COMMONWEALTH OF AUSTRALIA

Patents Act 1952

APPLICATION FOR A STANDARD PATENT OR A STANDARD PATENT OF ADDITION

Form 1
Regulation 9

608429

I/WE, MITSUBISHI JUKOGYO KABUSHIKI KAISHA,

of 5-1, Marunouchi 2-chome, Chiyoda-ku, Tokyo JAPAN

hereby apply for the grant of a standard patent ~~XXXXXXXXXX~~ for an invention entitled "METHOD FOR PRODUCING A CATALYST FOR OXIDIZING SULFUROUS ACID GAS"

which is described in the accompanying ~~provisional~~ complete specification.

**(To be included in the case of a Convention application)*

Details of basic application(s) –

Number of basic application 78212/1988

Name of Convention country in which basic application was filed JAPAN

Date of basic application April 1 1988

**(To be included in the case of an application made by virtue of section 51)*

Number of original application APPLICATION ACCEPTED AND AMENDMENTS

Person by whom made ALLOWED 10-1-91

**(To be included in the case of an application for a patent of addition)*

I request that the patent may be granted as a patent of addition to the patent applied for on Application No. ..... Patent No. .....

In the name of .....

I request that the term of the patent of addition be the same as that for the main invention or so much of the patent for the main invention as is unexpired.

My address for service is KELVIN LORD AND COMPANY, Patent & Trade Mark Attorneys, of 4 Douro Place, West Perth, Western Australia, Australia, 6005.

PO0161 Dated this 31/03/89 day of MARCH 19 89

AUSTRALIAN PATENT OFFICE LS

To :

THE COMMISSIONER OF PATENTS

(Signature)
MITSUBISHI JUKOGYO KABUSHIKI KAISHA
By their Patent Attorneys
KELVIN LORD AND COMPANY

This form must be accompanied by either a provisional specification (Form 9 and true copy) or by a complete specification (Form 10 and true copy).

* These sections are to be completed only where applicable..

C. J. THOMPSON, Commonwealth Government Printer

Australia Patent Declaration Form

Forms 7 and 8

AUSTRALIA

Patents Act 1952

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

Name(s) of Applicant(s)

In support of the application made by MITSUBISHI JUKOGYO KABUSHIKI KAISHA

Title

for a patent for an invention entitled METHOD FOR PRODUCING A PARALLEL GAS FLOW TYPE CATALYST FOR OXIDIZING SULFUROUS ACID GAS

Name(s) and address(es) of person(s) making declaration

I/We Shoji UEDA, Managing Director of MITSUBISHI JUKOGYO KABUSHIKI KAISHA, 5-1, Marunouchi 2-chome, Chiyoda-ku, Tokyo, Japan

do solemnly and sincerely declare as follows:-

1. I am/we are the applicant(s) for the patent, or am/are authorised by the abovementioned applicant to make this declaration on its behalf.

2. The basic application(s) as defined by Section 141 of the Act was/were made in the following country or countries on the following date(s) by the following applicant(s) namely:-

Country, filing date and name of Applicant(s) for the or each basic application

in Japan on April 1, 1988
by MITSUBISHI JUKOGYO KABUSHIKI KAISHA
in _____ on _____ 19__
by _____

3. The said basic application(s) was/were the first application(s) made in a Convention country in respect of the invention the subject of the application.

Name(s) and address(es) of the or each actual inventor

4. The actual inventor(s) of the said invention is/are Naohiko UKAWA, Kozo IIDA and Tsumoru NAKAMURA of c/o Hiroshima Technical Institute, MITSUBISHI JUKOGYO K.K., 6-22, Kanonshin-machi 4-chome, Nishi-ku, Hiroshima, Hiroshima Pref., Japan (all of them)

See reverse side of this form for guidance in completing this part

5. The facts upon which the applicant(s) is/are entitled to make this application are as follows:-

The applicant is the assignee of the actual inventors.

DECLARED at Tokyo, Japan this 14th day of April, 1989

MITSUBISHI JUKOGYO KABUSHIKI KAISHA

(12) PATENT ABRIDGMENT (11) Document No. AU-B-32298/89
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(54) Title
METHOD FOR PRODUCING A CATALYST FOR OXIDIZING SULFUROUS ACID GAS

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(56) Prior Art Documents
EP 82831
EP 19174

(57) Claim

1. A method for producing a catalyst for oxidizing sulfurous acid gas which uses vanadium pentaoxide as a main catalyst, potassium salts as subsidiary catalysts and diatomaceous earth as a carrier, said method is characterized in that after an ingredient mixture of a solution containing vanadium pentaoxide and potassium salts, diatom earth, water and a binder containing sodium polyacrylate is kneaded, the mixing ratio of these materials being 20-30 weight % of the solution containing vanadium pentaoxide and potassium salts (ph 9-12), 35-50 weight % of diatom earth, 10-25 weight % of water, and 10-20 weight % of the binder containing sodium polyacrylate, said ingredient mixture is molded by extrusion into a shape in which gas passage holes are arranged in a honeycomb or grid structure.

PATENTS ACT 1952

COMPLETE SPECIFICATION

(ORIGINAL)

608429

FOR OFFICE USE:

Application Number:
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Complete Specification Lodged:
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Name of Applicant(s): MITSIBUSHI JUKOGYO KABUSHIKI KAISHA

This document contains the amendments made under Section 49 and is correct for printing

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Complete Specification for the invention entitled:

"METHOD FOR PRODUCING A
CATALYST FOR OXIDIZING SULFUROUS ACID GAS"

AUSTRALIAN PATENT OFFICE LS

The following statement is a full description of this invention, including the best method of performing it known to me/us

SPECIFICATION

1. TITLE OF THE INVENTION

Method for Producing a ~~Parallel Gas Flow Type~~ Catalyst for Oxidizing Sulfurous Acid Gas

2. FIELD OF THE INVENTION AND RELATED ART STATEMENT

The present invention relates to a method for producing a catalyst for oxidizing sulfurous acid gas in a plant producing sulfuric acid and, in particular, to a method for producing a parallel gas flow type catalyst for oxidizing sulfurous acid gas.

A so-called contact process for producing sulfuric acid in which a catalyst is used for oxidizing sulfurous acid gas (SO_2) and in which sulfuric acid is obtained as sulfur trioxide (SO_3) has a long history; vanadium type catalysts were first produced in Japan about 50 years ago and have been used since then. There are several references for methods of producing vanadium type catalysts (for example, Ryusan Handbook, published by Ryusan Kyokai, December 15, 1977, pp.345-347), and a number of improved methods have been made public.

Although such catalysts used to be formed into particles or cylinders most commonly, the catalysts are formed in a ring shape or a flower petal shape in recent years. Furthermore, a reactor has been disclosed which uses a parallel gas flow type

catalyst whose gas passage holes are shaped into a honeycomb. While these improvements on the catalyst shape have been made in order to reduce a pressure loss at a catalyst layer, no production method has been established for the parallel gas flow type catalyst for oxidizing sulfurous acid gas.

It is generally more difficult to produce and mold the parallel gas flow type catalyst having gas passage holes of grid or honeycomb shapes, compared to the conventional catalyst that is formed into particles or cylindrical shapes. Since the conventional catalyst is simpler in shape and shorter in length, it can be molded rather easily whether it is formed by extrusion from a metal pattern or it is shaped using a mold. However, the parallel gas flow type catalyst is generally as long as the length of a catalyst layer and has to have a plurality of gas passage holes in its section. It is therefore necessary to form the catalyst into a continuous shape having a grid section or a honeycomb section by providing a plurality of nozzles on a metal mold for shaping the catalyst and by extruding an ingredient powder of the catalyst from these nozzles. In doing so, since the ingredient powder itself does not have a sufficient caking property to be formed into a desired shape, it has been a problem how to add such caking property to the ingredient for easier shaping. Since a catalyst for oxidizing sulfurous acid gas uses, as a carrier, diatom (diatomaceous) earth having silica, which is notorious for

being difficult to shape, as a major component, it is particularly difficult to form this catalyst into complicated shapes with a plurality of gas passage holes as mentioned above, and we have not seen any products of this type.

SUMMARY OF THE INVENTION

In accordance with the present invention there is provided a method for producing a catalyst for oxidizing sulfurous acid gas which uses vanadium pentaoxide as a main catalyst, potassium salts as subsidiary catalysts and diatomaceous earth as a carrier, said method is characterized in that after an ingredient mixture of a solution containing vanadium pentaoxide and potassium salts, diatom earth, water and a binder containing sodium polyacrylate is kneaded, the mixing ratio of these materials being 20-30 weight % of the solution containing vanadium pentaoxide and potassium salts (ph 9-12), 35-50 weight % of diatom earth, 10-25 weight % of water, and 10-20 weight % of the binder containing sodium polyacrylate, said ingredient mixture is molded by extrusion into a shape in which gas passage holes are arranged in a honeycomb or grid structure.

In the present invention, diatom earth is chosen for a catalyst carrier. A material containing silica as a main component is generally used as a carrier, and it also can be considered to use silica gel as a carrier. However, silica gel shows pronounced dilatancy when extruded, and it is difficult to correct this characteristic. Also, silica gel contains a large amount of water, and this water is released when a catalyst product is baked, causing the catalyst to crack or to deform. Therefore, silica gel cannot be used in the present invention. Since diatom earth, on the other hand, is easy to be extruded and the deformation during baking can be controlled, diatom earth is used as a carrier in the present invention.

Nevertheless, because diatom earth itself does not have a sufficient caking property and, although not so much as silica gel, still shows some dilatancy, it is impossible to mold by extrusion as it is. Therefore, a binding agent or binder has to be added in order to form into a desired shape.

Although, as a binder, a variety of organic materials are known and a combination of a plurality of such materials is usually used, a binder containing sodium polyacrylate is chosen for the binder in the present invention. While the inventors of the present invention have tested numerous binders to find almost all of them to be impossible to be formed into a desired shape, this particular binder provides a caking property with catalyst ingredients, making it possible to shape them, and

most of it decomposes and goes out of the product catalyst during the final production step of baking, leaving no ill effects to the catalyst performance. Also, although a sodium component would be left in the product catalyst, it is actually a subsidiary catalytic component of a catalyst for oxidizing sulfurous acid gas and does no harm to the catalyst performance unless its amount is excessive.

The reason for choosing, instead of sodium polyacrylate itself, a binder containing sodium polyacrylate here is that the binder may contain small amounts of materials such as alkali cellulose, sodium alginate, polyethylene glycol, which can also be a binder. Also, the binder may contain polyacrylates other than sodium polyacrylate, such as potassium polyacrylate and/or ammonium polyacrylate.

The catalyst forming materials in the present invention are a solution containing vanadium pentaoxide and potassium salts, diatom earth, water, and a binder containing sodium polyacrylate, which are kneaded together. The mixing ratio of these materials is 20-30 weight % of the solution containing vanadium pentaoxide and potassium salts (pH 9-12), 35-50 weight % of diatom earth, 10-25 weight % of water, and 10-20 weight % of the binder containing sodium polyacrylate.

4. BRIEF DESCRIPTION OF THE DRAWINGS

FIGS.1 and 2 show sections of parallel gas flow type catalysts for oxidizing sulfurous acid gas obtained by the method of the present invention; FIG.1 shows gas passage holes arranged in a grid structure, and FIG.2 shows gas passage holes arranged in a honeycomb structure.

In these figures, 1 denotes gas passage holes and 2 indicates walls.

5. DETAILED DESCRIPTION OF A PREFERRED EMBODIMENT

An embodiment will be shown below of the method for producing a parallel gas flow type catalyst for oxidizing sulfurous acid gas of the present invention. It should, however, be noted that the present invention is by no means restricted to this embodiment.

Vanadium pentaoxide (V_2O_5) was added to a solution containing 4 weight parts of potassium hydroxide and 6 weight parts of water so that the content of vanadium pentaoxide (V_2O_5) in the final product catalyst would become 6 weight %. Subsequently, sulfuric acid solution was added to this solution (called the original solution hereinafter) to make it weakly alkaline.

This original solution was added to diatom earth together with a binder containing sodium polyacrylate, and the resulting mixture was kneaded after a suitable amount of water was added. The amount of a binder added should be as small as possible:

here it was 17 weight % and the amount of diatom earth was 42 weight %. The amount of water added should be chosen as it is most suitable for extrusion. If it is too little the extrusion pressure suddenly increases, making it impossible to extrude. If it is too much, while it is possible to extrude, the catalyst cannot be shaped. Here, the amount of water was 13 weight %.

Next, the kneaded mixture was placed in an extrusion molding machine which contained a metal mold with 100 grid-like gas passage holes whose side was 5.7 mm. After the pressure was reduced to a vacuum, an extrusion product whose length is 500 mm was obtained. Subsequently, this extrusion product was dried and made acidic with SO_2 gas and then baked in an electric furnace up to a maximum temperature of 650°C to obtain a parallel gas flow catalyst for oxidizing sulfurous acid gas.

The content of V_2O_5 in the catalyst was 6.0 % and that of K_2O was 7.9 %. As an example to show that the catalyst thus obtained was in fact capable of oxidizing sulfurous acid gas, a part of the catalyst was cut out and placed in a reaction tube whose inner diameter is 49.5 mm so that the effective volume of the catalyst would be 165 cm^3. A gas mixture of a 10.5 % concentration of SO_2, a 10.2 % concentration of O_2 and N_2 to fill the rest at the entrance was passed through this tube with the catalyst while the tube was kept at a temperature of 470°C by a heater placed on the outside. SO_2 concentrations at the

inlet to and the outlet from the catalyst layer were measured and the ratio of conversion was found as shown in Table 1.

From Table 1 it is confirmed that the catalyst obtained in this embodiment can indeed oxidize sulfurous acid gas.

Table 1

Amount of Gas Passing (Nm^3/h)	Conversion Ratio (%)
0.493	29.8
0.331	44.4
0.165	74.6

According to the method of the present invention, it has now become possible to manufacture a parallel gas flow type catalyst for oxidizing sulfurous acid gas which has a grid or honeycomb structure of gas passage holes. When a catalyst obtained by the method of the present invention is used in a chemical plant for producing sulfuric acid, a pressure loss at a catalyst layer can be reduced considerably, and also the blockage by dust contained in an ingredient gas can be prevented. Therefore, the power consumption of a gas flow blower can be reduced.

The claims defining the invention are as follows:-

1. A method for producing a catalyst for oxidizing sulfurous acid gas which uses vanadium pentaoxide as a main catalyst, potassium salts as subsidiary catalysts and diatomaceous earth as a carrier, said method is characterized in that after an ingredient mixture of a solution containing vanadium pentaoxide and potassium salts, diatom earth, water and a binder containing sodium polyacrylate is kneaded, the mixing ratio of these materials being 20-30 weight % of the solution containing vanadium pentaoxide and potassium salts (ph 9-12), 35-50 weight % of diatom earth, 10-25 weight % of wacer, and 10-20 weight % of the binder containing sodium polyacrylate, said ingredient mixture is molded by extrusion into a shape in which gas passage holes are arranged in a honeycomb or grid structure.

2. The method for producing a catalyst for oxidizing sulfurous acid gas as claimed in Claim 1, which is further characterized in that said binder containing sodium polyacrylate consists of sodium polyacrylate only.

3. The method for producing a catalyst for oxidizing sulfurous acid gas as claimed in Claim 1, which is further characterized in that said binder containing sodium polyacrylate contains at least one of sodium polyacrylate, alkali cellulose, sodium alginate, and polyethylene glycol.

4. The method for producing a catalyst for oxidizing sulfurous acid gas as claimed in claim (1), which is further characterised in that said binder containing sodium polyacrylate comprises sodium polyacrylate and/or potassium

polyacrylate and/or ammonium polyacrylate.

5. A method for producing a catalyst for oxidizing sulfurous acid gas substantially as hereinbefore described with reference to Figure 1 or Figure 2 of the accompanying drawings.

DATED DECEMBER 31 1990

MITSUBISHI JUKOGYO KABUSHIKI KAISHA

By their Patent Attorneys

KELVIN LORD AND COMPANY

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FIG. 1

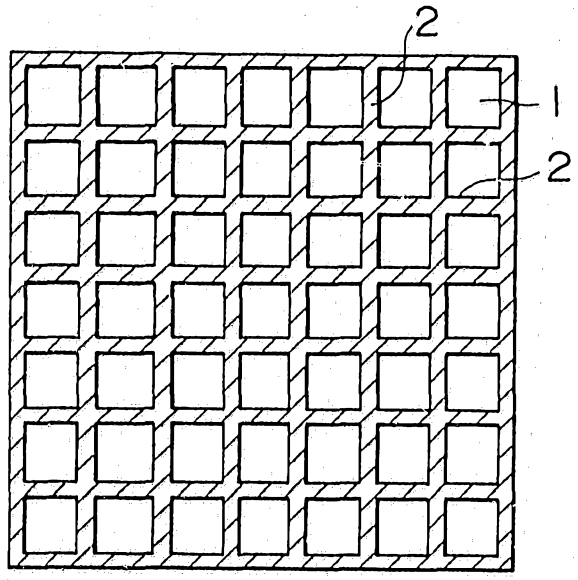

FIG. 2