



PATENT SPECIFICATION

NO DRAWINGS

1.078.897

1.078.897



Inventors: MAKATO, YOKOO, AKIZO KESHI,
NORISHINGE HASHIMOTO and SHIGENOBU NAKAHARA

Date of Application and filing Complete Specification: Sept. 18, 1964.

No. 57000/66.

(Divided out of No. 1,078,896.)

Complete Specification Published: Aug. 9, 1967.

© Crown Copyright 1967.

Index at acceptance:—C3 R(32B1, 32B2, 32D1, 32D2, 32D5, 32D6A, 32D6C, 32D6D, 32E1, 32E6, 32E9, 32G1, 32H5F, 32H8, 32J1, 32J2A, 32J2Y; C3 C1E2

Int. Cl.:—C 08 g 21/04, C 08 g 53/10 // C08 j

COMPLETE SPECIFICATION

A process for the production of Polyurethane Foam

We, TAKEDA CHEMICAL INDUSTRIES LIMITED, of 27, Doshomachi 2-chome, Higashi-ku, Osaka, Japan, a company organised under the laws of Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a process for the production of polyurethane foam, and in particular, to a process for the production of polyurethane foam using a novel catalyst.

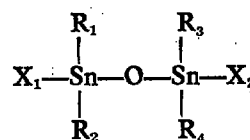
Many compounds have been proposed for use as catalysts for the reaction of isocyanates which compounds having active hydrogen atoms, tin compounds are generally preferred for practical purposes.

As a result of many investigations by the present inventors it was unexpectedly found that, as the catalyst for the reaction of an isocyanate with a compound having active hydrogen atoms, tetra-alkyldistannoxane derivatives having the general formula described below have a catalytic activity several tens times as high as the compounds which have been said to have the highest catalytic activity, exemplified by dibutyltindilaurate, tin (11)-2-ethylhexoate, triethylenediamine, and others and that the tetra-alkyldistannoxane derivative can be used as the sole catalyst for the production of polyurethane foam, moreover, they are more stable than the hitherto employed catalysts.

The catalytic activity of these tetra-alkyldistannoxane derivatives is indeed unexpected, since no other catalyst is known that can itself accelerate both the polymerisation reaction (i.e. the reaction between —NCO groups and —OH groups) and the foaming reaction (i.e. gas blowing reaction) so as to maintain a good balance between the two reactions to produce polyurethane foam having good properties and stability.

Due to the high activity of the tetra-alkyldistannoxanes as catalysts, it is possible to obtain polyurethane foam using only small amounts of the tetra-alkyldistannoxanes as catalysts, these amounts being about half to about one-tenth of the amounts previously used of previously proposed catalysts, without the necessity for heating the reactants. Moreover, the thus-obtained foam has good properties. As the tetra-alkyldistannoxanes are very stable, a so-called pre-mix including such compounds as catalysts can be stored for a long period without suffering any deterioration. The compounds are readily soluble in polyetherpolyols and polyesterpolyols so that polyurethane foam can be obtained by simple procedures. The tetra-alkyldistannoxanes are very effective catalysts for the production of polyurethane foam.

According to the invention there is provided a process for the production of a polyurethane foam, which comprises reacting a polyisocyanate with a polyetherpolyol or a polyesterpolyol in the presence of a blowing agent and a catalyst but without a carrier for said catalyst, said catalyst having the following general formula:



wherein X₁ is an hydroxyl radical an alkoxy radical, an acyloxy radical or an —NCX radical in which X is an oxygen or a sulphur atom and X₂ is a halogen atom, an hydroxyl radical, an alkoxy radical, an acyloxy radical or an —NCX radical in which X is an oxygen or sulphur atom, R₁, R₂, R₃ and R₄ are each an alkyl radical.

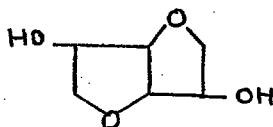
In the general formula, X₁ and X₂ are the same or different, and each stands for a

- halogen atom (e.g. chlorine, bromine or iodine), an hydroxyl radical, an alkoxy radical (e.g. a methoxy, ethoxy, propoxy or butoxy radical), a acyloxy radical (e.g. a formyloxy, acetoxy, propionoyloxy, butyroyloxy, hexanoyloxy, lauroyloxy or stearoyloxy radical). However, the compounds in which both X_1 and X_2 are halogen atoms, are excluded because such compounds have a low activity as catalysts.
- 10 R_1 , R_2 , R_3 and R_4 are same or different, and each stands for an alkyl radical such as a methyl, ethyl, propyl, butyl, isopropyl, heptyl or hexyl radical, preferably, an alkyl radical having one to four carbon atoms such as a methyl, ethyl, propyl, iso-propyl or butyl radical. In the $-NCX$ radicals, X is an oxygen or sulphur atom.
- 15 These compounds are prepared, for example, by the procedure described in "Journal of Organometallic Chemistry" Volume 1, No. 1, page 81—88 (1963). The preferred compounds of the general formula above are those wherein X_1 is a halogen atom and X_2 is an hydroxyl radical; both X_1 and X_2 are acyloxy radicals; X_1 is an acyloxy radical and X_2 is an hydroxyl radical; X_1 is an alkoxy radical and X_2 is a halogen atom; X_1 is an alkoxy radical and X_2 is an acyloxy radical; or both X_1 and X_2 are $-NCX$ radicals; that is, X_1 is an $-NCO$ radical and X_2 is an $-NCS$ radical or both X_1 and X_2 are $-NCO$ or $-NCS$ radicals. Carboxylic acyloxy radicals having one to eighteen carbon atoms, and alkoxy radicals have one to three carbon atoms are desirable.
- 20 Of these compounds described above, the preferred compounds for practical purposes are the following:—
- 25 Tetra - butyl - 1 - chloro - 3 - hydroxy-distannoxane
- 30 Tetra - butyl - 1,3 - diacetox-distannoxane
- 35 Tetra - butyl - 1,3 - distearoyloxy - distannoxane
- 40 Tetra - butyl - 1,3 - diformyloxy - distannoxane
- 45 Tetra - butyl - 1,3 - di - (2 - ethyl-hexanoyloxy) - distannoxane
- 50 Tetra - propyl - 1 - chloro - 3 - hydroxy-distannoxane
- 55 Tetra - butyl - 1,3 - dilauroyloxy - distannoxane
- 60 Tetra - ethyl - 1 - chloro - 3 - hydroxy-distannoxane
- 65 Tetra - methyl - 1 - chloro - 3 - hydroxy-distannoxane
- Tetra - propyl - 1,3 - diacetox - distannoxane
- Tetra - ethyl - 1,3 - diacetox - distannoxane
- Tetra - methyl - 1,3 - distearoyloxy - distannoxane
- Tetra - methyl - 1,3 - diacetox - distannoxane
- Tetra - butyl - 1 - hydroxy - 3 - acetoxy-distannoxane
- Tetra - butyl - 1 - hydroxy - 3 - formyloxy-distannoxane
- Tetra - propyl - 1 - hydroxy - 3 - acetoxy-distannoxane
- 70 Tetra - butyl - 1 - hydroxy - 3 - lauroyloxy-distannoxane
- Tetra - ethyl - 1 - hydroxy - 3 - acetoxy-distannoxane
- 75 Tetra - ethyl - 1 hydroxy - 3 - (2 - ethyl-hexanoyloxy) - distannoxane
- Tetra - butyl - 1 - chloro - 3 - methoxy-distannoxane
- Tetra - butyl - 1 - chloro - 3 - ethoxy-distannoxane
- 80 Tetra - butyl - 1 - chloro - 3 - propoxy-distannoxane
- Tetra - propyl - 1 - chloro - 3 - methoxy-distannoxane
- 85 Tetra - ethyl - 1 - chloro - 3 - methoxy-distannoxane
- Tetra - butyl - 1,3 - diisocyanate - distannoxane
- 90 Tetra - butyl - 1,3 - diisothiocyanate - distannoxane
- Tetra - propyl - 1,3 - diisocyanate - distannoxane
- 95 Tetra - ethyl - 1,3 - diisocyanate - distannoxane
- Tetra - ethyl - 1,3 - diisothiocyanate - distannoxane
- 100 Tetra - propyl - 1,3 - diformyloxy - distannoxane
- Tetra - ethyl - 1,3 - diformyloxy - distannoxane
- 105 Tetra - methyl - 1,3 - diformyloxy - distannoxane
- Tetra - methyl - 1 - acetoxy - 3 - hydroxy-distannoxane
- 110 Tetra - ethyl - 1 - formyloxy - 3 - hydroxy-distannoxane
- Tetra - propyl - 1 - formyloxy - 3 - hydroxy - distannoxane
- 115 Tetra - butyl - 1 - methoxy - 3 - formyloxy-distannoxane
- Tetra - butyl - 1 - ethoxy - 3 - acetoxy-distannoxane
- Tetra - butyl - 1 - ethoxy - 3 - stearoyloxy-distannoxane
- 120 Tetra - butyl - 1 - ethoxy - 3 - formyloxy-distannoxane
- Tetra - propyl - 1 - methoxy - 3 - acetoxy-distannoxane
- 125 Tetra - butyl - 1 - propoxy - 3 - acetoxy-distannoxane
- Tetra - propyl - 1 - methoxy - 3 - lauroyloxy - distannoxane
- 130 It is to be appreciated that this invention is not restricted to the compounds exemplified above. Compounds other than the above-mentioned compounds, so far as they can be represented by the general formula, have also a higher catalytic activity than previously proposed catalysts, and have a good effect on



the production of polyurethane foam by themselves.

The polyetherpolyols which may be used in the present process include products obtained by the addition polymerization of one or more alkylene oxides, such as ethylene oxide, propyleneoxide, butyleneoxide, or styreneoxide, to one or more polyols, as initiators, such as ethylene glycol, diethylene glycol, propylene glycol, glycerol, trimethylol propane, hexane triol, pentaerithritol, sorbitol, sucrose, mannitol, 1,4,3,6 - sorbide (a compound having the formula



derived from sorbitol by removal of 2 moles of water), mannitan, or sorbitan and amines such as ethylene diamine, propylene diamine, ethanol amine under alkaline or acidic conditions.

These polyetherpolyols may be prepared in the manner as described in "High polymers Vo. XIII. Polyethers Part 1" (1963) by Norman G. Gaylord published by Interscience Publishers.

Polyetherpolyols having three or more terminal hydroxyl groups are preferred.

The molecular weight of the polyetherpolyols may be varied according to the desired properties of polyurethane foam, and is generally selected to be from about 500 to about 10000.

The polyesterpolyols which may be used in the present process include products obtained by the reaction of one or more polyols, such as ethylene glycol, propylene glycol, diethylene glycol, triethylene glycol, butylene glycol, trimethylol propane, glycerol, hexane triol or pentaerithritol with one or more polycarboxylic acids, such as succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, maleic acid, fumaric acid, itaconic acid, phthalic acid, iso-phthalic acid, terephthalic acid, or their acid anhydrides.

These polyesterpolyols may be prepared in the manner described in "Polyesters and their Application" third printing. April 1959, published by Bjorksten Research Lab. Inc. New York, U.S.A.

Polyesterpolyols having two or more terminal hydroxyl groups are preferable.

The molecular weight of the polyesterpolyols may vary with the desired properties of polyurethane foam, and is generally selected to be from about 500 to 10000.

Polyetherpolyols or polyesterpolyols, especially polyetherpolyols having three or more terminal hydroxyl groups, preferably three to nine terminal hydroxyl groups, and a hydroxyl number of 350 to 700 are preferably used in

order to obtain so-called rigid polyurethane foam.

The polyisocyanates which may be employed in the present process include such aromatic polyisocyanates as tolylene diisocyanate (TDI), diphenylmethane diisocyanate (MDI), dianisidine diisocyanate, biphenylene diisocyanate, naphthylene diisocyanate, phenylene diisocyanate, triphenylmethane triisocyanate, or such aliphatic polyisocyanates as tetramethylene diisocyanate, hexamethylene diisocyanate, and addition products having two or more terminal —NCO groups such as those which are obtained by the reaction of an excess of the above mentioned polyisocyanates with lower molecular weight polyols, such as ethylene glycol, propylene glycol, glycerol, hexane triol or trimethylol propane, or with the polyetherpolyols or polyesterpolyols described above.

The preferred polyisocyanates are aromatic polyisocyanates, or addition products having two or more —NCO groups which are obtained by the reaction of an excess of the aromatic polyisocyanates with lower molecular weight polyols, or with polyetherpolyols or polyesterpolyols; especially tolylene diisocyanate (TDI), diphenylmethane diisocyanate (MDI) or the addition products of the lower molecular weight polyols or polyetherpolyols with excess TDI or MDI.

In carrying out the present process a polyetherpolyol or polyesterpolyol is allowed to react with a polyisocyanate in the presence of a blowing agent and a compound of the general formula mentioned above but without a carrier for said compound.

The blowing agents may be any of those known per se, for example, water, compounds generating water during the reaction, halogenated hydrocarbons having low boiling points, such as dichlorodifluoromethane, trichloromonofluoromethane, or compounds generating nitrogen gas during the reaction, such as azo compounds.

The amount of the compound having the general formula (I) used as a catalyst in the present process is half to one-tenth of that of previously proposed catalysts, that is, about 0.02 to about 0.1 part by weight to 100 parts of the total weight of polyisocyanate component and polyetherpolyol or polyesterpolyol component. However, if desired, the amount of the catalyst used in the present process may be more than that above mentioned, for example, about 0.1 to about 0.3 part by weight to 100 parts of the total weight of the two reactants described above.

The present process may be carried out in the presence of, for example, a foam stabilizer (e.g. a silicon oil- a poly-dimethyl siloxane or an alkylsilanepolyoxyalkylene copolymer- a non-ionic surface active agent, such as sorbitan monostearate or glycerol monooleate), a pigment, a reinforcing material, a dye, an anti-

oxidant, a fireproofing agent, or other known additives, or a mixture of any two or more of such additives.

5 The present process may be carried out using the procedure described in "High Polymers. Vol. XVI. Polyurethanes: Chemistry and Technology I, Chemistry" by Saunders and Frisch (1962) published by Interscience Publishers.

10 As stated above, since a compound having the general formula above has an extraordinary high activity as a catalyst, polyurethane foam can be easily obtained, without heating, using only a very small amount of the compound having the general formula above as a catalyst. Moreover, since the compounds having the general formula above are very stable themselves, so-called pre-mixes including these compounds as catalysts can be stored for a long while without suffering any deterioration. In addition to these advantages, since the compounds having the general formula above are readily soluble in polyetherpolyols and polyesterpolyols, only simple procedures are required for the production of polyurethane foams.

It is thus clear that the use of a compound having the general formula above as a catalyst has much value from the industrial viewpoint.

30 In our copending application No. 38270/64 (Serial No. 1078896) there is described and claimed a process for the production of a polyurethane foam, which comprises reacting a polyisocyanate with a polyetherpolyol or a

polyesterpolyol in the presence of a blowing agent, a tertiary amine and a compound of the general formula given above.

For the purpose of a better understanding of this invention, the following examples are given. However, it is to be understood that this invention is not limited to these examples.

In these examples, the amount of the compounds are expressed by parts by weight unless otherwise specified.

EXAMPLE 1

A catalyst as shown in the following table in an amount of 0.005 part, 1 ml. of dioxane, 7.0 parts of polypropylene glycol of molecular weight 460 obtained by addition polymerization of propylene oxide to propylene glycol as an initiator, and 2.4 parts of tolylene diisocyanate containing 80 parts of the 2,4-isomer and 20 parts of the 2,6-isomer were thoroughly mixed. Soon after the mixture was formed, a test tube was filled with the mixture and placed in a 70°C constant temperature bath.

The gelation time was measured from the instant the tube was placed in the bath, the gelation time being taken as the time when the mixture will no longer visibly flow when the test tube is inverted.

The results obtained are as follows: In Tables 1 and 2 Me, Pr, Bu, Ac, St, La and 2 - Et - Hex respectively denote methyl, propyl, butyl, acetyl, stearoyl, lauroyl and 2-ethyl-hexanoyl.

TABLE I

Catalyst	Gelation time min.
None	>200
Triethylene diamine	>200
$\text{Me}_2\text{-Sn-NO}_2(\text{OH})$	105
Me_2SnCl_2	>200
$\left(\text{C}_6\text{H}_5\right)_2\text{SnCl}_2$	>200
$\text{Cl-(Pr)}_2\text{SnOSn(Pr)}_2\text{-Cl}$	>200
$\text{Cl-(Bu)}_2\text{SnOSn(Bu)}_2\text{-Cl}$	>200
$\text{Cl-(Pr)}_2\text{SnOSn(Pr)}_2\text{-OH}$	2
$\text{Cl-(Bu)}_2\text{SnOSn(Bu)}_2\text{-OH}$	2
$(\text{AcO}) (\text{Pr})_2\text{SnOSn(Pr)}_2(\text{AcO})$	2
$(\text{AcO}) (\text{Bu})_2\text{SnOSn(Bu)}_2(\text{AcO})$	2
$(\text{AcO}) (\text{Bu})_2\text{SnOSn(Bu)}_2\text{OH}$	2
$\text{NCO(Bu)}_2\text{SnOSn(Bu)}_2\text{NCO}$	2 — 4
$\text{Cl-(Me)}_2\text{SnOSn(Me)}_2(\text{OCH}_3)$	2 — 4
$(\text{StO})(\text{Bu})_2\text{SnOSn(Bu)}_2(\text{OSt})$	2
$(\text{LaO})(\text{Bu})_2\text{SnOSn(Bu)}_2(\text{OLa})$	2
$(2\text{-Et-Hex-O})(\text{Bu})_2\text{SnOSn(Bu)}_2(\text{O-2-Et-Hex})$	2

EXAMPLE 2

Example 1 was repeated using the same reactants and the same procedure as in

5 Example 1 except that 3.5 parts of 4,4'-di-

phenylmethane diisocyanate was used in place of 2.4 parts of tolylene diisocyanate, and the dioxane was omitted. The gelation time was measured, the results being as follows:—

TABLE 2

Catalyst	Gelation time min.
None	60
Dibutyltindilaurate	14
Tin(II)-2-ethylhexoate	10
$(\text{AcO})(\text{Pr})_2\text{SnOSn(Pr)}_2(\text{OAc})$	0.5
$\text{Cl(Pr)}_2\text{SnOSn(Pr)}_2\text{OH}$	1.5
$\text{Cl(Bu)}_2\text{SnOSn(Bu)}_2\text{OH}$	1.0



Component A:

EXAMPLE 3

5 Polyethertriol of hydroxyl number 56 obtained by the addition polymerization of propylene oxide with trimethylol propane as an initiator

Tetra-butyl-1,3-diacetoxydi-
stannoxane — 100 parts
— 0.2 part

Component B:

10 Water — 4.0 parts
Silicone oil — 2.0 parts

Component C:

15 Tolylene diisocyanate (2,4-isomer 80 parts, 2,6-isomer 20 parts) — 49.6 parts

Components A, B and C were homogeneously mixed at 25°C. to allow reaction to take place, and a white elastic foam having a density of 0.026 g./cm³. was obtained; the rise time was 1 minute 50 seconds and the gelation time was 2 minutes 25 seconds.

EXAMPLE 4

Component A:

25 Polyetherpolyol of hydroxyl number 530 obtained by the addition polymerization of propylene oxide with sorbitol as an initiator

— 100 parts
Silicone oil — 1 part

1-1

30 Tetra - butyl - 1/chloro - 3-hydroxy - distannoxane — 0.35 part

Component B:

Prepolymer having an amine equivalent of 139 and an acid number of 0.005 obtained by the reaction of excess tolylene diisocyanate (2,4 - isomer 80 parts, 2,6 - isomer 20 parts) with the same polyetherpolyol as in component A — 139 parts

40 Components A and B were homogeneously mixed at 22°C. to allow a reaction to take place, a tough and rigid foam having a density of 0.033 g./cm³. being obtained; the cream time was 40 seconds, the rise time was 1 minute 25 seconds and the gelation time was 1 minute 30 seconds.

EXAMPLE 5

45 A polyethertriol of hydroxyl number 56, obtained by the addition polymerization of propylene oxide with hexane triol as an initiator (100 parts), water (4.0 parts), silicone oil (2.0 parts), tolylene diisocyanate containing 80 parts of 2,4-isomer and 20 parts of 2,6-isomer (49.6 parts) and tetrabutyl-1,3-diacetoxydistannoxane (0.17 part) were mixed to allow a reaction to take place at room temperature, and a white elastic foam having a density of 0.028 g./cm³, a tensile strength of 1.11 kg./cm². and an elongation of 220% was obtained.

EXAMPLE 6

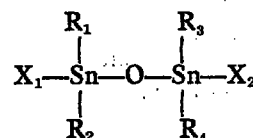
60 Example 5 was repeated except that 0.17 part of tetra - butyl - 1/acetoxo - 3 - hydroxy-distannoxane were used in place of 0.17 part of tetra - butyl - 1,3 - diacetoxydistannoxane,

and an elastic foam having a density of 0.027 g./cm³, a tensile strength of 1.19 kg./cm². and an elongation of 250% was obtained.

65

WHAT WE CLAIM IS:—

1. A process for the production of a polyurethane foam, which comprises reacting a polyisocyanate with a polyetherpolyol or a polyesterpolyol in the presence of a blowing agent and a catalyst but without a carrier for said catalyst, said catalyst having the following general formula:



75

wherein X₁ is an hydroxyl radical an alkoxy radical, an acyloxy radical or an —NCX radical in which X is an oxygen or sulphur atom, X₂ is a halogen atom, an hydroxyl radical, an alkoxy radical, an acyloxy radical or an —NCX radical in which X is an oxygen or sulphur atom, R₁, R₂, R₃ and R₄ are each an alkyl radical.

80

2. A process as claimed in Claim 1, wherein a polyetherpolyol having three or more terminal hydroxyl radicals is used.

85

3. A process as claimed in Claim 2, wherein the polyetherpolyol has a hydroxyl number of 350 to 700.

4. A process as claimed in any preceding claim, wherein the polyisocyanate is tolylene diisocyanate.

90

5. A process as claimed in any of claims 1 to 3, wherein the polyisocyanate is diphenylmethane diisocyanate.

95

6. A process as claimed in any preceding claim, wherein X₁ and X₂ in the general formula are each an —NCX group in which X is an oxygen or sulphur atom.

7. A process as claimed in any of claims 1 to 5, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1 - chloro - 3 - hydroxy - distannoxane.

100

8. A process as claimed in any one of Claims 1 to 5, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1 - acetoxo - 3 - hydroxy - distannoxane.

105

9. A process as claimed in any one of claims 1 to 5, wherein the compound having the general formula defined in Claim 1 is tetrabutyl - 1,3 - diacetoxy - distannoxane.

10. A process as claimed in any one of Claims 1 to 5, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1 - methoxy - 3 - acetoxo - distannoxane.

110

11. A process as claimed in any one of Claims 1 to 6, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1,3 - diisocyanate - distannoxane.

115



Component A:

EXAMPLE 3

5 Polyethertriol of hydroxyl number 56 obtained by the addition polymerization of propylene oxide with trimethylol propane as an initiator

Tetra-butyl-1,3-diacetoxydi-
stannoxane — 100 parts
— 0.2 part

Component B:

10 Water — 4.0 parts
Silicone oil — 2.0 parts

Component C:

15 Tolylene diisocyanate (2,4-isomer 80 parts, 2,6-isomer 20 parts) — 49.6 parts

Components A, B and C were homogeneously mixed at 25°C. to allow reaction to take place, and a white elastic foam having a density of 0.026 g./cm³. was obtained; the rise time was 1 minute 50 seconds and the gelation time was 2 minutes 25 seconds.

EXAMPLE 4

Component A:

25 Polyetherpolyol of hydroxyl number 530 obtained by the addition polymerization of propylene oxide with sorbitol as an initiator — 100 parts

Silicone oil — 1 part

[-]

30 Tetra - butyl - 1/chloro - 3-hydroxy - distannoxane — 0.35 part

Component B:

Prepolymer having an amine equivalent of 139 and an acid number of 0.005 obtained by the reaction of excess tolylene diisocyanate (2,4 - isomer 80 parts, 2,6 - isomer 20 parts) with the same polyetherpolyol as in component A — 139 parts

40 Components A and B were homogeneously mixed at 22°C. to allow a reaction to take place, a tough and rigid foam having a density of 0.033 g./cm³. being obtained; the cream time was 40 seconds, the rise time was 1 minute 25 seconds and the gelation time was 1 minute 30 seconds.

EXAMPLE 5

45 A polyethertriol of hydroxyl number 56, obtained by the addition polymerization of propylene oxide with hexane triol as an initiator (100 parts), water (4.0 parts), silicone oil (2.0 parts), tolylene diisocyanate containing 80 parts of 2,4-isomer and 20 parts of 2,6-isomer (49.6 parts) and tetrabutyl-1,3-diacetoxydistannoxane (0.17 part) were mixed to allow a reaction to take place at room temperature, and a white elastic foam having a density of 0.028 g./cm³, a tensile strength of 1.11 kg./cm². and an elongation of 220% was obtained.

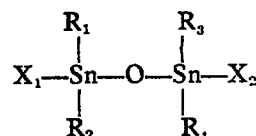
EXAMPLE 6

60 Example 5 was repeated except that 0.17 part of tetra - butyl - 1/acetoxo - 3 - hydroxy-distannoxane were used in place of 0.17 part of tetra - butyl - 1,3 - diacetoxydistannoxane,

and an elastic foam having a density of 0.027 g./cm³., a tensile strength of 1.19 kg./cm². and an elongation of 250% was obtained. 65

WHAT WE CLAIM IS:—

1. A process for the production of a polyurethane foam, which comprises reacting a polyisocyanate with a polyetherpolyol or a polyesterpolyol in the presence of a blowing agent and a catalyst but without a carrier for said catalyst, said catalyst having the following general formula: 70



75

wherein X₁ is an hydroxyl radical an alkoxy radical, an acyloxy radical or an —NCX radical in which X is an oxygen or sulphur atom, X₂ is a halogen atom, an hydroxyl radical, an alkoxy radical, an acyloxy radical or an —NCX radical in which X is an oxygen or sulphur atom, R₁, R₂, R₃ and R₄ are each an alkyl radical. 80

2. A process as claimed in Claim 1, wherein a polyetherpolyol having three or more terminal hydroxyl radicals is used. 85

3. A process as claimed in Claim 2, wherein the polyetherpolyol has a hydroxyl number of 350 to 700.

4. A process as claimed in any preceding claim, wherein the polyisocyanate is tolylene diisocyanate. 90

5. A process as claimed in any of claims 1 to 3, wherein the polyisocyanate is diphenylmethane diisocyanate. 95

6. A process as claimed in any preceding claim, wherein X₁ and X₂ in the general formula are each an —NCX group in which X is an oxygen or sulphur atom.

7. A process as claimed in any of claims 1 to 5, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1 - chloro - 3 - hydroxy distannoxane. 100

8. A process as claimed in any one of Claims 1 to 5, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1 - acetoxy - 3 - hydroxy-distannoxane. 105

9. A process as claimed in any one of claims 1 to 5, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1,3 - diacetoxy - distannoxane.

10. A process as claimed in any one of Claims 1 to 5, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1 - methoxy - 3 - acetoxy-distannoxane. 110

11. A process as claimed in any one of Claims 1 to 6, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1,3 - diisocyanate - distannoxane. 115



12. A process as claimed in any one of Claims 1 to 6, wherein the compound having the general formula defined in Claim 1 is tetra - butyl - 1,3 - diisothiocyanate - distannoxane.
13. A process for the production of a polyurethane foam in accordance with Claim 1 substantially as hereinbefore described in any one of the foregoing Examples.
14. A polyurethane foam produced by the process claimed in any preceding claim.

TREGEAR, THIEMANN & BLEACH,
Chartered Patent Agents,
Melbourne House, Aldwych, London, W.C.2.
Agents for the Applicants.

Leamington Spa: Printed for Her Majesty's Stationery Office, by the Courier Press.
—1967. Published by The Patent Office, 25 Southampton Buildings, London, W.C.2,
from which copies may be obtained.



* C L N S *