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(72) Inventeurs/Inventors:
THOONEN, SANDER, NL;
VAN KOTEN, GERARD, NL;
DEELMAN, BERTH JAN, NL

(73) Propriétaire/Owner:
ATOFINA VLISSINGEN B.V., NL

(74) Agent: BORDEN LADNER GERVAIS LLP

(54) Titre : PROCEDE DE PRODUCTION DE TRIHALOGENURES DE MONOALKYLE-ETAIN
(54) Title: PROCESS FOR THE PRODUCTION OF MONOALKYLTIN TRIHALIDES

(57) Abrégé/Abstract:
The present invention is directed to a process for the production of monoalkyltin trihalides of the formula $RSnX_3$ wherein R-alkyl or cycloalkyl and X-Cl, Br or I, involving a redistribution reaction between tetraorganotins, triorganotin halides or diorganotin halides and tin tetrahalides, said process comprising contacting tetra-(R_4Sn), tri-(R_3SnX) or diorganotin halides (R_2SnX_2) with SnX_4 to afford said monoorganotin trihalides in the presence of at least one transition metal complex, said complex comprising at least one transition metal, M, selected from Group VIII of the periodic Table of elements, at least one monodentate ligand or bidentate ligand, L, L' or L'', and optionally one or more anions, X, of an organic or inorganic acid, as a catalyst or catalyst precursor.

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(71) Applicant *(for all designated States except US)*: **ATOFINA VLISSINGEN B.V.** [NL/NL]; Haven 9850, NL-4389 PD Vlissingen-Oost (NL).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: **THOONEN, Sander** [NL/NL]; Balearen 69, NL-3524 EL Utrecht (NL). **VAN KOTEN, Gerard** [NL/NL]; Paltzerweg 180, NL-3734 CN Den Dolder (NL). **DEELMAN, Berth, Jan** [NL/NL]; Egelantier 38, NL-4102 XD Culemborg (NL).

(74) Agent: **PRINS, A.W.**; Vereenigde, Nieuwe Parklaan 97, NL-2587 BN The Hague (NL).

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(54) Title: PROCESS FOR THE PRODUCTION OF MONOALKYLTIN TRIHALIDES

(57) Abstract: The present invention is directed to a process for the production of monoalkyltin trihalides of the formula $RSnX_3$ wherein R-alkyl or cycloalkyl and X-C1, Br or I, involving a redistribution reaction between tetraorganotins, triorganotin halides or diorganotin halides and tin tetrahalides, said process comprising contacting tetra-(R_4Sn), tri-(R_3SnX) or diorganotin halides (R_2SnX_2) with SnX_4 to afford said monoorganotin trihalides in the presence of at least one transition metal complex, said complex comprising at least one transition metal, M, selected from Group VIII of the periodic Table of elements, at least one monodentate ligand or bidentate ligand, L, L' or L", and optionally one or more anions, X, of an organic or inorganic acid, as a catalyst or catalyst precursor.

Process for the production of monoalkyltin trihalides

The present invention concerns a transition metal-catalyzed process for the production of monoalkyltin trihalides involving a redistribution reaction between tetraorganotins, triorganotin halides or diorganotin halides and tin tetrahalides.

Monoalkyltin trichlorides can be prepared industrially from tetraalkyltins and $SnCl_4$ according to the stoichiometry of eq. 1 (Neumann, W. P.; Burkhardt, G. *Liebigs Ann. Chem.* **1963**, ***663***, 11).

$$R_4Sn + 2\ SnCl_4 \rightarrow 2\ RSnCl_3 + R_2SnCl_2 \qquad (1)$$

$$R_4Sn + 3\ SnCl_4 \rightarrow 4\ RSnCl_3 \qquad (2)$$

$$R_2SnCl_2 + SnCl_4 \rightarrow 2\ RSnCl_3 \qquad (3)$$

(R = alkyl or cycloalkyl)

This process was further improved by M&T chemicals (Natoli, J. G., U.S. Pat. 3432531, 1969; Larkin, W. A.; Bouchoux, J. W., U.S. Pat. 3931264, 1976). In this process substantial amounts of dialkyltin dichloride are formed as by-product (typically around 33 %).

The reason that the process according to eq. 1 is often used and not that of eq. 2, is that eq. 3 does not proceed under mild conditions for longer alkyl groups. Eq. 3 would be the last step in a process according to eq. 2.

Nothwithstanding the above remarks, Neumann (US 3459779, 1969) has described the redistribution reaction of dialkyltin halides with $SnCl_4$ in $POCl_3/P_2O_5$ to produce monoalkyltin trichlorides. Also Langer et al. reported the formation of $MeSnCl_3$ from dimethyltin dichloride and tin tetrachloride in

dimethylsulfoxide (DMSO) (*Tetrahedron Lett.*, 1967, *1*, 43-47; US 3454610, 1969, to Dow Chemical Co.). Also the redistribution of dialkyltin dihalides, trialkyltin halides or tetraalkyltins with tin tetrahalide catalyzed by quarternary ammonium salts at temperatures above 150°C has been reported (Kugele, T.G.; Parker, D.H., US Pat. 3862198,1975) and more recently, redistribution reactions of organotins catalyzed by SnF_2 were claimed by Buschhoff et al. (US Pat. 4604475, 1986, to Schering A.G.).

All the processes described above generally use harsh reaction conditions and yields are often less than desirable.

It is therefore an object of the current invention to provide for a catalytic process for the production of monoorganotin trihalides from tetraalkyltins or polyalkyltin halides and tin tetrahalide that can be operated under mild conditions ($T < 150$°C, $p \leq 5$ bar) and which affords the product in high yield (> 60% based on Sn).

The reaction may be carried out at a temperature between 0 and 200°C.

The reaction may be carried out at a pressure between 1 and 5 bar.

The monoalkyltin trihalide product may be obtain in > 60% yield (based on Sn) in less than 48 h at temperatures below 150°C.

It is also an object of the current invention to make use of transition metal complexes as (pre)catalyst as opposed to previously reported catalysts.

In its broadest form the present invention comprises a process for the production of monoalkyltin trihalides of the formula $RSnX_3$, wherein R = alkyl or cycloalkyl and X = C1, Br or I, involving a redistribution reaction between tetraorganotins, trioganotin halides or diorganotin halides and tin tetrahalides, said process comprising contacting tetra- (R_4Sn), tri- (R_3SnX) or diorganotin halides (R_2SnX_2) with SnX_4 to afford said monoorganotin trihalides in the presence of at least one transition metal complex, said complex comprising at least one transition metal, M, selected from Group VIII of the periodic Table of elements, at least one monodentate ligand or bidentate ligand, L or L', and optionally one or more anions, Y, of an organic or inorganic acid, as a catalyst or catalyst precursor.

According to one embodiment of the invention, the catalyst is based on the use of a complex having the formula

$$L'MY_2 \qquad (I)$$

wherein L' is a bidentate ligand, or

L_2MY_2; (II)

wherein L is a monodentate ligand, or

L_4M (III)

wherein L is a monodentate ligand.

According to another embodiment, the said complex is:

$[L"M(\mu\text{-}Y)]_2$ (IV)

wherein L" = a cyclometallated bidentate optionally substituted o-(diarylphosphino)benzyl ligand. Catalysts of type IV have been applied for the Heck-vinylation of aryl halides (EP 725049 A1).

The metal to be used is a Group VIII metal, and preferred metals are Pt, Pd and/or Ni. The anions may be of organic and/or inorganic nature. It is preferred to use C1, Br, I, acetate, triflate or tosylate anions.

The current invention thus involves the use of transition metal complexes according to formula (I), (II), (III), or (IV) as a (pre-)catalyst in the redistribution reaction of tetra- (R_4Sn), tri- (R_3SnX) or diorganotin halides (R_2SnX_2) with SnX_4 to afford monoorganotine trihalides ($RSnX_3$; R = alkyl or cycloalkyl; X = C1, Br or I).

In a preferred embodiment of the invention L in formula (II) or (III) is selected from phosphine, alkene, amine, organic sulfide, nitrile and imidazoline-2-ylidene. L' is selected from diphosphine, dialkene, diamine and bis(imidazoline-2-ylidene) ligands, preferably optionally substituted o-{di(2-tolyl)phosphino}benzyl. More in particular, L is triarylphosphine or triphenylphosphine or L' = a bidentate nitrogenligand or *N,N,N',N'*-tetramethylethylenediamine (TMEDA), M is Pd or Pt and for catalyst (I), X is C1.

The catalyzed redistribution reaction concerns the redistribution of Bu_2SnCl_2 or Bu_4Sn with $SnCl_4$ to afford $BuSnCl_3$. The use of (pre)catalysts according to formula (I), (II), (III) and (IV) allows the formation of $BuSnCl_3$ from redistribution of Bu_2SnCl_2 or Bu_4Sn with $SnCl_4$ under mild reaction

conditions (T ≤ 150 °C, p ≤ 1 bar) in less than 24 hours and better than 70 % yield (based on Sn).

The group R is preferably defined as an alkyl (linear or branched), cycloalkyl or aryl, having from 1 to 12 C-atoms. Preferably methyl, *n*-butyl or *n*-hexyl are used.

The reaction can be carried out with or without a solvent. In general inert organic and aprotic solvents are preferred, especially aromatic solvents, chloroaromatics, alkanes, dialkylacetamides, *N*-alkylpyrolidones, dialkylamides of aliphatic carboxylic acids and organic nitriles. In particular toluene, *o*-xylene, 1,2-dichlorobenzene and *n*-octane were found to be appropriate solvents.

In a specific embodiment of the invention the concentration of the tin reagents employed falls within the range of 0.01 to 5, more preferred 0.1-2.0 M

The catalyst loading based on the total amount of Sn used can be < 5 % and even < 0.1 %. The catalyst is preferably employed in the concentration range $5 \cdot 10^{-4}$-0.1 M.

In one specific example, which employed $PtCl_2(PPh_3)_2$ as catalyst (or catalyst precursor) in toluene at a reaction temperature of 85 °C, the yield of $BuSnCl_3$ was found to be as high as 92 % (based on Sn). $SnCl_2$ was the sole tin-containing by-product.

In another preferred embodiment of the invention the catalyzed redistribution reaction concerns redistribution between R_2SnCl_2 or R_4Sn (R is Me, Et, propyl, hexyl, more preferably Me and *n*-hexyl) and $SnCl_4$ to afford $RSnCl_3$. For R = Me and *n*-hexyl, the monoalkyltin trichloride was obtained starting from R_2SnCl_2 and $SnCl_4$ in 87 and 67 % yield, repectively, whereas in the blank experiment (no catalyst added) only unreacted starting materials were recovered.

The invention is now further demonstrated by the following examples.

Examples

Preparation of $BuSnCl_3$ from Bu_2SnCl_2 and $SnCl_4$.

Bu_2SnCl_2 was either dissolved in the solvent or used as such. $SnCl_4$ was added followed by the catalyst. The reaction mixture was stirred for 12 h at 110°C. Filtration of the reaction mixture gave a slightly yellow solution which was analyzed by ^{119}Sn NMR using $SnMe_4$ as external standard. After evaporation of the solvent, butyltin trichloride was obtained as a colorless liquid by vacuum distillation (90°C/11 mmHg) and analyzed by ^{1}H, $^{13}C\{^{1}H\}$ and $^{119}Sn\{^{1}H\}$ NMR spectroscopy. The solid material obtained from the filtration was washed with toluene (3 × 20 mL) and dried *in vacuo* to give an off-white solid which was indentified as $SnCl_2$ by ^{119}Sn NMR (δ = –235.1 in actone-d_6) and/or elemental analysis. The results of the different experiments are presented in Table 1.

In one typical experiment, 373 g (1.32 mol, 80%) of $BuSnCl_3$ and 44.5 g (0.23 mol, 14%) of $SnCl_2$ were obtained starting from Bu_2SnCl_2 (252 g, 0.83 mol) in toluene (500 mL), $SnCl_4$ (215 g, 0.83 mol) and $PtCl_2(PPh_3)_2$ (215 mg, 0.27 mmol) as catalyst following the above procedure. Elemental analysis of $SnCl_2$: Cl, 37.0%; Sn, 61.23 %. $SnCl_2$ requires: Cl, 37.4%; Sn, 62.60%.

Table 1. Results of the reaction of Bu_2SnCl_2 with $SnCl_4$ (1:1 molar ratio) in the presence of several catalysts.[a]

Entry	Catalyst	T (°C)	[Catalyst] (mol%)[b]	Solvent[c]	Yield of $BuSnCl_3$ (%)[d]	Selectivity (%)[e]
1	$Pd(PPh_3)_4$	110	5	no solvent	60 (25)	n.d.
2	$PdCl_2(PPh_3)_2$	110	1	no solvent	64	n.d.
3	$PdCl_2$(TMEDA)	110	5	no solvent	72 (70)	n.d.
4	$PtCl_2(PPh_3)_2$	110	0.1	no solvent	62	n.d.
5	$PtCl_2(PPh_3)_2$	110	1	toluene	72	n.d.
6	$PtCl_2(PPh_3)_2$	85	0.1	toluene	92[f]	93[f]
7	$PtCl_2(PPh_3)_2$	110	0.1	toluene	84 (80)	87
8	$PtCl_2(PPh_3)_2$	130	0.1	toluene	83	86
9	$PtCl_2(PPh_3)_2$	110	0.03	toluene	n.d. (80)	n.d.
10	$PtCl_2(PPh_3)_2$	110	0.1	*o*-xylene	70	79
11	$PtCl_2(PPh_3)_2$	110	0.1	1,2-dichlorobenzene	85	83
12	$PtCl_2(PPh_3)_2$	110	0.1	*n*-octane	76	89

[a] Reaction time = 12 h unless indicated otherwise. [b] Relative tot Bu_2SnCl_2. [c] $[Bu_2SnCl_2]_0 = [SnCl_4]_0 = 1.0$ M when toluene was used as solvent. [d] Yield based on total amount of Sn, determined by ^{119}Sn NMR analysis of the crude reaction mixture. Isolated yield in brackets. [e] By-product: $SnCl_2$. [f] After $t = 48$ h instead of 12 h. n.d. = not determined

Preparation of $MeSnCl_3$ from Me_2SnCl_2 and $SnCl_4$.

In this typical experiment, 3.40 g (14.1 mmol, 87 %) of $MeSnCl_3$ was obtained starting from Me_2SnCl_2 (1.80 g, 8.3 mmol) in toluene (5 mL), $SnCl_4$ (1.0 mL, 8.3 mmol) and $PtCl_2(PPh_3)_2$ (6.5 mg, 8.2 μmol) as catalyst following the above procedure for preparation of $BuSnCl_3$. After filtration volatiles were removed in vacuo (1 mm Hg) to afford the product as a white solid. No $SnCl_2$ formation was observed.

Preparation of (n-C_6H_{13})$SnCl_3$ from (n-C_6H_{13})$_2SnCl_2$ and $SnCl_4$.

In this typical experiment, 1.15 g (3.70 mmol, 67%) of (n-C_6H_{13})$SnCl_3$ was obtained starting from (n-C_6H_{13})$_2SnCl_2$ (1.0 g, 2.78 mmol) in toluene (10 mL), $SnCl_4$ (0.72 g, 2.78 mmol) and $PtCl_2(PPh_3)_2$ (23 mg, 27.8μmol) as catalyst following the above procedure for preparation of $BuSnCl_3$. After filtration volatiles were removed in vacuo (2 mm Hg) and the remaining liquid was vacuum-transferred (ca 200 °C, 1-2 mm Hg). $SnCl_2$ (0.23 g, 1.2 mmol, 21%) was isolated as side-product.

Preparation of $BuSnCl_3$ from Bu_4Sn and $SnCl_4$.

In this typical procedure, $SnCl_4$ (11.15 g, 0.043 mol) was dissolved in toluene (10 mL). Next, Bu_4Sn (4.96 g, 0.0143 mol) was added and reaction mixture was stirred for 2 h at 110°C. After cooling of the reaction mixture to room temperature, the catalyst $PtCl_2(PPh_3)_2$ (3.8 mg, 4.8 μmol) was added and the reaction mixture was stirred for another 12 h at 110°C. Filtration of the reaction mixture gave a slightly yellow solution which was analyzed by ^{119}Sn NMR using $SnMe_4$ as external standard. After evaporation of volatiles in *vacuo*, vacuum distillation (90°C/11 mmHg) afforded 13.5 g (83%) of butyltin trichloride as a colorless liquid. The solid material obtained from the filtration

was washed twice with toluene (5 mL) and dried *in vacuo* to give 0.40 g of an off-white solid. The solid was dissolved in acetone-d_6 and indentified as $SnCl_2$ by ^{119}Sn NMR ($\delta = -235.1$). Elemental analysis: Cl, 36.92%; Sn, 62.40%. $SnCl_2$ requires: Cl, 37.40%; Sn, 62.60%. The results of the different experiments are presented in Table 2.

Table 2. Results of the reaction of Bu_4Sn with $SnCl_4$ (1:3 molar ratio) in the presence of several catalysts.[a]

Entry	Catalyst	[Catalyst] (mol %)[b]	Solvent[c]	Yield of $BuSnCl_3$ (%)[d]
1	$Pd(PPh_3)_4$	5	toluene	n.d. (47)
3	$PdCl_2(PPh_3)_2$	1	no solvent	n.d. (72)
4	$PtCl_2(PPh_3)_2$	0.1	toluene	77 (75)
5	$PtCl_2(PPh_3)_2$	0.03	toluene	87 (83)
6	-		no solvent	n.d. (63)[e]

[a] Conditions: $T = 110°C$, $t = 12$ h. [b] Relative to Bu_4Sn. [c] $[SnCl_4]_0 = 1.0$ M when toluene was used as solvent. [d] Yield based on Sn, determined by ^{119}Sn NMR analysis of the crude reaction mixture. Isolated yield in brackets. [e] Bu_4Sn : $SnCl_4 = 1:2$. n.d. = not determined

CLAIMS:

1. Process for the production of monoalkyltin trihalides of the formula $RSnX_3$, wherein R = alkyl or cycloalky and X = Cl, Br or I, comprising a redistribution reaction between tetraorganotins, triorganotin halides or diorganotin halides and tin tetrahalides, said process comprising contacting tetra- (R_4Sn), tri- (R_3SnX) or diorganotin halides (R_2SnX_2) with SnX_4 to afford said monoalkyltin trihalides in the presence of at least one transition metal complex, said complex comprising at least one transition metal, M, which is from Group VIII of the periodic Table of elements, at least one ligand which is monodentate or bidentate, and optionally one or more anions, Y, of an organic or inorganic acid, as a catalyst, a catalyst precursor, or a catalyst system.

2. Process according to claim 1, wherein the said complex has the formula

$L'MY_2$, (I)

wherein L' is a bidentate ligand;

L_2MY_2, (II)

wherein L is a monodentate ligand; or

L_4M, (III)

wherein L is a monodentate ligand.

3. Process according to claim 1, wherein the said complex has the formula

$[L"M(\mu\text{-}Y)]_2$ (IV)

wherein L" = a cyclometallated bidentate optionally substituted o-(diarylphosphino)benzyl ligand.

4. Process according to any one of claims 1-3, wherein M is Pt, Pd or Ni.

5. Process according to any one of claims 1-4, wherein Y is Cl, Br, I, acetate, triflate or tosylate anion.

6. Process according to claim 1, wherein the ligand is a phosphine, an alkene, an amine, an organic sulfide, a nitrile , an imidazoline-2-ylidene, a diphosphine, a dialkene, a diamine or a bis(imidazoline-2-ylidene) ligand.

7. Process according to claim 2, wherein L is a phosphine, an alkene, an amine, an organic sulfide, a nitrile or an imidazoline-2-ylidene.

8. Process according to claim 2, wherein L' is a diphosphine, a dialkene, a diamine or a bis(imidazoline-2-ylidene) ligand.

9. Process according to claim 3, wherein L" is an optionally substituted o-{di(2-tolyl)phosphino}benzyl.

10. Process according to claim 2, wherein the catalyst, catalyst precursor or catalyst system is formed by combining (I), (II) or (III) with another ligand which is a phosphine, an alkene, an amine, an organic sulfide, a nitrile or an imidazoline-2-ylidene.

11. Process according to claim 1, wherein the ligand is a triarylphosphine or a bidentate nitrogen ligand.

12. Process according to claim 2, wherein L is a triarylphosphine.

13. Process according to claim 2, wherein L' is a bidentate nitrogen ligand.

14. Process according to any one of claims 1-13, wherein R is an alkyl having 1-12 carbon atoms.

15. Process according to any one of claims 1-14, wherein the reaction is carried out in the presence of a solvent.

16. Process according to any one of claims 1 to 3, wherein M is Pt or Pd.

17. Process according to any one of claims 1 to 16, wherein X is Cl.

18. Process according to any one of claims 1 to 17, wherein the reaction is carried out at a temperature between 0 and 200°C.

19. Process according to any one of claims 1 to 18, wherein the reaction is carried out at a pressure between 1 and 5 bar.

20. Process according to any one of claims 1 to 19, wherein the monoalkyltin trihalide product is obtained in > 60% yield, based on Sn, in less than 48 h at temperatures below 150°C.

21. Use of a transition metal complex comprising at least one transition metal, M, from Group VIII of the periodic Table of elements, at least one monodentate ligand or bidentate ligand, and optionally one or more anions, Y, of an organic or inorganic acid, as a catalyst or catalyst precursor for the production of monoalkyltin trihalides of the formula $RSnX_3$, wherein R = alkyl or cycloalky and X = Cl, Br or I, using a redistribution reaction between tetraorganotins, triorganotin halides or diorganotin halides and tin tetrahalides.

22. The process of claim 11, wherein the ligand is triphenylphosphine or N, N, N',N'-tetramethylethylenediamine (TMEDA).

23. The process of claim 12, wherein L is triphenylphosphine.

24. The process of claim 13, wherein L' is N, N, N',N'-tetramethylethylenediamine (TMEDA).

25. The process of claim 15, wherein said solvent is an aromatic solvent, a chloroaromatic, an alkane, a dialkylacetamide, an *N*-alkylpyrolidone, a dialkylamide of aliphatic carboxylic acid or an organic nitrile.