

PATENT SPECIFICATION

NO DRAWINGS

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COMPLETE SPECIFICATION

A process for the production of Polyurethane Foam

We, TAKEDA CHEMICAL INDUSTRIES LIMITED, a Company organised under the laws of Japan, of 27 Doshomachi, 2-chome, Higashi-ku, Osaka, 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 with compounds having active hydrogen atoms, tin compounds are generally preferred for practical purposes.

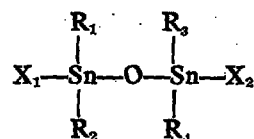
As a result of many investigations by the present inventors it was unexpectedly found that, as the catalyst for the reaction of an isocyanate with a compound having active hydrogen atoms, tetra-alkyldistannoxane derivatives having the general formula (1) described below have a catalytic activity several 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.

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.

Based on the foregoing, the present invention provides 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 tertiary amine, a blowing agent and a compound having the following general formula:



(Formula 1)

wherein, X_1 is an hydroxyl radical, an alkoxy radical, a acyloxy radical, or an —NCX radical in which X is an oxygen or sulphur atom, X_2 is a halogen atom, an hydroxyl radical, an alkoxy radical, a acyloxy radical, or an —NCX radical in which X is an oxygen or sulphur atom, R_1 , R_2 , R_3 , and R_4 are, each an alkyl radical.

In the general formula (1), X_1 and X_2 are 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, propionyloxy, butyroyloxy, hexanoyloxy, lauroyloxy or stearoyloxy radical). However,

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the compounds in which both X_1 and X_2 are halogen atoms, are excluded because such compounds have a low activity as catalysts.

- 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.

- 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 (1) 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 a 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 a 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.

- Of these compounds described above, the preferred compounds for practical purposes are the following:—

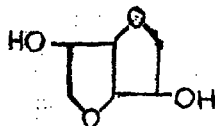
- Tetra - butyl - 1 - chloro - 3 - hydroxy-distannoxane
 Tetra - butyl - 1,3 - diacetoxo - distannoxane
 35 Tetra - butyl - 1,3 - distearoyloxy - distannoxane
 Tetra - butyl - 1,3 - diformyloxy - distannoxane
 Tetra - butyl - 1,3 - di - (2 - ethyl-hexanoyloxy) - distannoxane
 40 Tetra - propyl - 1 - chloro - 3 - hydroxy-distannoxane
 Tetra - butyl - 1,3 - dilauroyloxy - distannoxane
 45 Tetra - ethyl - 1 - chloro - 3 - hydroxy-distannoxane
 Tetra - methyl - 1 - chloro - 3 - hydroxy-distannoxane
 Tetra - propyl - 1,3 - diacetoxo - distannoxane
 50 Tetra - ethyl - 1,3 - diacetoxo - distannoxane
 Tetra - methyl - 1,3 - distearoyloxy - distannoxane
 55 Tetra - methyl - 1,3 - diacetoxo - distannoxane
 Tetra - butyl - 1 - hydroxy - 3 - acetoxo-distannoxane
 Tetra - butyl - 1 - hydroxy - 3 - formyloxy-distannoxane
 60 Tetra - propyl - 1 - hydroxy - 3 - acetoxo-distannoxane
 Tetra - butyl - 1 - hydroxy - 3 - lauroyloxy-distannoxane

- Tetra - ethyl - 1 - hydroxy - 3 - acetoxo-distannoxane
 Tetra - ethyl - 1 - hydroxy - 3 - (2 - ethyl-hexanoyloxy) - distannoxane
 Tetra - butyl - 1 - chloro - 3 - methoxy-distannoxane
 Tetra - butyl - 1 - chloro - 3 - ethoxy-distannoxane
 Tetra - butyl - 1 - chloro - 3 - propoxy-distannoxane
 Tetra - propyl - 1 - chloro - 3 - methoxy-distannoxane
 Tetra - ethyl - 1 - chloro - 3 - methoxy-distannoxane
 Tetra - butyl - 1,3 - diisocyanate - distannoxane
 Tetra - butyl - 1,3 - diisothiocyanate - distannoxane
 Tetra - propyl - 1,3 - diisocyanate - distannoxane
 Tetra - ethyl - 1,3 - diisocyanate - distannoxane
 Tetra - ethyl - 1,3 - diisothiocyanate - distannoxane
 Tetra - propyl - 1,3 - diformyloxy - distannoxane
 Tetra - ethyl - 1,3 - diformyloxy - distannoxane
 Tetra - methyl - 1,3 - diformyloxy - distannoxane
 Tetra - methyl - 1 - acetoxo - 3 - hydroxy-distannoxane
 Tetra - ethyl - 1 - formyloxy - 3 - hydroxy-distannoxane
 Tetra - propyl - 1 - formyloxy - 3 - hydroxy-distannoxane
 Tetra - butyl - 1 - methoxy - 3 - acetoxo-distannoxane
 Tetra - butyl - 1 - methoxy - 3 - formyloxy-distannoxane
 Tetra - butyl - 1 - ethoxy - 3 - acetoxo-distannoxane
 Tetra - butyl - 1 - ethoxy - 3 - stearoyloxy-distannoxane
 Tetra - butyl - 1 - ethoxy - 3 - formyloxy-distannoxane
 Tetra - propyl - 1 - methoxy - 3 - acetoxo-distannoxane
 Tetra - butyl - 1 - propoxy - 3 - acetoxo-distannoxane
 Tetra - propyl - 1 - methoxy - 3 - lauroyloxy - distannoxane

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 two 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 Vol. 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, or triphenylmethane triisocyanate; 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 compound having the general formula (1) mentioned above, a tertiary amine and a blowing agent.

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 (1) 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.

Tertiary amines which may be used in the present process include, for example, triethylamine, N-methyl morpholine, 1,4-diazabicyclo-(2,2,2) - octane, N,N,N',N' - tetra - methyl-1,3 - butane - diamine, N,N,N',N' - tetrakis (2-hydroxypropyl)-ethylene diamine, dimethylethanol amine, and N,N-(2-hydroxypropyl)-dimethyl morpholine.

The amount of these tertiary amines used may be about 0.03 to about 1.0 part by weight to 100 parts of the total weight of polyisocyanate component and polyetherpolyol or polyesterpolyol component, but, the amount,

if desired, may be more than that mentioned above.

The present process may be carried out in the presence of, for example, a foam stabilizer (e.g. a silicone oil- a poly-dimethyl siloxane or an alkylsilane-polyoxyalkylene copolymer-, a non-ionic surface active agent, such as sorbitan monostearate or glycerol monooleate), a pigment, a reinforcing material, a dye, an antioxidant, a fireproofing agent, or other known additives, or a mixture of any two or more of such additives.

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

As stated above, since a compound having the general formula (1) has an extraordinarily 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 (1) as a catalyst. Moreover, since the compounds having the general formula (1) 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 (1) are readily soluble in polyetherpolyols and polyesterpolyols, only simple procedures are required for the production of polyurethane foams.

In our copending application No. 57000/66 (Serial No. 1,078,897) 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 and a catalyst but without a carrier for said catalyst, said catalyst having the general formula (I).

It is thus clear that the use of a compound having the general formula (1) as a catalyst has much value from the industrial viewpoint.

The following Examples illustrate the invention.

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

EXAMPLE 1.

Component A:

Polyethertriol of hydroxyl number 56
obtained by the addition polymerization of propylene oxide with glycerol as an initiator. — 100 parts
Tetra-butyl-1,3-diacetoxystannoxane — 0.2 part

Component B:

Water — 4.0 parts
Silicone oil — 2.0 parts
1,4-diazabicyclo-(2,2,2)-octane — 0.07 part

Component C:

Toluene 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, a white elastic foam having a density of 0.025 g./cm³. being obtained; the cream time was 7 seconds, the rise time was 1 minute 20 seconds and the gelation time was 1 minute 45 seconds.

EXAMPLE 2.

Component A:

Polyether triol of hydroxyl number 56
obtained by the addition polymerization of propylene oxide with trimethylol propane as an initiator — 50 parts

Polyether Triol of hydroxyl number 160
obtained by the addition polymerization of propylene oxide with trimethylol propane as an initiator — 50 parts
Tetra-propyl-1,3-diacetoxystannoxane — 0.2 part

Component B:

Water — 4.0 parts
Silicone oil — 2.5 parts
1,4-diazabicyclo-(2,2,2)-octane — 0.07 part

Component C:

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

— 48.7 parts

Trichloromonofluoromethane

— 20 parts

- 5 Components A, B and C were homogeneously mixed at 23°C. to allow a reaction to take place, and a white elastic foam having a density of 0.016 g./cm³. was obtained; the rise time was 1 minute 50 seconds, and the gelation time was 2 minutes 23 seconds. 10

EXAMPLE 3.**Component A:**

- 15 Polyether triol of hydroxyl number 460 obtained by the addition polymerization of propylene oxide with glycerol as an initiator — 100 parts
Dimethylethanolamine — 1.5 parts
Silicone oil — 1.5 parts
Trichloromonofluoromethane — 30 parts
20 Tetra-butyl-1-acetoxy-3-hydroxy-distannoxane — 0.2 part

Component B:

- 25 Polyisocyanate compound having an amine equivalent of 108 and an acid number of 0.25 (Allied Chemical Corp., U.S.A., "Nacconate-4040"*) — 93 parts

- 30 Components A and B were homogeneously mixed at 21°C., to allow a reaction to take place, and a tough and rigid foam having a density of 0.032 g./cm³. was obtained; the cream time was 30 seconds, the rise time was 1 minute 20 seconds and the gelation time was 1 minute 20 seconds. 35

*"Nacconate-4040" is a registered Trade Mark of a phosgenation product of tolylene

diamine having the following properties: 35
Brown liquid free of sediment,
Amine equivalent 108,
Viscosity (Brookfield) 90 at 25°C.,
Specific gravity (25°C./25°C.) 1.26,
Solidification point lower than -15°C. and 40
Approximate flash point (Cleveland open cup) 132°C.

EXAMPLE 4.

- 45 **Component A:**
Polyethertriol of hydroxyl number 56 obtained by the addition polymerization of propylene oxide with trimethylol propane as an initiator — 100 parts
Tetra-butyl-1,3-diisothiocyanatedistannoxane — 0.1 part

- 50 **Component B:**
Water — 4.0 parts
Silicone oil — 2.0 parts
1,4-diazabicyclo-(2,2,2)-octane — 0.07 part

- 55 **Component C:**
Tolylene diisocyanate (2,4-isomer 80 parts, 2,6-isomer 20 parts) — 49.6 parts

- 60 Components A, B and C were homogeneously mixed at 20°C., and a white elastic foam having a density of 0.025 g./cm³. was obtained; the cream time was 10 seconds, the rise time was 1 minute 50 seconds and the gelation time was 2 minutes 50 seconds. 65

EXAMPLE 5.

100 Parts of a polyestertriol having a hydroxyl number of 57 which was obtained by the condensation of adipic acid (146 parts), diethylene glycol (127 parts) and trimethylol propane (13.4 parts) at 180°C., 2.5 parts of

water, 2.0 parts of a non-ionic surface active agent, 1.0 part of N-methyl-morpholine, 0.1 part of tetra-butyl-1,3-diacetoxydistannoxane and 35 parts of tolylene diisocyanate (2,4-isomer 80 parts, 2,6-isomer 20 parts) were homogeneously mixed at room temperature to allow a reaction to take place, and a beautiful white elastic foam having a density of 0.04 g./cm³. and a tensile strength of 1.5 kg./cm². was obtained; the rise time was 1 minute 40 seconds and the gelation time was 2 minutes 15 seconds.

EXAMPLE 6.

160 Parts of a polyestertriol having a hydroxyl number of 424 and an acid number of 6.1 which was obtained by the condensation of phthalic anhydride (296 parts), adipic acid (1168 parts), trimethylol propane (1980 parts) and 1,3-butylene glycol (360 parts), 165 parts of a prepolymer having an amine equivalent of 124.7 which was obtained by the reaction of tolylene diisocyanate containing 80 parts of 2,4-isomer and 20 parts of 2,6-isomer (48.5

parts), hexane triol (8 parts) and propylene glycol (2.0 parts), 52 parts of trichloromono-fluoromethane, 0.2 parts of tetra - butyl - 1,3-diacetoxydistannoxane, 0.5 part of dimethyl-ethanol amine, and 1.5 parts of silicone oil were thoroughly mixed to allow a reaction to take place, and a rigid foam having a density of 0.046 g./cm³., a compression load of 2.60 kg./cm². and a good resistance to chemicals was obtained; the rise time was 2 minutes and the gelation time was 3 minutes.

EXAMPLE 7.

So-called pre-mix formulations were prepared by mixing the components shown in Table 1 in the proportions specified therein. The thus prepared pre-mix formulations were stored at 13—15°C. for certain periods, a polyisocyanate component was added to each of these pre-mix formulations to produce a polyurethane foam, and both the cream time and the rise time were measured. The results are listed in Table 3.

TABLE 1

Formulation	1	2	3	4	5
Polyetherpolyol*	100	100	100	100	100
Silicone oil	1.5	1.5	1.5	—	—
Dimethylethanolamine	2.0	2.0	2.0	2.0	2.0
Tetra-butyl-1,3-diacetoxy-distannoxane	1.0	—	—	1.0	—
Dibutylindilaurate	—	1.0	—	—	1.0
Stannous octoate	—	—	1.0	—	—
Dibutyl phthalate	5.0	5.0	5.0	5.0	5.0
Trichloromonofluoromethane	30	30	30	30	30

*Polyetherpolyol of hydroxyl number 460 which was obtained by the addition polymerization of propylene oxide with sorbitol as an initiator.

initiator

TABLE 2

Formulation	1	2	3	4	5
Initial					
cream time (seconds)	25	50	28	25	50
rise time (seconds)	50	100	55	50	100
appearance of foam	G	G	G	G	G
After 70 days					
cream time (seconds)	25	X	65	25	50
rise time (seconds)	35	X	175	55	126
appearance of foam	G		G	G	G
After 100 days					
cream time (seconds)	23	X	75	26	48
rise time (seconds)	46	X	212	50	102
appearance of foam	G		C	G	G

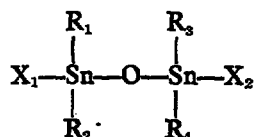
G: good

C: coarse

X: no foaming

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, a tertiary amine and a compound having the following general formula:



- wherein, X_1 is an hydroxyl radical, an alkoxy radical, a acyloxy radical, or an $-NCX$ radical in which X is an oxygen or sulphur atom, X_2 is a halogen atom, an hydroxyl radical, an alkoxy radical, a acyloxy radical, or an $-NCX$ radical in which X is an oxygen or sulphur atom, R_1 , R_2 , R_3 , and R_4 are each an alkyl radical.

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

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.

5. A process as claimed in anyone of Claims

1 to 3, wherein the polyisocyanate is diphenylmethane diisocyanate.

6. A process as claimed in any preceding claim, wherein X_1 and X_2 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 one of Claims 1 to 5, wherein the compound having the formula defined in Claim 1 is tetra - butyl - 1 - chloro - 3 - hydroxy - distannoxane.

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.

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.

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.

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 poly-



TABLE 2

Formulation	1	2	3	4	5
Initial					
cream time (seconds)	25	50	28	25	50
rise time (seconds)	50	100	55	50	100
appearance of foam	G	G	G	G	G
After 70 days					
cream time (seconds)	25	X	65	25	50
rise time (seconds)	35	X	175	55	126
appearance of foam	G		G	G	G
After 100 days					
cream time (seconds)	23	X	75	26	48
rise time (seconds)	46	X	212	50	102
appearance of foam	G		C	G	G

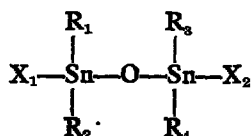
G: good

C: coarse

X: no foaming

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, a tertiary amine and a compound having the following general formula:



- 10 wherein, X_1 is an hydroxyl radical, an alkoxy radical, a acyloxy radical, or an $-NCX$ radical in which X is an oxygen or sulphur atom, X_2 is a halogen atom, an hydroxyl radical, an alkoxy radical, a acyloxy radical, or an $-NCX$ radical in which X is an oxygen or sulphur atom, R_1 , R_2 , R_3 , and R_4 are each an alkyl radical.

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

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.

5. A process as claimed in anyone of Claims

1 to 3, wherein the polyisocyanate is diphenylmethane diisocyanate.

6. A process as claimed in any preceding claim, wherein X_1 and X_2 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 one of Claims 1 to 5, wherein the compound having the formula defined in Claim 1 is tetra - butyl - 1 - chloro - 3 - hydroxy - distannoxane.

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.

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.

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.

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 poly-

urethane foam in accordance with Claim 1 substantially as hereinbefore described in any one of the foregoing Examples.

- 5 14. A polyurethane foam produced by the process claimed in any preceding claim.

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London, W.C.2.
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