

PATENT SPECIFICATION



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237,030

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COMPLETE SPECIFICATION.

A Process for Transforming Oxides of Carbon into Oxygenated Organic Compounds.

I, JAMES YATE JOHNSON, a British subject, of 47, Lincoln's Inn Fields, in the City of London, Gentleman, do hereby declare the nature of this invention (which has been communicated to me from abroad by the Badische Anilin & Soda Fabrik, of Ludwigshafen-on-Rhine, Germany, a company incorporated according to German laws), and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

My foreign correspondents have found that oxides of carbon can be readily transformed with good yield into oxygenated organic compounds, and more particularly into methanol, which in accordance with their prior Patent No. 229,715 was done with the aid of catalysts containing certain combinations of hydrogenating elements and elements selected from the fourth to the seventh group of the periodic system, by passing the said oxides in mixture with hydrogen or gases containing hydrogen, at an elevated temperature and pressure over catalysts not specifically mentioned and claimed in the said prior patent containing mixtures of two or more metals, the oxides of which are reduced either alone or in mixture, by the action of reducing gases such as hydrogen or carbon monoxide at ordinary pressure at or below about 550° Centigrade to substantial amounts of the metal, excluding however iron, nickel or cobalt which should not be present. As oxides of such metals copper, silver, lead and cadmium may be mentioned and also such metals, the oxides of which can be reduced under the above conditions, in the special mixture employed. Thus, for instance, zinc oxide is readily reduced in the presence of copper. The contact mass may also contain other

additions, for example a small amount of potassium carbonate, also suitable supports and so on. The process can be suitably carried out at temperatures varying between 200 degrees and 350 degrees Centigrade but higher or lower temperatures may also be used in some cases, the speed of the reaction being increased by the use of higher temperatures. The pressure may range from 100 atmospheres upwards, though lower pressures are also suitable, the greater the pressure the higher is the quantity of gas undergoing reaction in a unit of time. The most suitable gas mixtures are those containing a theoretical amount of hydrogen calculated on the amount of oxide of carbon present or an excess of hydrogen over the said quantity.

The following example illustrates more fully the manner of carrying out the invention into practice, but the invention is not limited to this example.

EXAMPLE.

A dilute aqueous solution containing 10 molecular proportions of copper nitrate for each molecular proportion of zinc nitrate is precipitated, white hot, by adding a solution of potassium carbonate. The carbonates formed are filtered off, carefully washed and dried. The powdery mass is then made into a paste with an addition of about 5 per cent., by weight, of sugar and of about 30 per cent., by weight, of a 30 or 40 per cent. formaldehyde solution, and, if necessary, a little water. The paste is then dried, broken into pieces and reduced with dry hydrogen at between 250 degrees and 300 degrees Centigrade. Over the contact mass thus prepared, a dry mixture of about 1 volume of pure carbon monoxide with about 9 volumes

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of hydrogen is passed at a velocity between 5 and 200 times the volume of the contact mass, calculated on compressed gas, per hour, and at about 300
 5 degrees Centigrade and at a pressure of about 150 atmospheres. The reaction gases leaving the apparatus are cooled while maintaining the pressure, and
 10 liquid methanol separates out with good yield. The contact mass may be brought into any desired shape or applied to a suitable support, or any other proper modifications may be made. Other con-
 15 tact masses may be employed in a similar manner, for example, such as contain two or more of the metals copper, silver, lead, zinc, cadmium, or others in vary-
 20 ing percentages. Carbon monoxide may be replaced wholly or in part by carbon dioxide with very good results, and other gases for example nitrogen may also be present.

Having now particularly described and ascertained the nature of my said inven-
 25 tion and in what manner the same is to be performed I declare that what I claim is:—

1. A process for transforming oxides of carbon into oxygenated organic com-
 30 pounds, and particularly into methanol,

consisting in passing carbon monoxide, or dioxide, or both, with hydrogen at an elevated temperature and pressure over a contact mass containing two or more
 35 metals, the oxides of which can be reduced, as hereinbefore set forth, excluding iron, nickel, or cobalt and the mixtures of the said two or more metals with the metals of the fourth to the seventh group of the periodic system, or
 40 boron.

2. The process for the manufacture of methanol or other oxygenated organic compounds, in accordance with the fore-
 45 going claiming clause; substantially as set forth in the foregoing example.

3. As an article of manufacture, methanol whether pure or mixed with other compounds, when prepared in
 50 accordance with the foregoing claiming clauses.

Dated this 28th day of April, 1924.

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Agents.

Reference has been directed, in pursu-
 60 ance of Section 7, Sub-section 4, of the Patents and Designs Acts, 1907 and 1919, to Specification No. 20,438 of 1913.