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FORM 1

SPRUSON & FERGUSON

COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952

APPLICATION FOR A STANDARD PATENT

Hercules Incorporated, Incorporated in Delaware, of 1313 N. Market Street, Wilmington, Delaware, 19894, UNITED STATES OF AMERICA, hereby apply for the grant of a standard patent for an invention entitled:

Rate Moderated Two Component Metathesis Catalyst System
which is described in the accompanying complete specification.

Details of basic application(s):-

Basic Applic. No: Country:

053429

US

Application Date:

18 May 1987

The address for service is:-

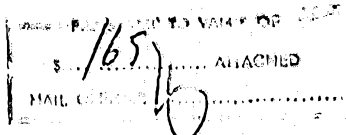
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DATED this SIXTEENTH day of MAY 1988

Hercules Incorporated

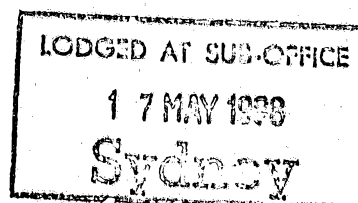
By:

H. J. Anderson



Registered Patent Attorney

TO: THE COMMISSIONER OF PATENTS
OUR REF: 57469
S&F CODE: 55410



5845/2

APPLICATION ACCEPTED AND AMENDMENTS

ALLOWED 24.5.90



DECLARATION IN SUPPORT OF A
CONVENTION APPLICATION FOR A PATENTAUSTRALIA
CONVENTION
STANDARD
& PETTY PATENT
DECLARATIONIn support of the Convention Application made for a
patent for an invention entitled:

Title of Invention Rate Moderated Two Component Metathesis Catalyst System

Full name(s) and
address(es) of
Declarant(s)I/We Michael Bernard Keehan, Assistant General Counsel,
Law Department of Hercules
of 2026 Harwyn Road
North Graylyn Crest
Wilmington, Delaware 19810
United States of America

do solemnly and sincerely declare as follows:-

Full name(s) of
Applicant(s)1. ~~I am/We are the applicant(s) for the patent.~~

(or, in the case of an application by a body corporate)

1. I am/~~We are~~ authorised by HERCULES INCORPORATEDthe applicant(s) for the patent to make this declaration on
its/~~their~~ behalf.2. The basic application(s) as defined by Section 141 of the
Act was/~~were~~ made

Basic Country(ies)

in U.S.A.

Priority Date(s)

on 18 May 1987

Basic Applicant(s)

by Lawrence Lee Nelson

Full name(s) and
address(es) of
inventor(s)3. ~~I am/We are the actual inventor(s) of the invention referred
to in the basic application(s)~~

(or where a person other than the inventor is the applicant)

3. Lawrence Lee Nelson

of 5532 Doral Drive, Fairway Falls
Wilmington, DE 19808
United States of America

(respectively)

is/~~are~~ the actual inventor(s) of the invention and the facts upon
which the applicant(s) is/~~are~~ entitled to make the application are
as follows:Set out how Applicant(s)
derive title from actual
inventor(s) e.g. The
Applicant(s) is/~~are~~ the
assignee(s) of the
invention from the
inventor(s)By Assignment dated 13 May 1987 from the actual
inventor to the said applicant4. The basic application(s) referred to in paragraph 2 of this
Declaration was/~~were~~ the first application(s) made in a Convention
country in respect of the invention(s) the subject of the application.

Wilmington,

Declared at Delaware, this 6 day of April 1988.
U.S.A.

(12) PATENT ABRIDGMENT (11) Document No. AU-B-16316/88
(19) AUSTRALIAN PATENT OFFICE (10) Acceptance No. 603669

(54) Title
RATE MODERATED TWO COMPONENT METATHESIS CATALYST SYSTEM

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(56) Prior Art Documents
US 4727125
US 4400340
US 3691095

(57) Claim

1. A process for preparing a polymer by polymerizing a metathesis-polymerizable polycyclic cycloolefin, in which the cycloolefin monomer is metathesis-polymerized in the presence of a metathesis-polymerization catalyst and a bidentate activator complex, said complex being prepared from an alkyl aluminium catalyst activator and a reaction rate moderator characterized in that the reaction rate moderator is a bidentate Lewis base having a first functional group that is chemically reactive with said catalyst activator and a second functional group containing an atom having unpaired electrons.

2. A process for preparing molded objects by polymerizing a metathesis-polymerizable polycyclic cycloolefin, in which the cycloolefin monomer, a metathesis-polymerization catalyst and a bidentate activator complex, said complex being prepared from an alkyl aluminium catalyst activator and a reaction rate moderator are charged into a mold in which polymerization takes place, characterized in that the reaction rate moderator is a bidentate Lewis base having a first functional group that is chemically reactive with said catalyst activator and a second functional group containing an atom having unpaired electrons.

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10. A polymerization composition comprising a metathesis polymerizable polycyclic cycloolefin, a metathesis polymerization catalyst and a bidentate activator complex, said complex being prepared from an alkyl aluminium catalyst activator and a reaction rate moderator, characterized in that the reaction rate moderator is a bidentate Lewis base having a first functional group that is chemically reactive with said catalyst activator and a second functional group containing an atom having unpaired electrons.

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S & F Ref: 57469

FORM 10

COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952

COMPLETE SPECIFICATION

This document contains the amendments made under Section 49 and is correct for printing.

(ORIGINAL)

FOR OFFICE USE:

Class Int Class

This document contains the

Complete Specification Lodged:

Accepted:

Published:

Priority:

Related Art:

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Sydney, New South Wales, 2000, Australia

Complete Specification for the invention entitled:

Rate Moderated Two Component Metathesis Catalyst System

The following statement is a full description of this invention, including the best method of performing it known to me/us

- 1 -

Rate Moderated Two Component
Metathesis Catalyst System

Abstract of Disclosure

Disclosed is a method of reducing the activation rate of an alkyl aluminum activator in the metathesis polymerization of polycyclic cycloolefins. The activation rate is reduced by the use of a bidentate Lewis base as the reaction rate moderator.

This invention relates to a process for increasing the gelation time in the polymerization of strained ring polycyclic cycloolefins under the influence of an aluminum alkyl-activated-metathesis catalyst system.

5 Such polymerization of strained ring polycyclic cycloolefins, as disclosed in U.S. Patent Nos. 4,400,340 and 4,520,181, has the disadvantage that polymerization takes place extremely rapidly. Even when the reaction injection molding (RIM) process of the said patents is used, in which
10 one stream including a tungsten or molybdenum halide catalyst and a second stream including an alkyl aluminum activator are combined in a mixing head and immediately injected into a mold, it is not unusual for the monomer to polymerize to a solid, immobile condition before the mixed streams can be
15 transferred to the mold.

To overcome that difficulty, the activator stream in the known process includes a reaction rate moderator to delay the catalyst activation until the reaction mass is totally within the mold. The total time from mixing until polymerization is
20 substantially complete is still just a matter of seconds.

Because of the extremely rapid rate of reaction of strained ring polycyclic cycloolefins, even in the presence of the rate-moderator, the short gelation times limit the application of the RIM process to relatively small items and
25 molds with a minimum of sharp corners or sections that tend to trap pockets of air, and in any case the polymerization of strained ring polycyclic cycloolefins under the influence of

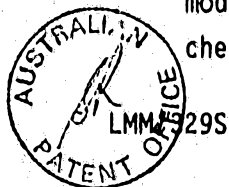
a metathesis catalyst system as disclosed in the said patents cannot be readily employed in other thermoset molding techniques that require relatively long mold filling times, such as pour, rotational and resin transfer (RTM) molding.

5 A process for increasing the gelation time to as much as ten minutes at room temperature is disclosed in U.S. Patent No. 4,458,037, by the use of a dialkyl aluminium iodide activator moderated by an ether. When heated to 80°C, that mixture polymerizes in about 15 seconds, but it is also unsatisfactory in procedures where filling of the mold takes place
10 relatively slowly, since the mold temperature must be held low enough during the filling operation to keep the exothermic reaction mixture in a fluid state until the mold is entirely filled and the mold temperature can be allowed to rise to the reaction temperature. To achieve commercially practical production rates, the differential between mold filling
15 temperature and polymerization reaction temperature must be smaller than is possible using the process of U.S. Patent No. 4,458,037.

There is therefore a need for a process for preparing molded objects from metathesis-polymerizable polycyclic cycloolefins that meets that requirement for a smaller temperature differential by significantly
20 increasing the gelation time at normal molding temperatures.

In accordance with a first embodiment of the present invention there is provided a process for preparing a polymer by polymerizing a metathesis-polymerizable polycyclic cycloolefin, in which the cycloolefin monomer is metathesis-polymerized in the presence of a metathesis-polymerization
25 catalyst and a bidentate activator complex, said complex being prepared from an alkyl aluminium catalyst activator and a reaction rate moderator characterized in that the reaction rate moderator is a bidentate Lewis base having a first functional group that is chemically reactive with said catalyst activator and a second functional group containing an atom having
30 unpaired electrons.

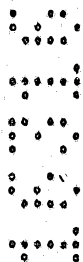
According to a second embodiment of the present invention there is provided a process for preparing molded objects by polymerizing a metathesis-polymerizable polycyclic cycloolefin, in which the cycloolefin monomer, a metathesis-polymerization catalyst and a bidentate activator
35 complex, said complex being prepared from an alkyl aluminium catalyst activator and a reaction rate moderator are charged into a mold in which polymerization takes place, characterized in that the reaction rate moderator is a bidentate Lewis base having a first functional group that is chemically reactive with said catalyst activator and a second functional



group containing an atom having unpaired electrons.

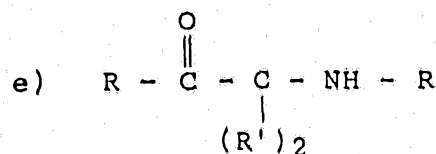
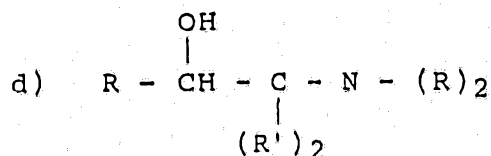
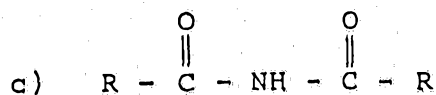
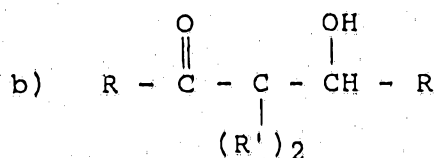
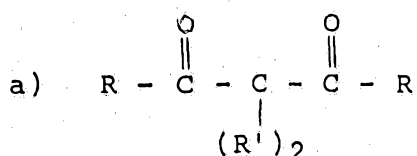
According to a third embodiment of this invention there is provided a polymerization composition comprising a metathesis polymerizable polycyclic cycloolefin, a metathesis polymerization catalyst and a bidentate activator complex, said complex being prepared from an alkyl aluminium catalyst activator and a reaction rate moderator, characterized in that the reaction rate moderator is a bidentate Lewis base having a first functional group that is chemically reactive with said catalyst activator and a second functional group containing an atom having unpaired electrons.

5



A bidentate Lewis base is an organic compound having one functional group that is chemically reactive with aluminum and a second functional group containing an atom having unpaired electrons whereby the Lewis base can form an adduct with aluminum. The preferred bidentate Lewis bases are those having the two functional groups in 1,3 relationship.

Typical 1,3-bidentate Lewis bases include materials of the general formula:

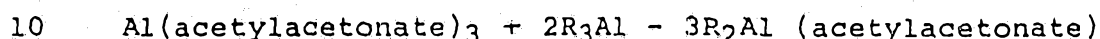


Where R is a 1 to 5 carbon alkyl radical or a phenyl or benzyl radical, R' is a 1 to 5 carbon alkyl radical or hydrogen, and in instances where a plurality of R or R' groups are present in a structure, they may be the same or different.

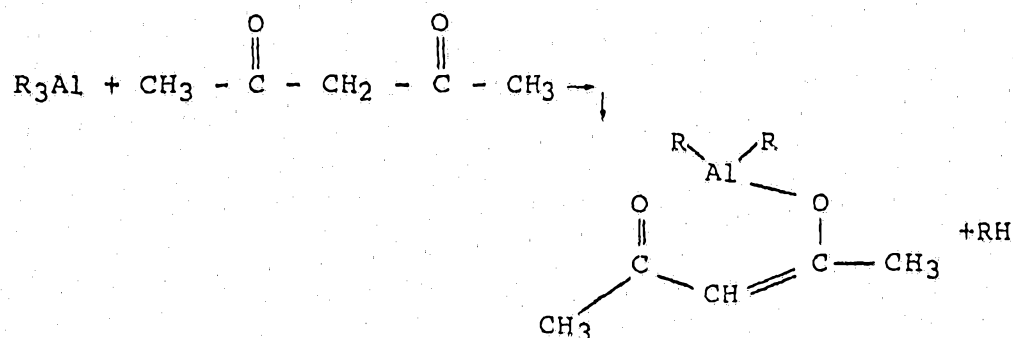
Examples of compounds conforming to the above structures are 4-hydroxypentanone-2, 3-methyl-2,4-pentanedione, acetyl-

acetone, dibenzoyl methane, N,N-dimethylisopropanolamine, N,N-dimethyl acetone amine, diacetoneamine, diacetonalcohol, and diacetamide. Other useful bidentate Lewis bases are tetrahydrofurfuryl alcohol and 8-hydroxyquinoline wherein the two reactive groups are present in a ring structure.

The bidentate activator complexes can be prepared by disproportionation of the corresponding aluminum trichelate with an aluminum alkyl or aluminum alkyl halide according to the reaction:



The activator complex can also be prepared by reacting the bidentate Lewis base with an aluminum trialkyl or a dialkyl aluminum halide according to the reaction:



15 The other bidentate Lewis base species are believed to complex aluminum in a similar manner.

The bidentate Lewis base compounds can be employed in conjunction with conventional metathesis catalysts to polymerize any metathesis polymerizable polycyclic olefin.

20 Most strained-ring nonconjugated polycyclic cycloolefins are metathesis polymerizable. These include, for example, dicyclopentadiene, methyl dicyclopentadiene, dicyclopentadiene oligomers, e.g., tricyclopentadiene, norbornene, norbornadiene, alkylidene norbornenes, dimethanooctahydro-

naphthalene, dimethanohexahydronaphthalene and substituted derivatives of these compounds. The preferred cyclic olefin monomer is dicyclopentadiene or a mixture of dicyclopentadiene with other strained ring polycyclic cycloolefins in ratios of 1 to 99 mole % of either monomer, preferably about 75 to 99 mole % dicyclopentadiene.

The catalyst component of the two-part-metathesis catalyst system can be either a molybdenum or a tungsten halide or such a halide having two valences satisfied by oxygen rather than halogen. The preferred catalyst component is a tungsten halide, and preferably a mixture or complex of tungsten hexachloride (WCl_6) and tungsten oxytetrachloride ($WOCl_4$) in a molar ratio of $WOCl_4$ to WCl_6 of about 1:9 to 2:1. This mixture or complex is prepared by known processes, for instance, as disclosed in Klosiewicz, U.S. 4,568,660 by contacting essentially pure WCl_6 with a controlled amount of an oxygen donor. Useful oxygen donors include, e.g., a hydrated salt, water, a wet molecular sieve, or an alkyl alcohol. The most preferred oxygen donor is t-butanol.

The tungsten or molybdenum compound is not normally soluble in the monomer, but can be solubilized by complexing it with a phenolic compound. The compound is first suspended in a small amount of an inert diluent such as benzene, toluene, xylene or chlorinated benzene to form a 0.1 to 1 mole per liter slurry. The phenolic compound is added to the slurry in a molar ratio of about 1:1 to 1:3 catalyst compound to phenolic compound and a stream of dry inert gas is passed through the agitated solution to remove hydrogen chloride gas. Preferred phenolic compounds include phenol, alkyl phenols, halogenated phenols or phenolic salts such as lithium or sodium phenoxide. The most preferred phenolic compounds are t-butyl phenol, t-octyl phenol and nonyl phenol.

To prevent premature polymerization of the catalyst component/monomer solution, which would occur within a matter of hours, about 1 to 5 moles of a Lewis base or a chelating agent are added per mole of catalyst compound. Preferred
5 chelants include acetylacetones, dibenzoyl methane and alkyl acetoacetates, where the alkyl group contains from 1 to 10 carbon atoms. Preferred Lewis bases are nitriles and ethers such as benzonitrile and tetrahydrofuran. The improvement in
10 stability and shelf-life of the catalyst component/monomer solution is obtained whether the complexing agent is added before or after the phenolic compound. When this complexed catalyst component is added to purified cycloolefin, for example dicyclopentadiene, it forms a solution that is stable and has a shelf-life of several months in the absence of an
15 activator.

The activator part of the metathesis catalyst system is an alkyl aluminum or an alkyl tin compound, which according to the invention is moderated with the bidentate Lewis base. The alkyl aluminum compounds, either trialkyl aluminum or
20 dialkyl aluminum halide, are preferred. Particularly preferred is a dialkyl aluminum halide with an alkyl moiety containing 1 to 12 carbon atoms and iodide as the halide. The activator, which is readily soluble in the monomer, is prepared by mixing the alkyl aluminum compound or mixture of
25 alkyl aluminum compounds with the bidentate Lewis base or mixture of bidentate Lewis bases at a 1:1 to 1:5 molar ratio. While either order of addition, i.e., Lewis base to alkyl aluminum compound or alkyl aluminum compound to Lewis base, can be used, it is preferred to add the Lewis base to the
30 alkyl aluminum compound with agitation. The reaction is highly exothermic, and it is desirable to control the rate of Lewis base addition to the alkyl aluminum compound so as to maintain the temperature at less than approximately 50°C to prevent decomposition of the aluminum Lewis base complex. In

the case of solid bidentate Lewis bases, the base can be added as a solid or dissolved in a suitable nonreactive solvent such as toluene. Alternatively, the activator can be prepared by dissolving or suspending the bidentate Lewis base in the monomer and adding the alkyl aluminum component to the solution.

When the two parts of the catalyst system are combined, the resulting ratio of the monomer (for example dicyclopentadiene) to the catalyst compound will preferably be from about 500:1 to about 15,000:1 on a molar basis, more preferably 2,000:1 and the catalyst compound to alkyl aluminum ratio will be preferably be from about 1:2 to about 1:5.

A reaction mixture moderated by a bidentate Lewis base according to this invention remains fluid for a relatively long time at room temperature. As long a time as 1 to 4 hours can be required for gel formation at room temperature. Thus, the catalyst components need not be mixed and immediately injected into a mold. While the RIM technique can be employed, processing is not limited to the RIM technique. Moreover, the RIM technique can be used with a premixed reactive solution (i.e., a monomer containing both catalyst and activator) and materials can be charged directly into a heated mold without using a mix head on the molding machine.

The great advantage of using the moderators of this invention is the extended gel time that they provide at convenient molding temperatures. At 80°C, the gel time can be extended to as long as three minutes and more, whereas solutions containing conventional rate moderators usually gel within a maximum of 15 to 20 seconds. That extended gel time, during which the reaction mixture remains highly fluid, allows the reaction mixture to be used in techniques in which molds are filled slowly. For example, the mixture can be employed in rotational molding, using centrifugal force to distribute the mixture before polymerization. The mixtures

are also useful in preparing polymer articles containing glass or other fibrous mat reinforcement, which must be completely impregnated while the mixture remains fluid. Also, in the manufacture of large objects, where the volume of the mold requires relatively long filling time, the bidentate Lewis base moderators are useful. Using the process according to the invention, molds can usually be charged at the polymerization temperature.

In the known RIM processing, the components are most conveniently combined by mixing equal parts of two reactive streams, one of which contains twice the desired concentration of the catalyst component and the other contains twice the desired concentration of the activator. That procedure is possible but not necessary in the process according to the invention, in which it may be convenient to add one part of the system, the catalyst component or the activator component, to substantially all of the monomer, and then to add a concentrate of the other part just prior to the polymerization and molding step.

The invention is illustrated by the following examples. In these examples, the catalyst component was prepared by suspending a $WCl_6/WOCl_4$ complex in toluene, reacting it with nonylphenol to solubilize it and complexing it with acetylacetone. This product was then diluted to a 0.1 molar concentration by adding sufficient additional toluene. A 1.0 molar toluene solution of an 85:15 molar mixture of tri-n-octyl aluminum (TNOA) and dioctyl aluminum iodide (DOAI) was prepared. One equivalent of bis(methoxyethyl) ether (diglyme) per mole of combined TNOA and DOAI was added to form a standard, control activator for dicyclopentadiene polymerization.

Control Examples

To a nitrogen-sparged vessel was charged 5 volumes of dicyclopentadiene. To this was added 0.06 volume of the

standard 85:15 TNOA/DOAI mixture, and the mixture was mixed well. After the mixing, 0.2 volume of the tungsten catalyst component solution was injected and mixed well. The vessel was immersed in a constant temperature bath maintained at 80°C.

The same procedure was simultaneously carried out using a 1.0 M TNOA solution free of DOAI as the activator.

The time from addition of the tungsten catalyst component until formation of a non-fluid gel was noted and recorded as the gel time. Similarly the time from addition of the catalyst until the temperature reached one-half of the final exotherm temperature was noted and recorded as the induction time or cure time. For these control examples, the values are recorded in Table I.

1.5

Examples 1 to 9

The procedure used to test the control or standard activators was followed using similar molar quantities of various bidentate Lewis base as the moderator in place of diglyme. Moderators tested and the gel and cure times achieved therewith are recorded in Table I.

20

Table I

	Example No.	Activator	Moderator	Gel Time (sec.)	Cure Time (sec.)
5	Control	TNOA/DOAI	diglyme	15	22
	Control	TNOA	diglyme	3	15
10	1	DOAI	dibenzoylmethane	135	191
	2	DOAI	dibenzoylmethane*	105	154
	3	DOAI	acetylacetone	140	210
	4	DOAI	acetylacetone**	103	139
	5	DOAI	HFA***	146	292
	6	DOAI	8-hydroxyquinoline****	225	260
	7	TNOA	THFA	78	123
15	8	TNOA/DOAI	acetyl acetone	115	156
	9	TNOA/DOAI	dibenzoyl methane	136	205

*In form of $Al(DBM)_3$.¹

**In form of $Al(AcAc)_3$.¹

***Hexafluoroacetylacetone.

****In form of $Al(HQ)_3$.¹

20 THFA = tetrahydrofurfuryl alcohol

HQ = 8-hydroxyquinoline

1 These complexes were prepared by reacting one equivalent of TNOA with three equivalents of the bifunctional Lewis base moderator.

The data from Examples 1 through 9 clearly show the significant increase in gel and cure times of the dicyclopentadiene solutions containing bidentate Lewis base rate moderators.

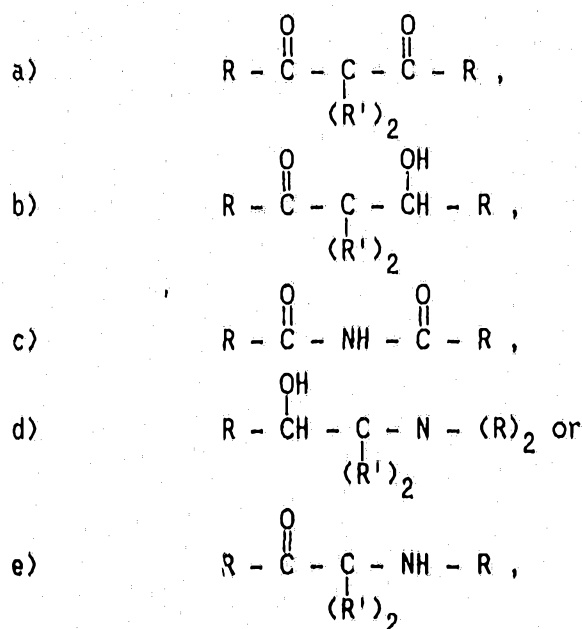
The claims defining the invention are as follows:

1. A process for preparing a polymer by polymerizing a metathesis-polymerizable polycyclic cycloolefin, in which the cycloolefin monomer is metathesis-polymerized in the presence of a metathesis-polymerization catalyst and a bidentate activator complex, said complex being prepared from an alkyl aluminium catalyst activator and a reaction rate moderator characterized in that the reaction rate moderator is a bidentate Lewis base having a first functional group that is chemically reactive with said catalyst activator and a second functional group containing an atom having unpaired electrons.

2. A process for preparing molded objects by polymerizing a metathesis-polymerizable polycyclic cycloolefin, in which the cycloolefin monomer, a metathesis-polymerization catalyst and a bidentate activator complex, said complex being prepared from an alkyl aluminium catalyst activator and a reaction rate moderator are charged into a mold in which polymerization takes place, characterized in that the reaction rate moderator is a bidentate Lewis base having a first functional group that is chemically reactive with said catalyst activator and a second functional group containing an atom having unpaired electrons.

3. A process as claimed in claim 1 or claim 2, in which said two functional groups are in a 1,3 relationship, whereby the Lewis base can form an adduct with aluminium.

4. A process as claimed in any one of claims 1 to 3, in which the bidentate Lewis base has the general formula:



wherein R is a C₁-C₅ alkyl radical or a phenyl or benzyl radical, R' is a C₁-C₅ alkyl radical or hydrogen, and if there is a plurality of R or R' groups in the structure, they may be the same or different.

5. A process as claimed in any one of claims 1 to 4, in which the bidentate Lewis base is tetrahydrofurfuryl alcohol, 8-hydroxyquinoline, acetylacetone, or dibenzoylmethane.

6. A process as claimed in any one of claims 1 to 5, in which the alkyl aluminium catalyst activator is combined with the bidentate Lewis base at a 1:1 to 1:5 molar ratio.

7. A process as claimed in any one of claims 1 to 6, which includes the step of adding the Lewis base to the alkyl aluminium catalyst activator with agitation while maintaining the reaction temperature at less than approximately 50°C.

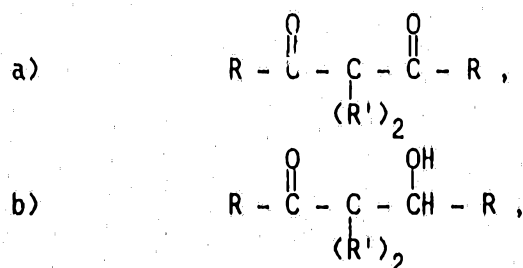
8. A process as claimed in any one of claims 1 to 7, in which the cycloolefin monomer is dicyclopentadiene.

9. A process for preparing a polymer by polymerizing a metathesis polymerizable polycyclic cycloolefin, which process is substantially as hereinbefore described with reference to any one of Examples 1 to 9.

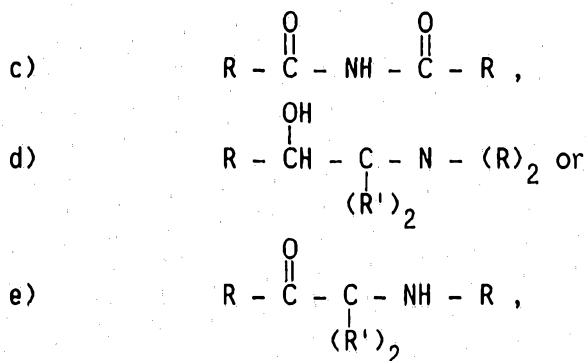
10. A polymerization composition comprising a metathesis polymerizable polycyclic cycloolefin, a metathesis polymerization catalyst and a bidentate activator complex, said complex being prepared from an alkyl aluminium catalyst activator and a reaction rate moderator, characterized in that the reaction rate moderator is a bidentate Lewis base having a first functional group that is chemically reactive with said catalyst activator and a second functional group containing an atom having unpaired electrons.

11. A composition as claimed in claim 10, in which said two functional groups are in a 1,3 relationship, whereby the Lewis base can form an adduct with aluminium.

12. A composition as claimed in claim 10 or claim 11, in which the bidentate Lewis base has the general formula:



LMM/329S



where R is a C₁-C₅ alkyl radical or a phenyl or benzyl radical, R' is a C₁-C₅ alkyl radical or hydrogen, and if there is a plurality of R or R' groups in the structure, they may be the same or different.

13. A composition as claimed in any one of claims 10 to 12, in which the bidentate Lewis base is tetrahydrofurfuryl alcohol, 8-hydroxyquinoline, acetylacetone, or dibenzoylmethane.

14. A composition as claimed in any one of claims 10 to 13, in which the bidentate activator complex was prepared from an alkyl aluminium catalyst activator and a bidentate Lewis base in a molar ratio of 1:1 to 1:5.

15. A composition as claimed in any one of claims 10 to 14, wherein the Lewis base is added to the alkyl aluminium catalyst activator with agitation while maintaining the reaction temperature at less than approximately 50°C.

16. A composition as claimed in any one of claims 10 to 15, in which the cycloolefin monomer is dicyclopentadiene.

17. A polymerization composition substantially as hereinbefore described with reference to any one of Examples 1 to 9.

18. The product of the process of any one of claims 1 to 9.

DATED this SEVENTEENTH day of AUGUST 1990
Hercules Incorporated

Patent Attorneys for the Applicant
SPRUSON & FERGUSON



LMM/329S