiment, the one or more organic oxygenate(s) is selected from dimethyl ether, diethyl ether, methylethyl ether, diisopropyl ether, methylpropyl ether, ethylpropyl ether, dipropyl ether, methylpropyl ether, ethylpropyl ether, isopropylpropyl ether, acetone, and methylethyl ketone.

[0069]

In an embodiment, the one or more organic oxygenate(s) comprises an alcohol selected from methanol, ethanol, propanol, isopropanol, butanol isomers, and isobutanol isomers.

[0070]

In another embodiment, the one or more organic oxygenate(s) is selected from methanol, dimethyl ether, acetone, diethyl ether, diisopropyl ether, and methyl ethyl ketone."

(C1-f) "[0073]

<Initiator>

Initiators useful in this disclosure are those initiators which are capable of being complexed in a suitable diluent with the chosen Lewis acid to yield a complex which rapidly reacts with the olefin to thereby form a propagating polymer chain.

...

[0077]

In a preferred embodiment the initiator comprises one or more of a hydrogen halide, a carboxylic acid, a carboxylic acid halide, a sulfonic acid, an alcohol, a phenol, a tertiary alkyl halide, a tertiary aralkyl halide, a tertiary alkyl ester, a tertiary aralkyl ester, a tertiary alkyl ether, a tertiary aralkyl ether, alkyl halide, aryl halide, alkylaryl halide, and arylalkylacid halide.

...

[0078]

Preferred hydrogen halide initiators include hydrogen chloride, hydrogen bromide, and hydrogen iodide. A particularly preferred hydrogen halide is hydrogen chloride.

...

[0087]

Catalyst system compositions useful in this disclosure typically comprise (1) a Lewis acid co-initiator and (2) at least two modifiers including at least one initiator (which may be a polymorphogenate) and at least one polymorphogenate (which may be an initiator)."

(C1-g) "[0097]

<Diluents>

Suitable diluents in the present disclosure include halogenated hydrocarbons, especially chlorinated and/or fluorinated hydrocarbons and the like.

...

[0105]

When HFC is employed in the diluent, the HFC is typically present at 1 to 100 volume percent based upon the total volume of the diluents, alternatively from a lower limit of 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, 97, 98, or 99 volume percent.

In a preferred embodiment the HFC is blended with one or more chlorinated hydrocarbons. In another preferred embodiment the HFC is selected from the group consisting of difluoromethane, trifluoromethane, 1,1-difluoroethane, 1,1,1-trifluoroethane, and 1,1,1,2-tetrafluoroethane and mixtures thereof."

(C1-h) "[0111]

<Polymerization Process>

FIG. 1 is a schematic flow diagram of a polymerization system or process according to

an embodiment of the disclosure.

Isobutylene in a line 10, comonomer(s) in a line 12, diluent 14a from a diluent storage 14, and any polymorphogenate in a line 16 are blended in a feed blend unit 18. The feed blend unit 18 is conventional except with respect to the polymorphogenate addition.

As is known in the art, it is typical to dry and fractionate the isobutylene and/or comonomer(s) if necessary to remove impurities in advance of the feed blend unit 18.

The catalyst preparation unit 20 generally includes conventional apparatus for mixing the initiator 22 and the Lewis acid 24, and diluting the catalyst complex with the desired amount of diluent 14b from the diluent storage 14. If desired, polymorphogenate may optionally be introduced into the catalyst preparation unit 20 via a line 26.

[0112]

Monomer is fed from the feed blend unit 18 via a line 28, optionally through a chiller 30, and then to a polymerization reactor 32. Catalyst complex is similarly fed from the catalyst preparation unit 20 via a line 34, optionally through a chiller 36, to the reactor 32. The invention may be practiced in continuous and batch processes. Further, the reactor 32 can be a plug flow reactor and/or stirred tank reactors. The chillers 30 and 36 are optional and can be used upstream or downstream from the feed blend unit 18 and/or the catalyst preparation unit 20.

...

[0115]

The polymer-containing effluent 56 from the reactor 32 can, if desired, be quenched by a quench medium such as steam and/or hot water introduced via a line 58. The polymer is then separated from the slurry in a polymer recovery unit 60, using the heat from the quench medium to vaporize diluent and unreacted monomers in a flash tank or stripper, for example.

The vapor stream 62 is then dried and fractionated in a diluent recovery unit 64 to obtain recycled diluent 66, recovered monomer 68, which may also be recycled, and one or more polymorphogenate or waste stream(s) 70.

...

It is also possible for all or part of the polymorphogenate to be produced with the recycled diluent 66 and recovered monomer 68, provided there is a purge or bleed mechanism to prevent excessive levels from accumulating in the process.

...

[0119]

This invention may also be practiced in batch reactors where the monomers, diluent, catalyst, and oxygenate are charged to the reactor 32 and then polymerization proceeds

to completion (such as by quenching) and the polymer is then recovered.

[0120]

The reacted monomers within the reactor 32 form part of a slurry. In one embodiment, the concentration of the solids in the slurry is equal to or greater than 10 vol%, and in other embodiments equal to or greater than 25 vol%, less than or equal to 75 vol%, or in a range from 1 to 70 vol%, 5 to 70 vol%, 10 to 70 vol%, 15 to 70 vol%, 20 to 70 vol%, 25 to 70 vol%, 30 to 70 vol%, or 40 to 70 vol%.

[0121]

Typically, a continuous flow stirred tank-type reactor 32 may be used. The reactor 32 is generally fitted with an efficient agitation means, such as a turbo-mixer or impeller(s), an external cooling jacket and/or internal cooling tubes and/or coils, or other means of removing the heat of polymerization to maintain the desired reaction temperature, inlet means (such as inlet pipes at the termini of the lines 28 and 34) for monomers, diluents, catalysts and polymorphogenate (combined or separately), temperature sensing means, and an effluent overflow or outflow pipe which withdraws polymer, diluent, and unreacted monomers among other components, to a holding drum or quench tank (not shown).

Preferably, the reactor 32 is purged of air and moisture. A person skilled in the art will recognize proper assembly and operation.

...

[0132]

The overall residence time in the reactor 32 can vary, depending upon, e.g., catalyst activity and concentration, monomer concentration, feed injection rate, production rate, reaction temperature, and desired molecular weight, and generally will be between about a few seconds and five hours, and typically between about 10 and 60 minutes. Variables influencing residence time include the monomer and diluent feed injection rates and the overall reactor volume."

(C1-i)

[0135]

In one embodiment, the polymerization of cationically polymerizable monomers (such as polymerization of isobutylene and isoprene to form butyl rubber) comprises several steps.

...

[0136]

In one embodiment, the resultant polymer from one embodiment of the disclosure is a polyisobutylene/isoprene polymer (butyl rubber) that has a MWD of about 2 to 5, and an

unsaturation of 0.5 to 2.5 moles per 100 mole of monomer."

(C1-j) "[Examples]

[0141]

Polymerizations were conducted in a laboratory-scale continuous reactor constructed of stainless steel and designed to permit the introduction of monomer and catalyst feeds and the continuous removal of the polymer product. Mixing was provided by a three bladed impeller mounted on a stainless steel shaft and driven by an external electric motor.

...

The reactor was cooled to the desired reaction temperature, listed in the examples, by immersing the assembled reactor into a pentane or isohexane bath in an inert atmosphere glove box. The temperature of the stirred hydrocarbon bath was controlled to $\pm 2^\circ\text{C}$. All apparatus in liquid contact with the reaction medium were dried at 120°C and cooled in a nitrogen atmosphere before use.

Isobutylene (Matheson or ExxonMobil) and methyl chloride (Air Gas) were dried by passing the gas through three stainless steel columns containing barium oxide and were condensed and collected as liquids in the glove box. Alternatively, methyl chloride was dried by the addition of triethylaluminum to the liquid at low temperature and subsequently distilling the methyl chloride from this solution under its own vapor pressure.

1,1,1,2-tetrafluoroethane (134a) (National Refrigerants) was dried by passing the gas through three stainless steel columns containing 3 angstroms molecular sieves and was condensed and collected as a liquid in the glove box.

Isoprene (Aldrich) was either distilled prior to use or used as received.

HCl solutions were prepared in either methyl chloride or 134a by dissolving gaseous HCl (Aldrich, 99% pure) into the condensed liquid at low temperature. The concentration of the HCl in these prepared solutions was determined by standard titration techniques. In the examples below, the diluent composition referred to as the "blend" is a 50/50 wt/wt mixture of 134a and methyl chloride.

[0142]

The slurry copolymerizations were performed by first preparing the monomer and catalyst feeds. The monomer feed was prepared in a glass or metal reservoir and comprised isobutylene, isoprene, the selected diluent, and other oxygenates that are optional components.

The monomer feed comprised 20 wt% isobutylene for Polymerization Runs 1 and 3 to 11.

...

Isoprene was charged to the monomer feed at 2.8 mol% with respect to isobutylene.

When methanol was intentionally added, it was added to the monomer feed so that the concentration in the monomer feed was as shown in Table 1.

Terms used in the Table and FIG. 2 are defined as follows: MeOH = methanol; DME = dimethyl ether; Et₂O = diethyl ether; DIPE = diisopropyl ether; MEK = methyl ethyl ketone; 134a = 1,1,1,2-tetrafluoroethane; MeCl = methyl chloride; Blend = 50/50 wt/wt mixture of 134a and MeCl; IB = isobutylene.

[0143]

For Polymerization Runs 4 to 11, the methyl chloride used for the polymerization was dried by passing the gas through a desiccant bed as described above. The desiccant was not completely effective at removing traces of methanol, dimethyl ether, and acetone that are adventitiously present in the methyl chloride.

The concentration of these components in the methyl chloride was determined by gas chromatography (GC). The determined values for these organic oxygenates were used to calculate the total concentration of these components in the final feed blend. For these examples, the total organic oxygenate concentration is listed in Table 1.

The methyl chloride used for Polymerization Runs 12 to 15 was collected by distillation from liquid methyl chloride treated with triethylaluminum. This procedure removes adventitious methanol, dimethyl ether, and acetone. As shown in Table 1, therefore, the methyl chloride used for these polymerization runs does not contain any of these organic oxygenates unless they are purposely added. This aspect also is reflected in the Table. HFC 134a does not contain these organic oxygenates.

[0144]

A catalyst feed was also prepared for each copolymerization in a separate reservoir. The catalyst feed was prepared by adding a predetermined amount of the stock HCl solution, a hydrocarbon solution of ethylaluminum dichloride (EADC), and optionally an organic oxygenate. The EADC/HCl molar ratio was 3.0.

...

[0145]

An initial monomer feed was also prepared and charged into the reactor for the purpose of starting the polymerization run. The concentration of isobutylene monomer in the initial monomer was 10 wt% isobutylene. Isoprene was also charged to this initial monomer feed at 2.8 mol% relative to isobutylene. All feeds were chilled to the same temperature as the reactor using the chilled hydrocarbon bath of the glove box.

Polymerizations in methyl chloride were conducted at a reactor temperature of -95°C ± 3°C. Polymerizations in 134a or the blend were conducted at a reactor temperature of -

75°C ± 3°C.

Near the beginning of the polymerization, the temperature of the bath was lowered a few degrees to provide an initial difference in temperature between the bath and the reactor contents. The copolymerizations were begun by introducing the catalyst. The catalyst flow rate was controlled to provide for a constant differential temperature between the reactor and the bath and achieve the target polymerization temperature for the run. Optionally, the temperature of the bath was lowered to achieve the polymerization temperature target.

Addition of monomer feed from the reservoir was introduced into the reactor approximately 10 minutes after the reaction commenced as evidenced by the formation of precipitated polymer particles (slurry particles). The run was continued until the monomer feed in the reservoir was exhausted or until the desired amount of monomer feed was consumed. Generally, the average monomer conversion in these polymerization runs was better than 75% and at times as high as 95%.

[0146]

A sample of the polymer was collected from the reactor shortly before the end of the polymerization run for molecular weight determination. This sample represents the product produced under a set of polymerization conditions at steady-state. This polymer sample was analyzed by Size Exclusion Chromatography to determine the number-average and weight-average molecular weights as well as the MWD of the sample. The MWD of each example is calculated by dividing the weight-average molecular weight by the number-average molecular weight.

...

[0149]

The data for each example in the Table represent the average of at least 3 polymerizations conducted under the conditions listed in the Table; namely, diluent and organic oxygenate concentrations.

The data for average monomer feed rate and average MWD were calculated as simple average for the total number of runs conducted under that conditions.

...

[Table 1]

表 1

重合 実験	希釈剤	モノマー 供給速度 (ml/分)	MeOH (wppm)	DME (wppm)	アセトン (wppm)	Et ₂ O (wppm)	DIPE (wppm)	MEK (wppm)	平均 M _w /M _n
1	134a	11.3	0	0	0	0	0	0	2.6
2	134a	18.9	0	0	0	0	0	0	2.4
3	134a	11.6	5.0	0	0	0	0	0	3.1
4	ブレンド	11.4	0.2	1.2	0.2	0	0	0	3.0
5	ブレンド	11.9	5.7	1.2	0.2	0	0	0	3.3
6	ブレンド	10.3	5.7	1.2	0.2	3.1	0	0	3.7
7	ブレンド	9.9	5.7	1.2	0.2	8.1	0	0	3.7
8	ブレンド	10.3	5.7	1.2	3.2	0	0	0	4.1
9	ブレンド	9.2	5.7	1.2	7.0	0	0	0	4.9
10	ブレンド	10.3	5.7	1.2	0.2	0	10.3	0	4.6
11	ブレンド	9.7	5.7	1.2	0.2	0	0	9.8	4.6
12	ブレンド	19.1	5.0	6.0	1.0	0	0	0	3.4
13	MeCl	15.0	0	0	0	0	0	0	2.5
14	MeCl	16.1	5.0	0	0	0	0	0	2.6
15	MeCl	16.8	5.0	6.0	1.0	0	0	0	3.4

重合実験

Polymerization run

希釈剤

Diluent

モノマー供給速度 (ml/分)

Monomer feed rate (ml/min)

アセトン

Acetone

平均

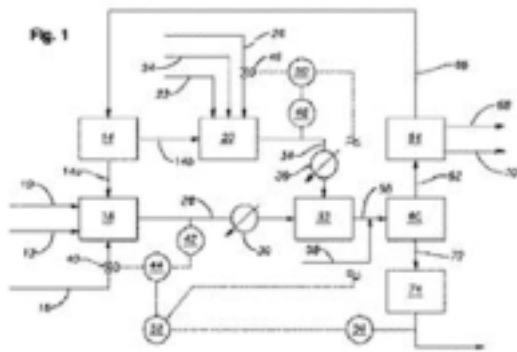
Average

ブレンド

Blend

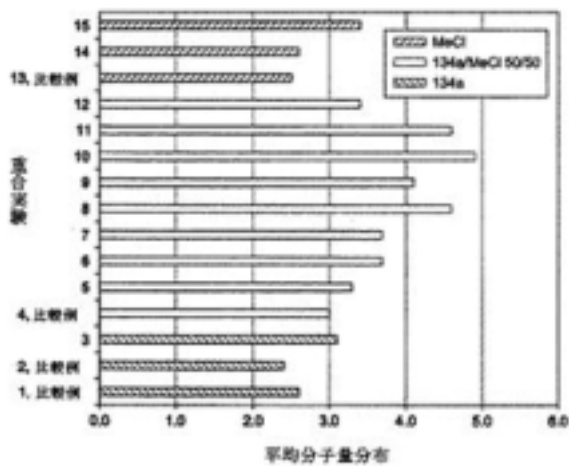
"

(C1-k) "[FIG. 1]



"

(C1-1) "[FIG. 2]



比較例

Comparative Example

重合実験

Polymerization run

平均分子量分布

Average Molecular Weight Distribution

"

No. 5 Judgment by the body

1 Invention described in Cited Document 1

Paragraph [0145] of Cited Document 1, which is included in the above described matter (C1-j), describes the follows:

"An initial monomer feed was also prepared and charged into the reactor for the purpose of starting the polymerization run. The concentration of isobutylene monomer in the

initial monomer was 10 wt% isobutylene. Isoprene was also charged to this initial monomer feed at 2.8 mol% relative to isobutylene. All feeds were chilled to the same temperature as the reactor using the chilled hydrocarbon bath of the glove box. ... Near the beginning of the polymerization, the temperature of the bath was lowered a few degrees to provide an initial difference in temperature between the bath and the reactor contents. The copolymerization reactions were begun by introducing the catalyst. ... Addition of monomer feed from the reservoir was introduced into the reactor approximately 10 minutes after the reaction commenced as evidenced by the formation of precipitated polymer particles (slurry particles). The run was continued until the monomer feed in the reservoir was exhausted or until the desired amount of monomer feed was consumed."

What should be noted in the descriptions in paragraph [0149] and "Polymerization run 5" in [Table 1] is that the "blend" is used as the "diluent" and the contents of "MeOH," "DME," and "acetone" are "5.7 wppm," "1.2 wppm," and "0.2 wppm," respectively. As is evident from the description in paragraph [0142], "MeOH = methanol; DME = dimethyl ether; ... Blend = 50/50 wt/wt mixture of 134a and MeCl," the "blend" can be a "50/50 wt/wt mixture of 134a and MeCl," "MeOH" and "DME" are "methanol" and "dimethyl ether," respectively. In addition, as is evident from the description in "1e" (see paragraph [0070] in particular), "MeOH," "DME," and "acetone" can be "organic oxygenates."

The "monomer feed" is described in paragraph [0142] in (C1-j) such that "the monomer feed was prepared in a glass or metal reservoir and comprised isobutylene, isoprene, the selected diluent, and other oxygenates that are optional components" "the monomer feed comprised 20 wt% isobutylene for Polymerization Runs 1 and 3 to 11;" and "isoprene was charged to the monomer feed at 2.8 mol% with respect to isobutylene." The "diluent" is described in paragraph [0141] of (C1-j) such that "the diluent composition referred to as the 'blend' is a 50/50 wt/wt mixture of 134a and methyl chloride;" including "1,1,1,2-tetrafluoroethane (134a)."

The "catalysts" can be "the HCl solution, or a hydrocarbon solution of ethylaluminum dichloride (EADC)" as is evident from the description in paragraph [0144] in (C1-j), "the catalyst feed was prepared by adding a predetermined amount of the stock HCl solution, a hydrocarbon solution of ethylaluminum dichloride (EADC)"

As is evident from the descriptions of paragraph [0002] in (C1-b) and (C1-i), "isobutylene" and "isoprene," which are the "monomer feeds," can be polymerized to produce a "butyl rubber."

Based on the descriptions above, it can be said that Cited Document 1 describes the following invention:

" A method for producing a butyl rubber, comprising:

preparing an initial monomer feed and charging it into a reactor for starting polymerization, wherein

the concentration of isobutylene monomer in the initial monomer is 10 wt% isobutylene;

charging isoprene to the initial monomer feed at 2.8 mol% relative to isobutylene;

chilling all feeds to the same temperature as the reactor using the chilled bath of the glove box;

lowering the temperature of the hydrocarbon bath a few degrees before the beginning of the polymerization to provide an initial difference in temperature between the bath and the reactor contents;

introducing a catalyst into the reactor to initiate a copolymerization reaction;

introducing addition of monomer feed from a reservoir into the reactor approximately 10 minutes after the reaction commenced as evidenced by the formation of precipitated polymer particles (slurry particles), wherein the monomer feed comprises 20 wt% isobutylene;

charging isoprene to the monomer feed at 2.8 mol% with respect to isobutylene;

and

continuing the polymerization run until the monomer feed in the reservoir is exhausted or until the desired amount of monomer feed is consumed,

wherein

the monomer feed comprises isobutylene, isoprene, a diluent, and other oxygenates that are optional components,

the diluent is a 50/50 wt/wt mixture of 1,1,1,2-tetrafluoroethane and methyl chloride,

the other oxygenates are

5.7 wppm of methanol, 1.2 wppm of dimethyl ether, and 0.2 wppm of acetone, and

the catalyst includes a HCl solution and a hydrocarbon solution of ethylaluminum dichloride (EADC)."

(hereinafter referred to as "the Cited Invention").

2 Comparison / Judgment for Invention and Cited Invention

(1) Comparison

The Invention and the Cited Invention are compared.

Since "isobutylene" in the Cited Invention and "isobutene" in the Invention are the

same compound, "isobutylene" and "isoprene" in the Cited Invention are "two monomers" "wherein at least one monomer is an isobutene and at least one monomer is a isoprene" in the Invention.

Furthermore, the "butyl rubber" obtained by polymerizing "isobutylene" and "isoprene" in the Cited Invention is an "elastomer" in light of the following description in paragraph [0002] of the Specification (S-a): "Rubbers, in particular those containing repeating units derived from isoolefins, are industrially prepared by carbocationic polymerization processes. an elastomer of isobutylene and a smaller amount of a multiolefin such as isoprene."

The "diluent," which is "a 50/50 wt/wt mixture of 1,1,1,2-tetrafluoroethane and methyl chloride," in the Cited Invention can correspond to "an organic diluent" comprising "methyl chloride" and "1,1,1,2-tetrafluoroethane" in the Invention.

The "50/50 wt/wt mixture of 1,1,1,2-tetrafluoroethane and methyl chloride," as a "diluent," in the Cited Invention contains no other component. Thus, it can be said "the methyl chloride and the 1,1,1,2-tetrafluoroethane add up to 90 to 100 vol.% of the total volume of the organic diluent."

Then, in the Cited Invention, the steps of "preparing an initial monomer feed and charging it into a reactor for starting polymerization, wherein the concentration of isobutylene monomer in the initial monomer is 10 wt% isobutylene; charging isoprene to the initial monomer feed at 2.8 mol% relative to isobutylene;" "wherein the monomer feed is comprised of isobutylene, isoprene, a diluent, and ... ; the diluent is a 50/50 wt/wt mixture of 1,1,1,2-tetrafluoroethane and methyl chloride" can correspond to,

in the Invention, "a) providing a reaction medium comprising an organic diluent and at least two monomers, wherein at least one monomer is an isobutene and at least one monomer is an isoprene;" and "the organic diluent comprises

- methyl chloride and
- 1,1,1,2-tetrafluoroethane, and

the methyl chloride and the 1,1,1,2-tetrafluoroethane add up to 90 to 100 vol.% of the total volume of the organic diluent."

The "initiator system" in the Invention is described in paragraph [0035] of the Specification (S-d) such that "Initiator systems in particular for elastomers obtained by cationic polymerizations typically comprise at least one Lewis acid and at least one

initiator." Thus, the "initiator system" can "comprises one Lewis acid and at least one initiator."

In the Specification ("S-d"), paragraphs [0037] to [0038] describe the use of "ethyl aluminum dichloride (EtAlCl₂ or EADC)" for the "lewis acid" and paragraphs [0062] to [0063] describe the use of "hydrogen chloride" for the "initiator", and paragraph [0114] describes that "the initiator system solution was prepared in 80 mL methylchloride by adding 11 mL of a 0.18 M HCl/CH₂Cl₂ solution and 8 mL of a 1.0 M hexane solution of ethylaluminum dichloride (EADC)."

The "catalyst" in the Cited Invention includes "an HCl solution" and "a hydrocarbon solution of ethylaluminum dichloride (EADC)." As can be seen from (C1-f) and the above described matters in the Specification, "HCl" (hydrogen chloride) in the "HCl solution" is "an initiator." As can be seen from the (C1-d) and the above described matters in the Specification, "ethylaluminum dichloride (EADC)" is "lewis acid." Thus, the "catalysts" in the Cited Invention correspond to the "initiator systems" in the Invention.

Paragraph [0115] in (C1-h) of Cited Document 1 includes the following description: "The polymer-containing effluent 56 from the reactor 32 can if desired be quenched by a quench medium such as steam and/or hot water introduced via a line 58. The polymer is then separated from the slurry in a polymer recovery unit 60, using the heat from the quench medium to vaporize diluent and unreacted monomers in a flash tank or stripper, for example." Thus, the "effluent 56" after the polymerization reaction can include the "polymer," "diluent," and "unreacted monomers." It can therefore be said that in the Cited Invention, "continuing the polymerization run until the monomer feed in the reservoir is exhausted or until the desired amount of monomer feed is consumed," "an effluent," which is "an organic fluid" containing "a polymer" and "a diluent," is produced.

Then, the steps of: "introducing a catalyst into the reactor to initiate a copolymerization reaction;

introducing addition of monomer feed from a reservoir into the reactor approximately 10 minutes after the reaction commenced as evidenced by the formation of precipitated polymer particles (slurry particles), wherein

the monomer feed comprises 20 wt% isobutylene;

charging isoprene to the monomer feed at 2.8 mol% with respect to isobutylene; and

continuing the polymerization run until the monomer feed in the reservoir is exhausted or until the desired amount of monomer feed is consumed" in the Cited Invention correspond to the step of "b) polymerizing the monomers within the reaction medium in the presence

of an initiator system to form an organic medium comprising the copolymer and the organic diluent" in the Invention.

Thus, the Invention and the Cited Invention are identical in that "A process for the preparation of elastomers, the process comprising at least the steps of: a) providing a reaction medium comprising an organic diluent and at least two monomers, wherein at least one monomer is an isobutene and at least one monomer is an isoprene; and b) polymerizing the monomers within the reaction medium in the presence of an initiator system to form an organic medium comprising the copolymer and the organic diluent, wherein the organic diluent comprises

- methyl chloride and
- 1,1,1,2-tetrafluoroethane, and

the methyl chloride and the 1,1,1,2-tetrafluoroethane add up to 90 to 100 vol.% of the total volume of the organic diluent."

However, the Invention and the Cited Invention are different from each other in the following features:

Different Feature 1: In the Invention, the percentages of "methyl chloride" and "1,1,1,2-tetrafluoroethane" in the "organic diluent" are "48 to 62 vol.%" and "48 to 62 vol.%" respectively. In the Cited Invention, in contrast, the ratio of "1,1,1,2-tetrafluoroethane" and "methyl chloride" is "50/50 wt/wt."

Different Feature 2: In the Cited Invention, the "reaction medium" contains "5.7 wppm of methanol, 1.2 wppm of dimethyl ether, and 0.2 wppm of acetone." In the Invention, in contrast, such a matter is not specified.

(2) Judgment

The above Different Features 1 and 2 will be respectively examined.

(A) Regarding Different Feature 1

The "diluent" used in the Cited Invention has a ratio of "1,1,1,2-tetrafluoroethane" to "methyl chloride" of "50/50 wt/wt", or contains 50% by weight of "1,1,1,2-tetrafluoroethane." In other words, a diluent in which the ratio of "1,1,1,2-tetrafluoroethane" to "methyl chloride" is 1: 1 in terms of weight is used. Paragraph

[0105] of Cited Document 1 ("C1-g") describes the following matters:

"When hydrofluorocarbon (HFC) is employed in the diluent, the HFC is typically present at 1 to 100 volume percent based upon the total volume of the diluents, alternatively from a lower limit of 5, ... , 45, 50, 55, ... , or 99 volume percent." as well as "In a preferred embodiment the HFC is blended with one or more chlorinated hydrocarbons. In another preferred embodiment the HFC is selected from the group consisting of difluoromethane, trifluoromethane, 1,1-difluoroethane, 1,1,1-trifluoroethane, and 1,1,1,2-tetrafluoroethane and mixtures thereof"; and, the "HFC" should be "50% by volume," or "50% by volume," relative to the total volume of the diluent,. in other words, the "HFC" is used in a concentration of "50% by volume" and the "hydrocarbon chloride" is used in concentration of "50% by volume."

In general, the volume of a fluid changes depending on the temperature. Thus, supplying the two fluids "1,1,1,2-tetrafluoroethane" and "methyl chloride" to the "respirator" or "reactor" can be preferably performed depending on a volume ratio rather than a weight ratio that changes with temperature, as the flow rate can be controlled more easily. In the Cited Invention, therefore, those skilled in the art could easily conceive of supplying "1,1,1,2-tetrafluoroethane" in a concentration of "50% by volume" and "methyl chloride" in a concentration of "50% by volume" in the Cited Invention.

(B) Regarding Different Feature 2

In the Invention, the "reaction medium" is "a reaction medium comprising an organic diluent and at least two monomers" and may further comprise other components. Thus, the Invention in interpretation by the description includes an aspect comprising "5.7 wppm of methanol, 1.2 wppm of dimethyl ether, and 0.2 wppm of acetone" in the "monomer feed" in the Cited Invention. Therefore, it cannot be a different feature.

Further, paragraphs [0061] to [0062] and [0069] in (S-d) of the Specification describe that, though as the "initiator", not as the "monomer feed," "alcohol" such as "methanol" or "ether" can be used as an "initiator." Thus, the Invention can include an aspect in which the polymerization reaction is carried out in a system containing "alcohol" and "ether."

In the Cited Invention, furthermore, "methanol," "dimethyl ether," and "acetone," which are "oxygenates," are included in the total of only 7.1 wppm (weight ppm) in the "monomer feed," or $7.1 \times 10^{-4}\%$. The "methanol", "dimethyl ether", and "acetone" in the Cited Invention can be impurity components contained in a very small amount in "methyl chloride" in light of the following descriptions:

"... methyl chloride (Air Gas) were dried by passing the gas through three stainless steel columns containing barium oxide and were condensed and collected as liquids in the glove box" in paragraph [0141] of "C1-j"; and

"For Polymerization Runs 4 to 11, the methyl chloride used for the polymerization was dried by passing the gas through a desiccant bed as described above. The desiccant was not completely effective at removing traces of methanol, dimethyl ether, and acetone that are adventitiously present in the methyl chloride. ... For these examples, the total organic oxygenate concentration is listed in Table 1." in paragraph [0143] of "C1-j."

On the other hand, paragraph [0108] in (S-f) of the Specification describes " High purity isobutene and methyl chloride were received from a manufacturing facility and used as is." In the examples in the Specification, therefore, "methanol," "dimethyl ether," and "acetone" can be highly likely to be contained as trace amounts of impurities.

Even if the above matters are taken into consideration, Different Feature 2 cannot be a substantial different feature.

(C) Regarding Effects of the Invention

As for "a process for the preparation of elastomers," the Invention intends to "provide a process allowing a higher throughput compared to prior art processes." (see, (S-a) of the Specification). The Specification describes that "It is apparent and surprising that for the 50/50 v/v ratio of methyl chloride and 1,1,1,2-tetrafluoroethane the highest temperature rise and thus the highest rate of polymerization is observed during the reaction and the yield of butyl rubber produced is at a maximum at this diluent ratio." (paragraph [0117]) and "... butyl rubber produced by slurry polymerization reactions performed in various ratios of methyl chloride and HFC-134A displayed a minimum in the polydispersity (PDI) for a 50/50 ratio of methyl chloride and 1,1,1,2-tetrafluoroethane." (paragraph [0118]). Also, [Table 1] in paragraph [0116] describes that the conversion rate is "89 mass%" and "Mw/Mn" is "3.3" in "Example 3" where the "diluent" is of "the 50/50 v/v ratio of methyl chloride and 1,1,1,2-tetrafluoroethane."

However, paragraph [0145] in (C1-f) of Cited Document 1 describes that "Generally, the average monomer conversion in these polymerization runs was better than 75% and at times as high as 95%." This means that the Cited Invention could achieve a high monomer conversion rate. Also, a high monomer conversion rate suggests that a polymerization rate is high. Furthermore, Polymerization run 5 in [Table 1] of " C1-f" describes that the "average Mw/Mn" is "3.3," which is about the same as that of the above Example 3 in the Specification.

Therefore, the effects of the Invention would be within the range predicted by those skilled in the art from the descriptions in Cited Document 1, and thus cannot be evaluated as particularly remarkable as compared with the Cited Invention.

(3) Summary

Therefore, the Invention could have been easily invented by a person skilled in the art on the basis of the invention disclosed in the Cited Invention.

3 The Appellant's allegation

(A) Appellant's allegation in the written request for appeal

In the written request for appeal dated February 28, 2020, the Appellant makes the allegations as described below.

Note that the Appellant makes the same allegations in the written opinion dated June 25, 2019.

Allegation 1: "In the Invention, the organic diluent comprises 48 to 52 vol.% Methyl chloride and 48 to 52 vol.% 1,1,1,2-tetrafluoroethane. Note that the density of methyl chloride is 1.003 g/ml and the density of liquid 1,1,1,2-tetrafluoroethane is 1.206 g/ml. The 50/50 wt/wt blend described in Cited Document 1 corresponds to a blend of 54.6 vol.% 1,1,1,2-tetrafluoroethane and 45.4 vol.% methyl chloride. This is outside the scope of claim 1 of the Application. At least in this respect, the invention recited in each claim of the Application is different from the invention described in Cited Document 1. Furthermore, Cited Document 1 describes that the amount of hydrofluorocarbon contained in the diluent is 15 to 100 vol.% (claim 21, etc. in Cited Document 1). This range of the volume described is very wide and allows a diluent to be included in various blend ratios. In contrast, the Invention defines that the blend contains 48 to 52 vol.% methyl chloride and 48 to 52 vol.% 1,1,1,2-tetrafluoroethane, each of which is limited to a very narrow range where the effects of the Invention have been confirmed. The disclosure of a very wide range of the volume, such as that in Cited Document 1, does not effectively disclose the Invention. It is therefore clear that the range in the Invention is different from the embodiment of Cited Document 1."

Allegation 2: "A person skilled in the art who has come into contact with Cited Document 1 would have difficulty predicting the effects of the Invention obtained by using an organic diluent containing 48 to 52 vol.% methyl chloride and 48 to 52 vol.% 1,1,1,2-

tetrafluoroethane. For example, highly increased temperature, high polymerization rate, and narrow molecular weight distribution demonstrated in the examples of the Application are the advantageous effects found in the Invention. ... Among the ratios tested this time, the conversion rate was the highest and the delta T was also large (FIG. 1). The monomer conversion was also high (FIG. 2). These results can be a very important advantage for maximizing the throughput of the polymerization reactor. In addition, as shown in FIG. 3, the molecular weight distribution is also the narrowest and can result in improved processing properties, thereby being advantageous for mixing operations for producing tire inner liner compounds (paragraph [0118])."

(B) Examination on Allegation 1

Regarding the above Allegation 1, assuming that the "diluent" in the Cited Invention, which is the "50/50 wt/wt mixture of 1,1,1,2-tetrafluoroethane and methyl chloride," the "diluent" is "a blend of 54.6 vol.% 1,1,1,2-tetrafluoroethane and 45.4 vol.% methyl chloride" as alleged by the Appellant and different from one in the Invention, which is "the diluent comprises 48 to 52 vol.% of methyl chloride and 48 to 52 vol.% of 1,1,1,2-tetrafluoroethane."

However, as examined in the above 2 (2) (A), the Cited Invention describes the use of "1,1,1,2-tetrafluoroethane" and "methyl chloride" in the "diluent" at a ratio of "50/50 wt/wt," or the use thereof at a ratio of 1: 1. Cited Document 1 also describes that "HFC is typically present at 1 to 100 volume percent based upon the total volume of the diluents, alternatively from a lower limit of 5, ..., 45, 50, 55, ..., or 99 volume percent" (paragraph [0105] in (C1-g)), including that the volume of "HFC" is "50 volume percent," or "50 vol.%" Furthermore, when supplying two fluids, "1,1,1,2-tetrafluoroethane" and "methyl chloride," to the "reservoir" or "reactor," the flow rate can be easily controlled in the supply with a determined volume ratio rather than the supply with a weight ratio that changes with temperature. In the Cited Invention, therefore, a person skilled in the art could easily conceive of the supply in which the ratio of "1,1,1,2-tetrafluoroethane" and "methyl chloride" is not 1 : 1 in terms of weight ratio but "50 vol.%" which is 1 : 1 in terms of volume ratio.

Therefore, the Appellant's Allegation 1 cannot be adopted.

(C) Examination on Allegation 2

Regarding the above Allegation 2, as examined in the above 2 (2) (C), Cited Document 1 describes that "Generally, the average monomer conversion in these

polymerization runs was better than 75% and at times as high as 95%" (paragraph [0145] in (C1-f)). Even in the Cited Invention, a high monomer conversion rate can be achieved, suggesting a high polymerization rate. Furthermore, Cited Document 1 also describes that the "average M/Mn" was "3.3" in the polymerization run 5 of [Table 1] in (C1-f), which is similar to that of Example 3 of the present specification. Therefore, the effects of the Invention cannot be evaluated as particularly remarkable.

In addition, the results of the examples of the Specification indicate that there is no significant difference in "polymerization conversion rate" in FIG. 1 whether "MeCl/R134a" is "50/50," which results in "89%," or "25/75," which results in "88%." These tendencies indicate that the difference is smaller when the "MeCl/R134a" of the Cited Invention is "45.4/54.5".

The "monomer conversion rate" in [FIG. 2] of the Specification indicates that there is no significant difference between "50/50" and "25/75" for "MeCl/R134a." These tendencies indicate that the difference is smaller when the "MeCl/R134a" of the Cited Invention is "45.4/54.5".

The value of "Mw/Mn" when "MeCl/R134a" in FIG. 3 of the Specification is "50/50" is "3.3" as in the "average Mw/Mn" in the polymerization run 5 of Cited Document 1, which is the Cited Invention.

Therefore, even if the examples of the Specification are examined, the effects of the Invention cannot be evaluated to be more remarkable than those of the Cited Invention.

As described above, therefore the Appellant's Allegation 2 cannot be adopted.

No. 6 Closing

As described above, the Invention could have been easily invented by a person who had ordinary skill in the art of the Invention before the priority date, based on the invention described in Cited Document 1, and accordingly, the patent should not be granted under the provisions of Article 29 (2) of the Patent Act.

Accordingly, the present application should be rejected without considering the inventions according to the other claims.

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

June 21, 2021

Chief administrative judge: SATO, Takefumi
Administrative judge: SUGIE, Wataru
Administrative judge: KATO, Tomoya