

Process for the manufacture of dinitrile compounds

The present invention relates to a process for the manufacture of compounds comprising two nitrile functional groups.

It relates more particularly to a process for the manufacture of dinitrile compounds from compounds comprising a nitrile functional group and an ethylenic unsaturation.

Dinitrile compounds, in particular adiponitrile, are important chemical intermediates in the synthesis of numerous compounds. Thus, adiponitrile is used in the manufacture of hexamethylenediamine, one of the monomers of polyamides. It can also be used in the manufacture of aminocapronitrile or caprolactam, monomers important in the production of various polyamides.

Among the processes provided for the synthesis of adiponitrile, the process using butadiene as starting material and the hydrocyanation reaction with hydrogen cyanide is the most extensively used industrially.

This process consists, in a first stage, in carrying out the hydrocyanation of an olefin, such as butadiene, to produce compounds comprising a nitrile functional group and an ethylenic unsaturation. This stage is generally carried out in the presence of a catalytic system comprising an organometallic complex formed by a metal, such as nickel, and an organophosphorus ligand.

After separation of the unsaturated mononitrile compounds and optionally purification, the latter are converted to dinitrile compounds by a second hydrocyanation reaction also carried out in the presence of a catalytic system comprising an organometallic complex formed by a metal, such as nickel, and

an organophosphorus ligand. Furthermore, the catalytic system comprises a cocatalyst generally composed of a Lewis acid.

- 5 The term "Lewis acid" is understood to mean, according to the usual definition, compounds which accept electron pairs. Lewis acids are generally salts of metal elements, as is described below.
- 10 In the current processes, the Lewis acid is maintained in the reaction medium, in particular during the stage of extraction of the nitriles. The Lewis acid is subsequently removed with the distillation bottoms, in particular when it is not separated from the medium in
- 15 conjunction with the organometallic complex used as catalytic system.

The presence of the Lewis acid during the distillation of the dinitrile can promote the generation of

20 impurities in the medium, which impurities may be present in the distilled dinitrile. In addition, the removal of the Lewis acid and its discharge as effluent can be harmful to the economics of the process and to the environment. One of the aims of the present

25 invention is to provide a process for the manufacture of dinitrile compounds which does not comprise these disadvantages.

To this end, the invention provides a process for the

30 manufacture of dinitrile compounds by hydrocyanation with hydrogen cyanide of compounds comprising a nitrile functional group and an ethylenic unsaturation in the presence of a catalytic system comprising an organo-metallic complex and a cocatalyst formed by a metal

35 compound.

According to the invention, the process comprises the following successive stages, after having carried out the hydrocyanation stage:

- I. treating the reaction medium E_1 obtained after the stage of hydrocyanation of the unsaturated nitrile compounds in order to extract at least the organometallic complex from said medium and to obtain a second medium E_2 ;
- II. treating the said second medium E_2 by passing over an ion-exchange resin in order to extract at least the metal forming the cocatalyst and to obtain a third medium E_3 , and
- III. separating the dinitriles formed from the said medium E_3 ,

and in that the order of stages II) and III) can be reversed.

According to a preferred characteristic of the invention, the cocatalyst is a Lewis acid. More specifically, the Lewis acids mentioned in the work edited by G.A. Olah, "Friedel-Crafts and Related Reactions", Volume 1, pages 191 to 197 (1953), are suitable for the invention.

The Lewis acids which can be employed as cocatalysts in the present process are chosen from compounds of the elements from Groups Ib, IIb, IIIa, IIIb, IVa, IVb, Va, Vb, VIb, VIIb and VIII of the Periodic Table of the Elements. These compounds are generally salts, in particular halides, such as chlorides or bromides, sulphates, sulphonates, haloalkylsulphonates, perhaloalkylsulphonates, in particular fluoroalkylsulphonates or perfluoroalkylsulphonates, haloacetates, perhaloacetates, carboxylates and phosphates.

Mention may be made, as non-limiting examples of such Lewis acids, of zinc chloride, zinc bromide, zinc iodide, manganese chloride, manganese bromide, cadmium chloride, cadmium bromide, stannous chloride, stannous bromide, stannous sulphate, stannous tartrate, indium chloride, indium trifluoromethylsulphonate, indium trifluoroacetate, the chlorides or bromides of rare earth elements, such as lanthanum, cerium, praseodymium, neodymium, samarium, europium, gadolinium, terbium, dysprosium, hafnium, erbium, thallium, ytterbium and lutetium, cobalt chloride, ferrous chloride or yttrium chloride.

Use may also be made, as Lewis acid, of compounds such as triphenylborane or titanium isopropoxide.

It is possible, of course, to employ mixtures of several Lewis acids.

Preference is very particularly given, among Lewis acids, to zinc chloride, zinc bromide, stannous chloride, stannous bromide, triphenylborane, indium trifluoromethylsulphonate, indium trifluoroacetate and zinc chloride/stannous chloride mixtures.

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The Lewis acid cocatalyst employed generally represents from 0.01 to 50 mol per mole of transition metal compound, more particularly of nickel compound.

30 According to a preferred embodiment of the invention, stage II) of treatment over ion-exchange resins is carried out before the separation of the dinitrile compounds from the medium E_3 .

35 This is because the separation of the dinitrile compounds is generally carried out by distillation and thus by heating the medium E_3 comprising these compounds. It is advantageous to remove the metal elements present in the medium E_3 before carrying out

such a heating in order to prevent these metal elements from promoting side reactions or decomposition of the dinitriles. These side or decomposition reactions generate by-products with a structure similar to that of the dinitriles which are difficult to separate from the latter.

Thus, in the case of adiponitrile, certain impurities, such as 1-imino-2-cyanocyclopentane (ICCP), can be produced. These impurities are reencountered in part in the distilled adiponitrile. The impurities and their hydrogenation products can also be reencountered in the hexamethylenediamine (HMD) obtained by hydrogenation of this adiponitrile and even in the polyamide obtained from the HMD.

Thus, these impurities can produce defects in the processes for forming these polyamides, in particular in spinning processes (increase in the number of breakages of the yarns) or with regard to the stability and the colour of the polyamide.

According to a preferred characteristic of the invention, the organometallic complex forming the catalytic system is generally a coordination complex between a metal element chosen from transition metals and ligands, generally organophosphorus ligands.

Such organometallic complexes are disclosed in numerous publications and numerous patents, such as US 3 496 215, DE19953058, FR 1 529 134, FR 2 069 411, US 3 631 191, US 3 766 231, FR 2 523 974, WO 99/06355, WO 99/06356, WO 99/06357, WO 99/06358, WO 99/52632, WO 99/65506, WO 99/62855, US 5 693 843, WO 96/1182, WO 96/22968, US 5 981 772, WO 01/36429, WO 99/64155 or WO 02/13964.

The metal elements are generally chosen from the group consisting of nickel, cobalt, iron, ruthenium, rhodium,

palladium, osmium, iridium, platinum, copper, silver, gold, zinc, cadmium and mercury. Among these metals, nickel is the preferred metal.

- 5 In these metal complexes, the metal is encountered in a specific oxidation state, in particular zero for nickel, for example.

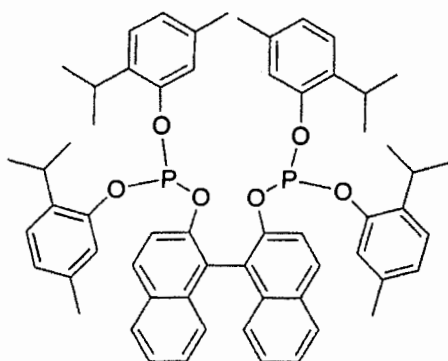
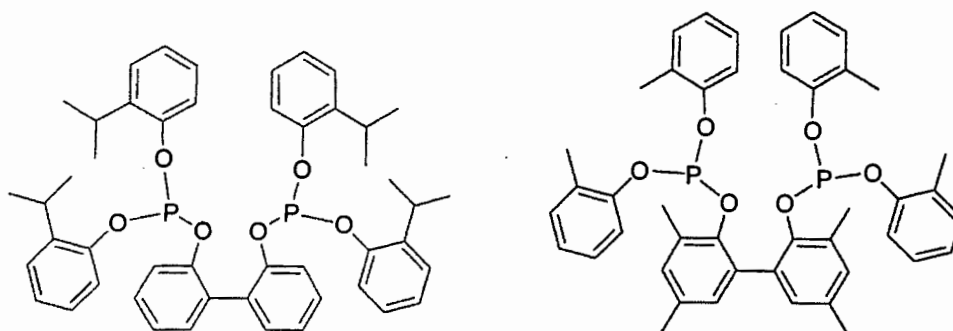
10 During the hydrocyanation reaction, a portion of the metal element (for example nickel) can be oxidized to a higher oxidation state, for example state 1 or 2, and may thus no longer be extractable from the reaction medium E₁ during stage I) of the process of the invention. Its presence in the reaction media E₂ or E₃
15 can result in disadvantages similar to those of the metal elements originating from the cocatalyst.

This disadvantage is overcome by the process of the invention, which makes it possible, according to a
20 preferred characteristic, to coextract these metal elements during the stage II) of treatment over ion-exchange resins by choosing an appropriate resin and appropriate extraction conditions. It is also possible, without departing from the scope of the present
25 invention, to use two or more resins in combination or successively in order to make possible the concomitant or successive extraction of these metal elements.

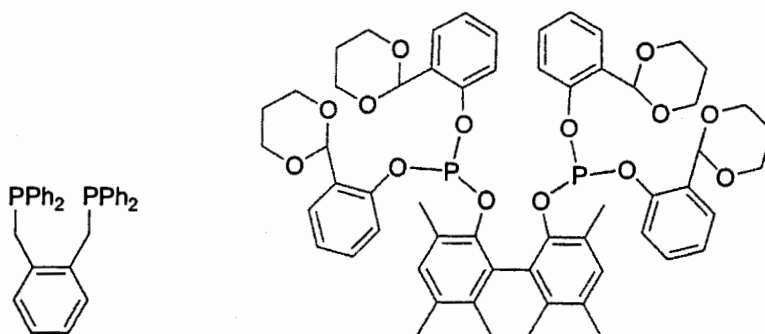
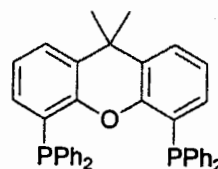
Mention may be made, as organometallic complexes
30 suitable for the invention, of those obtained from nickel compounds and from organophosphorus compounds belonging to the families of the monodentate or polydentate organophosphites, organophosphinites, organophosphonites and organophosphines. It is also possible
35 to use organometallic complexes obtained from stilbene or arsine in combination with nickel or one of the metals mentioned above. Examples of organophosphorus compounds are disclosed in numerous publications and numerous patents. Mention may be made, for example, as

examples of monodentate compounds, of triphenyl phosphite, tritolyl phosphite, trithymyl phosphite, phenyl diphenylphosphinite, tolyl ditolylphosphinite, thymyl dithymylphosphinite, diphenyl phenylphosphonite, 5 ditolyl tolylphosphonite, dithymyl thymylphosphonite, triphenylphosphine, tritolylphosphine or trithymylphosphine.

Mention may be made, as bidentate compounds, by way of 10 examples, of the following structures, in which Ph indicates phenyl:



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15 More generally, all organophosphorus ligands are

suitable for the invention.

Mention may also be made, by way of examples, of the catalytic systems and ligands disclosed in patents
5 WO 95/30680, WO 96/11182, WO 99/06358, WO 99/13983, WO 99/64155, WO 01/21579 and WO 01/21580.

The organometallic complexes can be prepared before their addition to the reaction medium or in situ.

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The organometallic complexes can be prepared by bringing a compound of the chosen metal into contact with a solution of an organophosphorus compound.

15 The compound of the metal can be dissolved in a solvent.

The metal can be present in the compound employed either in the oxidation state which it will have in the organometallic complex or in a higher oxidation state.
20

By way of examples, it may be indicated that, in the organometallic complexes of the invention, rhodium is in the oxidation state (I), ruthenium is in the oxidation state (II), platinum is in the oxidation state (0), palladium is in the oxidation state (0), osmium is in the oxidation state (II), iridium is in the oxidation state (I) and nickel is in the oxidation state (0).
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If, during the preparation of the organometallic complex, the metal is employed in a higher oxidation state, it can be reduced in situ.

35 Mention may be made, as nonlimiting examples of metal compounds used in the preparation of these complexes, of metal powders, such as nickel powder, and the following compounds:

- compounds in which the nickel is in the zero

oxidation state, such as potassium tetracyanonickelate $K_4[Ni(CN)_4]$, bis(acrylonitrile)nickel(0), bis(1,5-cyclooctadiene)nickel (also known as $Ni(cod)_2$) and the derivatives comprising ligands, such as tetrakis-
5 (triphenylphosphine)nickel(0),

- nickel compounds, such as the carboxylates (in particular the acetate), carbonate, bicarbonate, borate, bromide, chloride, citrate, thiocyanate, cyanide, formate, hydroxide, hydrophosphite, phosphate,
10 phosphate and derivatives, iodide, nitrate, sulphate, sulphite, arylsulphonates and alkylsulphonates.

When the nickel compound used corresponds to an oxidation state of the nickel of greater than 0, a
15 reducing agent for the nickel is added to the reaction medium, which reducing agent preferably reacts with the nickel under the conditions of the reaction. This reducing agent can be organic or inorganic. Mention may be made, as nonlimiting examples, of borohydrides, such
20 as $NaBH_4$ or KBH_4 , Zn powder, magnesium or hydrogen.

When the nickel compound used corresponds to the 0 oxidation state of the nickel, a reducing agent of the type of those mentioned above can also be added but
25 this addition is not essential.

When an iron compound is used, the same reducing agents are suitable.

30 In the case of palladium, the reducing agents can, in addition, be components of the reaction medium (phosphine, solvent, olefin).

The cocatalyst or Lewis acid is present in the
35 catalytic system according to an amount of between 0.01 and 50 mol of Lewis acid per mole of metal element of the organometallic complex, such as nickel, for example, and preferably between 0.05 and 10 mol/mol. The Lewis acid can be added directly to the reaction

medium or with the organometallic complex.

The hydrocyanation reaction is generally carried out at a temperature of between 10°C and 200°C, preferably
5 between 30°C and 120°C.

The hydrocyanation reaction can be carried out without solvent but it can be advantageous to add an inert organic solvent. This solvent can be a solvent for the
10 catalytic system which is miscible with the phase or with the medium comprising the compound to be hydrocyanated, at least at the hydrocyanation temperature.

15 This reaction and the process of the invention can be carried out continuously or batchwise.

This reaction can also be carried out in the presence of a two-phase system comprising in particular an
20 aqueous phase in which the organometallic complex is soluble. In this embodiment, the aqueous phase is separated with most of the organometallic complex at the end of the reaction. The organic phase comprising the dinitriles and the unreacted unsaturated nitriles
25 is the medium E₁ within the meaning of the present invention. This is because it is advantageous to treat this organic phase according to the process of the invention in order to extract the small portion of organic complex and of Lewis acid present in the said
30 phase.

The reaction medium E₁ obtained at the outlet of the hydrocyanation reactor is, according to a preferred embodiment of the invention, fed to a stage I) for
35 extraction of the organometallic complex.

This stage can consist of an extraction of the said complex by a solvent in a liquid/liquid extraction plant. Mention may be made, by way of examples, as

extraction solvent, of alkanes comprising from 5 to 9 carbon atoms, such as pentane, hexane or heptane, cycloalkanes comprising from 5 to 8 carbon atoms, such as cyclohexane, methylcyclohexane or cyclooctane, 5 halogenated hydrocarbons comprising from 1 to 5 carbon atoms, such as chloroform, dichloroethane, carbon tetrachloride, chloropropane or dichloromethane, or substituted or unsubstituted aromatic compounds comprising from 6 to 9 carbon atoms, such as benzene, 10 toluene, xylene, ethylbenzene or isopropylbenzene.

This separation can also be brought about by distillation of the unsaturated nitriles and the production of a two-phase medium, one of the phases of 15 which comprises the organometallic complex, the other phase of which comprising mainly the dinitriles. The latter can be subjected to liquid/liquid extraction with the solvents described above in order to extract the traces of organometallic complex.

20 The organometallic complex thus extracted can be recycled in a hydrocyanation reaction.

At the end of stage I), a reaction medium E_2 no longer 25 comprising organometallic complex is obtained. The term "no longer comprises" should be understood as meaning that the maximum amount of the organometallic complex has been extracted but traces of the said complex may remain in the medium without, however, departing from 30 the scope of the invention.

This reaction medium E_2 comprises the dinitriles formed and also the Lewis acid and optionally compounds of the metal element originating from the organometallic 35 complex which may or may not have undergone oxidation.

According to the preferred embodiment of the invention, this reaction medium E_2 is subjected to a treatment over ion-exchange resins which makes it possible to fix and

extract the metal ions of the Lewis acid and optionally the oxidized metal ions which originate from the organometallic complex and which are found in the said medium E₂.

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Suitable ion-exchange resins of the present invention are chosen according to the nature of the metal elements to be extracted. Thus, these resins can be one of those belonging to the group consisting of strong or
10 weak cationic resins, adsorbent resins, chelating resins and catalytic resins. Mention may be made, as examples of resins, of sulphonic resins, carboxylic resins, iminodiacetic resins or resins sold under the tradenames Amberlite and Amberlyst by Rohm & Haas,
15 Dowex by Dow and Levatite and Ionac by Bayer.

This stage II) can be carried out in any device known to a person skilled in the art. Thus, use may be made of columns comprising these resins in the form of a
20 fixed bed or of a fluidized bed or membrane systems formed by the said resin.

After treatment on a resin, a reaction medium E₃ is obtained which comprises essentially the dinitrile
25 organic compounds formed. These various compounds will advantageously be separated in a distillation stage III). However, other separation processes can be used without, however, departing from the scope of the invention.

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Furthermore, the stage II) of treatment with the resins also comprises a stage of elution of the resins laden with the extracted metal elements in order thus to regenerate the resins and to recover these metal
35 elements.

This elution is a conventional and standard stage for processes for treatment on ion-exchange resins. It can be carried out in particular using strong acids, such

as sulphuric acid or hydrochloric acid, or strong organic acids and preferably the acid corresponding to the anion of the Lewis acid to be regenerated.

5 In the context of the present invention, this elution stage can make it possible to recover and regenerate the Lewis acid and can thus make possible its reuse in a hydrocyanation stage. This possibility is of great advantage in making possible economical operation of
10 the process and respect for the environment, in particular when the compound used as Lewis acid is expensive and/or exhibits a toxic nature with respect to the environment.

15 The process of the invention applies in particular to aliphatic nitriles comprising an ethylenic unsaturation and more particularly to linear pentenenitriles, such as 3-pentenenitrile or 4-pentenenitrile.

20 These pentenenitriles can comprise minor amounts of other compounds, such as 2-methyl-3-butenitrile, 2-methyl-2-butenitrile, 2-pentenenitrile, valeronitrile, adiponitrile, 2-methylglutaronitrile or 2-ethylsuccinonitrile. These compounds are present in the
25 pentenenitriles, in particular when the latter originate from a first stage of hydrocyanation of butadiene to give unsaturated mononitriles.

This first stage is generally carried out in the
30 presence of a catalyst comprising an organometallic complex but in the absence of cocatalyst. The organometallic complex used in this first stage can be different from or identical to that used in the process of the invention. In the case where it is identical,
35 the organometallic complex recovered in stage I) of the process of the invention can be recycled in the hydrocyanation reactor for the first stage of hydrocyanation of butadiene.

The mononitriles introduced in the process of the invention can also originate from an isomerization stage, generally in association with the first stage above, which consists in maintaining the compounds
5 obtained in the first stage above in the presence of a catalytic system comprising an organometallic complex, advantageously identical to that of the first stage, in the absence of hydrogen cyanide.

10 This object of this isomerization stage is to improve the selectivity of the process for linear unsaturated mononitriles.

Other advantages and details of the invention will
15 become more clearly apparent in the light of the examples given below solely by way of indication and of illustration.

Example 1:

20

A solution of indium trifluoroacetate (0.5 g, 1.10 mmol) in a mixture of 3-pentenitrile (36.4 g, 448 mmol) and adiponitrile (63.6 g, 588 mmol) is prepared. The composition of the medium is determined
25 by elemental analysis.

4 ml fractions of this solution are brought into contact with 2 ml of each of the following resins sold by Rohm & Haas under the tradenames indicated below:

30

- a sulphonic resin (Amberlite 252H),
- a chelating resin (IRC 748),
- an adsorbent resin (Amberlite XAD7)
- and a cationic resin (IRC 50).

35

Each of the media is stirred at ambient temperature (20°C) for 4 h. The resin is separated by filtration and the composition of the filtrate is determined by elemental analysis.

The compositions of the starting media and of the filtrates are given in Table I below:

		In (ppm)
Composition of the starting medium		1190
Composition of the filtrate	Amberlite 252H Sulphonic resin	Undetectable
	IRC 748 Chelating resin	Undetectable
	Amberlite XAD7 Adsorbent resin	Undetectable
	IRC 50 Cationic resin	Undetectable

5 Example 2:

A glass column is filled with 100 ml of sulphonic resin (Amberlite 252H). A solution of 3-pentenitrile and of adiponitrile (30/70 in moles) comprising indium tri-fluoroacetate with a concentration, expressed as indium
 10 element, of 3300 ppm is fed continuously via the top of the column at a flow rate of 340 g/h. Samples are withdrawn every 10 minutes at the column outlet. The concentration of indium in the samples is measured by elemental analysis.
 15 During the three hours of operation of the separating column, the concentration of resin in the samples was always less than 20 ppm.

Example 3:

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A solution of zinc chloride (0.1 g, 0.73 mmol) in a mixture of 3-pentenitrile (36.4 g, 448 mmol) and of adiponitrile (63.6 g, 588 mmol) is prepared. The composition of the medium is determined by elemental
 25 analysis.

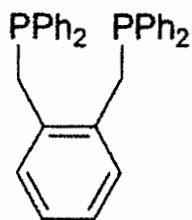
4 ml of this solution are stirred at ambient temperature (20°C) for 4 h in the presence of 2 ml of

sulphonic resin (Amberlite 252H). Analysis of the solution before and after treatment over resin indicates that the concentration of zinc in the solution changes from 480 ppm to 8 ppm.

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Example 4:

The reaction for the hydrocyanation of 3-pentenitrile to give adiponitrile, catalyzed by the Ni(0)/organo-phosphorus ligand/Lewis (In) acid system, is carried
10 out. The organophosphorus ligand is the compound with the following formula:



15 3 ml fractions of this reaction medium are stirred at ambient temperature (approximately 20°C) for 4 hours in the presence of 3 ml of one of the following resins:

- a sulphonic resin (Amberlite 252H),
- a chelating resin (IRC 748),
- 20 - an adsorbent resin (Amberlite XAD7).

The composition of the reaction medium before and after treatment over resin is given in Table II below:

	Resin ref.	%Ni	%In
Starting composition	/	0.459	1.0
Composition after treatment	Amberlite 252H	0.46	<0.02
	Amberlite XAD-7	0.28	0.42
	IRC 748	<0.008	<0.17

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These tests show that it is possible to extract the indium originating from the Lewis acid but also, under

- 17 -

certain conditions, the oxidized nickel originating from the organometallic complex used as catalyst.

Claims

1. Process for the manufacture of dinitrile compounds
by hydrocyanation of mononitrile compounds
comprising an ethylenic unsaturation by reaction
with hydrogen cyanide in the presence of a
catalytic system comprising an organometallic
complex and a cocatalyst, characterized in that it
comprises the following successive stages:
 - I) treating the reaction medium E_1 obtained after
hydrocyanation of unsaturated nitrile
compounds in order to separate the
organometallic complex from the catalytic
system and to obtain a second medium E_2 ;
 - II) treating the said second medium E_2 with an
ion-exchange resin in order at least to
extract the metal ions originating from the
cocatalyst in order to obtain a third medium
 E_3 , and
 - III) separating the dinitriles formed from the
third medium E_3 ,and in that stage III) can be carried out before
stage II).
2. Process according to Claim 1, characterized in
that the cocatalyst is a Lewis acid.
3. Process according to Claim 1 or 2, characterized
in that the metal ions extracted in stage II) are
recovered by elution of the resin.
4. Process according to Claim 3, characterized in
that the metal ions recovered are recycled in
order to form the cocatalyst of the catalytic

system of the hydrocyanation stage.

5. Process according to one of Claims 2 to 4, characterized in that the Lewis acid is a compound of a metal element belonging to Group Ib, IIb, IIIa, IIIb, IVa, IVb, Va, Vb, VIb, VIIb or VIII of the Periodic Table of the Elements.
5
6. Process according to one of Claims 2 to 5, characterized in that the Lewis acid is chosen from the group consisting of halides, such as chlorides or bromides, sulphates, sulphonates, haloalkylsulphonates, perhaloalkylsulphonates, in particular fluoroalkylsulphonates or perfluoroalkylsulphonates, haloacetates, perhaloacetates, carboxylates and phosphates of the metal elements.
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15
7. Process according to one of Claims 2 to 6, characterized in that the Lewis acid is chosen from the group consisting of zinc chloride, zinc bromide, zinc iodide, manganese chloride, manganese bromide, cadmium chloride, cadmium bromide, stannous chloride, stannous bromide, stannous sulphate, stannous tartrate, indium chloride, indium trifluoromethylsulphonate, indium trifluoroacetate, the chlorides or bromides of rare earth elements, such as lanthanum, cerium, praseodymium, neodymium, samarium, europium, gadolinium, terbium, dysprosium, hafnium, erbium, thallium, ytterbium and lutetium, cobalt chloride, ferrous chloride and yttrium chloride.
20
25
30
8. Process according to one of the preceding claims, characterized in that the resin for extraction of the metal ions used in stage II) is chosen from the group consisting of strong or weak cationic resins, adsorbent resins, chelating resins and catalytic resins.
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9. Process according to Claim 8, characterized in that the ion-exchange resin is chosen from the group consisting of sulphonic resins, carboxylic resins and iminodiacetic resins.
- 5
10. Process according to one of the preceding claims, characterized in that the extraction of the organometallic complex in stage I) is carried out by liquid/liquid extraction with an extraction
10 solvent chosen from the group consisting of alkanes comprising from 5 to 9 carbon atoms, cycloalkanes comprising from 5 to 8 carbon atoms, halogenated hydrocarbons comprising from 1 to 5 carbon atoms and substituted or unsubstituted
15 aromatic compounds comprising from 6 to 9 carbon atoms.
11. Process according to one of the preceding claims, characterized in that the separation of the
20 dinitriles in stage III) is carried out by distillation of the said dinitriles.
12. Process according to one of the preceding claims, characterized in that the unsaturated mononitrile
25 compounds are pentenenitriles.
13. Process according to Claim 12, characterized in that the pentenenitriles are obtained by hydrocyanation of butadiene.

Process for the manufacture of dinitrile compounds

The present invention relates to a process for the manufacture of compounds comprising two nitrile functional groups.

It relates more particularly to a process for the manufacture of dinitrile compounds from compounds comprising a nitrile functional group and an ethylenic unsaturation in the presence of a catalytic system comprising an organometallic complex and a cocatalyst of the Lewis acid type. The process of the invention comprises a stage of treatment of the reaction medium resulting from the hydrocyanation which makes it possible to extract and recover the metal element of the Lewis acid.