

(CONVENTION, By one or more persons and/or a Company.)

623711 Form 4.

COMMONWEALTH OF AUSTRALIA

Patents Act 1952-1969

CONVENTION APPLICATION FOR A PATENT

(1) Here insert (in full) Name or Names of Applicant or Applicants, followed by Address(es).

X (1) EXXON CHEMICAL PATENTS INC.
We OF Florham Park, New Jersey, United States of America

(2) Here insert Title of Invention.

hereby apply for the grant of a Patent for an invention entitled: (2) MINIMISING CATALYST LOSS IN THE PRODUCTION OF ALCOHOLS

(3) Here insert number(s) of basic application(s).

which is described in the accompanying complete specification. This applications is a Covention application and is based on the application numbered (3) 8828695.0

(4) Here insert Name of basic Country or Countries, and basic date or dates.

for a patent or similar protection made in (4) United Kingdom on 8th December 1988

My Our address for service is WATERMARK PATENT & TRADEMARK ATTORNEYS 290 Burwood Road, Hawthorn, Victoria, Australia.

DATED this 8th day of December 1989

(6) Signature(s) of Applicant(s) or Seal of Company and Signatures of its Officers as prescribed by its Articles of Association.

14466 081289

(5) EXXON CHEMICAL PATENTS INC. by D. B. Mischlewski Registered Patent Attorney

To: THE COMMISSIONER OF PATENTS.

COMMONWEALTH OF AUSTRALIA

Patents Act 1952-1969

DECLARATION IN SUPPORT OF A CONVENTION APPLICATION FOR A PATENT OR PATENT OF ADDITION

(1) Here insert (in full) Name of Company.

In support of the Convention Application made by(1) EXXON CHEMICAL PATENTS INC.

(hereinafter referred to as the applicant) for a Patent

(2) Here insert title of Invention.

for an invention entitled:(2)

'MINIMISING CATALYST LOSS IN THE PRODUCTION OF ALCOHOLS'

(3) Here insert full Name and Address of Company official authorized to make declaration.

I, (3) ROBERT FRANK NORTHOVER of EXXON CHEMICAL LIMITED, EXXON CHEMICAL TECHNOLOGY CENTRE, PO BOX 1, ABINGDON, OXFORDSHIRE, OX13 6BB, UNITED KINGDOM.

do solemnly and sincerely declare as follows:

1. I am authorised by the applicant for the patent to make this declaration on its behalf.

2. The basic application as defined by Section 141 of the Act was made in(4) THE UNITED KINGDOM

(4) Here insert basic Country or Countries followed by date or dates and basic Applicant or Applicants.

on the Eighth day of December 1988, by EXXON CHEMICAL PATENTS INC.

on the day of 19 by

(5) Here insert (in full) Name and Address of Actual Inventor or Inventors.

- (5) VAN DRIESSCHE, Eddy - Krekelmuitstraat 8, 9900-Eekloo, België
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is/are the actual inventors of the invention and the facts upon which the applicant is entitled to make the application are as follow:

The applicant is the assignee of the said Eddy Van Driessche; Raf Caers; Matthew Derk Olijve; Nicolaas Anthony De Munck; Jean Alexandre Andre Hanin; Arie Van Vliet - by virtue of their contracts of employment

4. The basic application referred to in paragraph 2 of this Declaration was the first application made in a Convention country in respect of the invention the subject of the application.

DECLARED at Oxfordshire, England. this Twenty-Sixth day of January 1990

(6) Signature.

(6)

Handwritten signature

To: THE COMMISSIONER OF PATENTS.

(12) PATENT ABRIDGMENT (11) Document No. AU-B-46050/89
(19) AUSTRALIAN PATENT OFFICE (10) Acceptance No. 623711

- (54) Title
MINIMISING CATALYST LOSS IN THE PRODUCTION OF ALCOHOLS
- International Patent Classification(s)
(51)⁴ C07C 029/16 C07C 031/00 C07C 031/02
- (21) Application No. : 46050/89 (22) Application Date : 08.12.89
- (30) Priority Data
- (31) Number (32) Date (33) Country
8828695 08.12.88 GB UNITED KINGDOM
- (43) Publication Date : 14.06.90
- (44) Publication Date of Accepted Application : 21.05.92
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- (56) Prior Art Documents
FR 2480276
US 4041082
US 4041057

- (57) The invention leads to an acidic waste water stream of reduced volume containing cobalt species generally below 20 ppm Co, and does not require further expensive end-of-pipe treatment for cobalt removal. Furthermore the invention produces cobalt solids, which can be recycled back to the oxonation section via the catalyst makeup step.

The present invention therefore provides a process for the recovery of cobalt from the oil/water mixed reaction product of cobalt catalysed oxonation by increasing the carbon dioxide level by leaving 1 to 3 volume % carbon dioxide in the fresh synthesis gas fed to oxonation and/or recycling hydrogen containing 10-15 volume % carbon dioxide to the oxonation section to enhance the carbon dioxide level in the oil water phase to promote the formation of cobalt carbonate and allowing the cobalt carbonate to settle for removal.

The process is applicable to the production of alcohols from any olefines which may be subjected to hydroformylation, but is particularly suited to the hydroformylation of C₄ to C₁₆, preferably C₄ to C₁₂ olefines for the production of C₅ to C₁₃ alcohols.

CLAIM

1. A process for the recovery of cobalt from the oil/water mixed reaction product of cobalt catalysed oxonation by increasing the carbon dioxide level by leaving 1 to 3 volume % carbon dioxide in the fresh synthesis gas fed to oxonation and/or recycling hydrogen containing 10-15 volume % carbon dioxide to the oxonation section to enhance the carbon dioxide level in the oil water phase to promote the formation of cobalt carbonate and allowing the cobalt carbonate to settle for removal.

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2. A process according to claim 1 in which the product of oxonation is that of hydroformylation of C₄ to C₁₆ branched or linear olefines.
3. A process according to claim 1 or claim 2 in which the settled cobalt carbonate is recycled for use in the oxonation catalyst.

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COMMONWEALTH OF AUSTRALIA
PATENTS ACT 1952-69

COMPLETE SPECIFICATION

(ORIGINAL)

Class

Int. Class

Application Number: 46050/89
Lodged: 8th December 1989

Complete Specification Lodged:
Accepted:
Published:

Priority :

Related Art :

Name of Applicant : EXXON CHEMICAL PATENTS INC.

Address of Applicant : Florham Park, New Jersey, United States of America

Actual Inventor : EDDY VAN DRIESSCHE, RAF CAERS, MATTHEW DERK OLIJVE, NICOLAAS ANTHONY DE MUNCK, JEAN ALEXANDRE ANDRE HANIN and ARIE VAN VLIET

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290 Burwood Road, Hawthorn, Victoria, Australia

Complete Specification for the invention entitled:

MINIMISING CATALYST LOSS IN THE PRODUCTION OF ALCOHOLS

The following statement is a full description of this invention, including the best method of performing it known to :- US

MINIMISING CATALYST LOSS IN
THE PRODUCTION OF ALCOHOLS

This invention relates to the production of alcohols by hydroformylation processes and in particular the production of alcohols with cobalt catalysed hydroformylation in which the amount of waste cobalt is reduced. This brings an economic benefit and also reduces the amount of cobalt in the waste water stream leading to significant environmental benefits.

The hydroformylation process, in general terms, is a process involving the preparation of oxygenated organic compounds by the reaction of carbon monoxide and hydrogen (synthesis gas) with carbon compounds containing olefinic unsaturation. The reaction is performed under hydroformylation conditions in the presence of a carbonylation catalyst or catalyst precursor such as dicobalt octacarbonyl, and results in the formation of a compound, e.g., an aldehyde which has one more carbon atom in its molecular structure than the feedstock. Subsequent hydrogenation of the primary product leads to higher alcohols which may be used for example for conversion into plasticisers.

Typically in higher alcohol production the feedstock for a hydroformylation process is a commercial C₄-C₁₂ olefine fraction and the desired end product is the respective C₅-C₁₃ saturated alcohol or derived mixed alcohol product, produced by hydrogenation of the aldehyde oxonation product. By virtue of the nature of the feedstock commonly available to industry, and indeed of the catalyst and reaction parameters employed, the oxonation reaction yields a range of products due to the numerous secondary reactions which take place. The main commercial products of the hydroformylation reaction are alde-

hydes and alcohols, with side reactions in the oxonation, demetalling and hydrogenation sections of the process system producing some 5 to 20 wt.% of high boiling materials by condensation, esterification and dehydration reactions.

In a conventional higher oxo alcohol process, the feedstock as described above is fed together with synthesis gas into an oxonation unit where catalytic hydroformylation takes place using, e.g., hydro cobaltcarbonyl as the active catalyst species.

After oxonation the product goes through a hydrogenation step to convert aldehydes into alcohols.

The product mixture after hydrogenation comprising the higher alcohol, the high boiling materials mentioned above and a low boiling fraction is then passed to a distillation unit where low boiling materials, high boiling materials and the desired alcohol product are separated. The low boiling material passing off overhead is a low value product, typically containing unreacted olefine feed and paraffins. The high boiling material usually contains dimers such as ethers and ether-alcohols, (e.g., C₂₀ compounds in C₁₀ alcohol production) and trimers such as acetals, (e.g., C₃₀ compounds in C₁₀ alcohol production), and heavier.

Although substantially alcohol free (apart from the heavy ether alcohols), it may contain a minor amount of alcohol which has not been removed in the distillation stage where the higher alcohol product of the hydroformylation process is separated. In our European patent publication 0183545 we describe a process for upgrading these heavy fractions to more useful alcohol.

In the production of higher alcohols cobalt catalyst is used for the reaction of the olefins with synthesis gas. After completion of the oxonation reaction the cobalt catalyst has to be removed from the reaction products and is recycled back to the feed for the oxonation reaction section.

Several technologies to remove the cobalt catalyst from the oxonation reaction products are being practised. Examples are the oxidation of the cobalt catalyst with air/acetic acid to form cobalt acetate, thermal decobalting to form cobalt metal or treatment with dilute caustic to produce sodium cobaltcarbonyl. The last method is called the Kuhlmann catalyst cycle technology and involves two main process steps: first recovery of the sodium cobaltcarbonyl and second the regeneration of the hydro cobaltcarbonyl.

The first step of this Kuhlmann Cycle consists of high pressure decobalting in which oil soluble hydro cobaltcarbonyl is converted into the water soluble sodium cobaltcarbonyl. This is typically done at high temperature (100-180°C) and high pressure (180-300 barg) by thoroughly mixing the oxonation products with a dilute caustic solution. After cooling and depressuring the sodium cobaltcarbonyl water is separated from the oil, and after washing the oil with water for removal of cobalt traces, both water phases are combined and stored.

In the second step, the water soluble sodium cobaltcarbonyl is converted back into the oxonation catalyst hydro cobaltcarbonyl by acidification of the cobalt water with dilute sulphuric acid. The volatile hydro cobaltcarbonyl is stripped from the water by a countercurrent flow of absorbing gas, frequently syngas, which is subsequently passed through an absorber column to recover the hydro cobaltcarbonyl from the stripping gas.

In the first step of this decobalting, the reaction of hydro cobaltcarbonyl with caustic is the main reaction. However, a minor part of the cobalt catalyst is converted into cobalt (II) salts which cannot be recycled back to the olefin feed stream and is lost via the acidic waste water stream into the environment. In the conventional Kuhlmann technology, the possibilities of excessive precipitation of these cobalt salts are deliberately minimised by maintaining a low partial pressure of carbon dioxide in the oxo off-gas streams and by avoiding large excesses of caustic. This avoids solids formation and plugging of the process piping and control valves. Furthermore, by avoiding major water holdups and maximising fluid velocities, solids settling is prevented. This leaves the cobalt salts mainly dissolved in the water stream, which is then acidified and stripped to produce a cobalt containing acidic waste water stream. Typically the waste water contains 60-200 ppm of cobalt.

In a typical industrial process, cobalt losses via this waste water stream are significant (15-30 kg/Co/day), and cause environmental problems. An end-of-pipe treatment process, which changes the pH of the water from acidic to alkaline by caustic addition, has been developed so that the cobalt hydroxide is formed and is removed from the water by flocculation and centrifuging. This process, although effective, is very costly, it requires significant investment, high operating cost and a lot of operator attention.

We have now developed a process which effectively removes cobalt salts, such as cobalt carbonate and cobalt hydroxide, from the aqueous sodium cobaltcarbonyl stream prior to contacting with sulphuric acid and stripping, without incurring additional cobalt losses or experiencing operating problems. This is accomplished by precipitation of
5 solid cobalt carbonate with carbon dioxide which is then settled and filtered from the sodium cobaltcarbonyl water stream.

The invention leads to an acidic waste water stream of reduced volume containing cobalt species generally below 20 ppm Co, and does not require further expensive end-of-pipe treatment for cobalt removal. Furthermore the invention produces cobalt solids,
10 which can be recycled back to the oxonation section via the catalyst makeup step.

The present invention therefore provides a process for the recovery of cobalt from the oil/water mixed reaction product of cobalt catalysed oxonation by increasing the carbon dioxide level by leaving 1 to 3 volume % carbon dioxide in the fresh synthesis gas fed to oxonation and/or recycling hydrogen containing 10-15 volume % carbon dioxide to
15 the oxonation section to enhance the carbon dioxide level in the oil water phase to promote the formation of cobalt carbonate and allowing the cobalt carbonate to settle for removal.

The process is applicable to the production of alcohols from any olefines which may be subjected to hydroformylation, but is particularly suited to the hydroformylation of C₄ to C₁₆, preferably C₄ to C₁₂ olefines for the production of C₅ to C₁₃ alcohols.
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Conventional hydroformylation conditions may be used in the process of this invention and the operating temperatures, pressures and other conditions, such as synthesis gas composition, may be controlled in accordance with the usual expertise of the person skilled in the art to maximise yield of the desired higher alcohol product. For example, the hydroformylation reaction may be carried out at a pressure of 150-300 atm, and a temperature of from 120°C -190° C.

The catalyst may be used in desired active form for example in a concentration of from 0.05-3 wt.% preferably 0.05 and 1 wt.% as metal based on the olefinic feed. Typically the synthesis gas used might have a H₂:CO volume ratio in the range 0.9:1-1.5:1.

The cobalt separation technique of the invention may conveniently be divided into four subsections: (i) Precipitation of cobalt salts (ii) Settling of cobalt salts (iii) Physical separation of the cobalt salts (iv) Recycle of recovered cobalt salts. Cobalt salts are defined as a mixture of mainly cobalt (II) carbonate, cobalt (II) hydroxide and a minor amount of cobalt metal.

The process is illustrated by reference to Figure 1 in which oxo product and dilute caustic containing sodium cobaltcarbonyl are coming from the decobalting section and flow into low pressure gas-liquid separator 1. The liquids from separator 1 flow into water-oxo product separator 2. The separated oxo product is mixed with recycled wash water from tower 6 and sent to a second water-oxo product separator 7.

The oxo product from separator 7 is fed to wash tower 6, while the water phase is combined with the one from separator 3 and sent to settlers 3 and 4.

In wash tower 6 the oxo product is water washed and after separation sent to hydrogenation. The wash water from tower 6 bottoms is partly recycled over tower 6 itself, partly sent

to separator 7, while the remaining goes to settlers 3 and 4.

In settlers 3 and 4 entrained oxo product and solids are separated from the sodium cobalt carbonyl water, which is pumped through cobalt solids filter 8 to stripper column 5. The cobalt solids which build up in filter 8 which can be a solid/liquid separator, a 5 centrifuge, a cyclone, or a membrane are regularly backflushed into settlers 3 and 4.

In stripper column 5 acid is added to the sodium cobaltcarbonyl water and is countercurrently stripped with gas. This stripgas containing hydro cobaltcarbonyl is contacted with oxo feed to recycle the cobalt catalyst back to oxonation. The stripped water from column 5 is sent to conventional end-of-pipe treatment or to a sewer.

10 Details of the cobalt separation technique:

(i) Precipitation of Cobalt Salt

15 The presence of carbon dioxide in the high pressure oxo offgas promotes the formation of cobaltcarbonate. This is accomplished by leaving 1-3 volume % carbon dioxide in the fresh synthesis gas as opposed to earlier processes when carbon dioxide has been deliberately removed from the syn-gas feed, the carbon dioxide level may be further enhanced by recycling hydrogen containing 10-15 volume % carbon dioxide to the oxonation section. We have found that contrary to the general belief, the presence of excess carbon dioxide is not harmful to the oxonation process and does not create additional cobalt losses. To facilitate the formation of cobalt hydroxide, an excess of caustic may be injected into the decobalting section in order to reach a pH of 8-8.5 in the water leaving the decobalting section.

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An alternative is to use fresh synthesis gas free of carbon dioxide, and to recycle hydrogen containing 10-15 volume % carbon dioxide to the oxonation section.

Cobalt carbonate is less water soluble than cobalt hydroxide, therefore the presence of carbon dioxide in the high pressure oxo offgas is more effective in removing water soluble cobalt (II) species than the addition of excessive amounts of caustic.

To maximise the cobaltcarbonyl recovery the oxo product is washed with a water stream in a wash tower 6. Carbon dioxide may then be injected into the water stream preferably as it leaves the wash tower and passing to the cobalt water storage drums 3 and 4 to further promote the formation of cobalt carbonate. A flocculant, for instance a poly methyl methacrylate, may be injected to facilitate the cobalt carbonate particle growth and the settling in the cobalt water storage drums. Another method of stimulating cobalt carbonate particle growth is the recycle of wash tower 6 bottoms water back to the decobalters or to the product settlers 2 and 7, since the cobalt species present in this recycle stream enhance the formation of larger cobalt carbonate particles.

ii. Settling of Cobalt Salts

The addition of a flocculant to sodium cobaltcarbonyl water steams helps the settling of the cobalt carbonate and is not harmful to the subsequent volatilization step in stripper column 5. The flocculant does not cause foaming or other process problems during the stripping of the acidified water stream.

After the formation of the cobalt salts as much as possible of the solids shall be allowed to settle from the sodium cobalt carbonyl water stream prior to pumping it to the stripper column 5. This may be accomplished by leading the water from the product separation step to the two parallel and/or run in series settlers 3 and 4, which also act as sodium cobalt carbonyl water storage drums. These two drums are designed for maximum settling performance by creating a residence time of 5-10 hours. Internals, such as floating suctions, may be installed to prevent cobalt particles entraining from the drums and to avoid stirring up of the already settled particles. Another important aspect of maximising settling times is the reduction of the water flows entering the settlers. This may be achieved by

minimising the net water flow going to the wash column 6, which also proceeds via partly recycling over wash column 6 itself. The wash bottoms stream is also used for prewashing the oxo product going to the oxo product separator 7.

iii. Physical Separation of Cobalt Salts

The settled water containing sodium cobaltcarbonyl is pumped from settlers 3 and 4 to the volatilization column 5 to liberate hydro cobaltcarbonyl. To minimise the risk of cobalt salts entering this tower, the water stream is preferably first passed through filter 8 to remove any entrained solids. The flush water from the filter is sent back to the settler drums. An important aspect of filter operation is the stability of the water flow to the volatilization tower 5, which is not affected by the filter operation.

An alternate way of removing entrained cobalt particles is to pass the water stream via a centrifuge, while avoiding air contact to prevent oxidation of the cobalt carbonyls. The addition of a flocculant is required for centrifuging, but does not harm the volatilization operation.

iv. Recycle of Cobalt Sludge

The cobalt salts, which settle out in the drums 3 and 4, build up as sludge and require periodical removal. During this removal the watery sludge is centrifuged or filtered to concentrate the sludge, the water with sodium cobalt carbonyl is fed back to the process.

The recovered cobalt salts, mainly cobalt carbonate, can be recycled back to the oxonation process as make up catalyst. This is done via the conventional process route by returning it to the cobalt supplier, who reworks it into cobalt oxide. Another method of recycling is to first wash the sludge with a carbon dioxide and/or carbonate containing water stream to remove sodium salts from the sludge and then pump it to the so-called preforming reactors to convert cobalt carbonate into hydro cobaltcarbonyl. Normally these reactors preform cobalt oxide into hydro cobaltcarbonyl. The recycle of the cobalt sludge would not be possible in the conventional process, since caustic treatment of the acidic waste water stream leads to the formation of a mixture of cobalt hydroxide/sulphate, which is very difficult to convert into hydro cobaltcarbonyl.

The process described above has led to a significant reduction of the cobalt losses into the acidic waste water stream. Historically they have been in the order of 15-30kg Co/day (based on a monthly average), while after implementation of the invention they have been reduced to below 7-8kg Co/day. In the meantime the average cobalt circulation rate has been increased from 30 kg Co/hr to 100kg Co/hr, which would otherwise have caused a significant increase of cobalt losses

Example 1

In this example the role of carbon dioxide on the cobalt removal process is demonstrated.

At the start of the experiment, the oxonation unit was operated with a synthesis gas stream free of carbon dioxide, while no hydrogen containing 10-15 volume % carbon dioxide was recycled to the oxo section. A fairly high proportion of the cobalt (II) remaining dissolved in the liquid going via filter 8 to stripping column 5. Although cobalt circulation rate was low (30 kg/hr) typically 10 kg Co/day were lost to the sewer.

By addition of carbon dioxide to the fresh synthesis gas (about 2 volume %), cobalt losses dropped to 7 kg Co/day. Start-up of the recycle compressor and feeding hydrogen/carbon dioxide reduced the losses even further down to 6 kg Co/day.

In another comparative example, the fresh syngas was left free of carbon dioxide, but by recycling hydrogen/carbon dioxide the cobalt losses were reduced in the same manner down to 6 kg Co/day.

Example 2

This example shows the effectiveness of the filter 8 in removing solids from the water stream which is fed to stripping column 5.

Using the process of Example 1, liquid was pumped from drum 4 to column 5, while by-passing filter 8. Under these circumstances, a constant 10-12 kg Co/day was lost to the environment via the sewer. These losses could not be attributed to malperformance of stripping column 5.

Without changing any of the operating parameters, filter 8 was taken in service with the backflush liquid of the filter 8 going to drum 3. In the improved operation losses decreased to an average of 6-8 kg Co/day, which demonstrates that the filter is capable of removing 4-6 kg/day cobalt solids from the aqueous feed going to column 5.

At a lower cobalt circulation than above, the same test was repeated. Without the filter, 5 kg Co/day emitted via the discharge of column 5, startup of the filter reduced the losses down to 2-3 kg Co/day.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A process for the recovery of cobalt from the oil/water mixed reaction product of cobalt catalysed oxonation by increasing the carbon dioxide level by leaving 1 to 3 volume % carbon dioxide in the fresh synthesis gas fed to oxonation and/or recycling hydrogen containing 10-15 volume % carbon dioxide to the oxonation section to enhance the carbon dioxide level in the oil water phase to promote the formation of cobalt carbonate and allowing the cobalt carbonate to settle for removal.
2. A process according to claim 1 in which the product of oxonation is that of hydroformylation of C_4 to C_{16} branched or linear olefines.
3. A process according to claim 1 or claim 2 in which the settled cobalt carbonate is recycled for use in the oxonation catalyst.
4. A process according to any one of the preceding claims in which the pH of the water leaving the decobalting section is adjusted to 8-8.5.
5. A process according to any one of the preceding claims in which the product of oxonation is washed with water and carbon dioxide injected into the wash water.
6. A process according to any one of the preceding claims in which a flocculant is added to promote the settling of the cobalt carbonate.
7. A process according to any one of the preceding claims in which the water from the product separation is fed to settling drums.
8. A process according to any one of the preceding claims in which after separation of the settled cobalt salts the water is passed to volatilisation to liberate hydro cobaltcarbonyl for subsequent catalyst use.
9. A process according to any one of the preceding claims in which after separation of the settled cobalt salt the water is filtered.



10. A process according to claim 9 in which the filter is a solid/liquid separator, a centrifuge, a cyclone, or a memberane.

11. A process according to any one of the preceding claims in which the settled cobalt salts are recycled to the oxonation process as make up catalyst.

12. A process according to claim 11 in which the sludge of settled cobalt salts is washed with a water stream containing carbon dioxide and/or carbonates.

DATED this 19th day of February, 1991.

EXXON CHEMICAL PATENTS INC.

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