PATENT SPECIFICATION

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COMPLETE SPECIFICATION

Process for Regenerating Catalysts for Benzine Synthesis

We, RUHRCHEMIE AKTIENGESELLSCHAFT, of Oberhausen-Holten, Germany, a body corporate organised and existing under the laws of the German State, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

It is known that the catalysts necessary for benzine synthesis from carbon monoxide and hydrogen lose in efficiency after a period of use. This reduction in activity is mainly due to the fact that high molecular organic substances settle on the catalysts. The catalytic efficiency of such catalysts is successfully restored by methods of treatment by which the high-molecular organic deposits are removed from the catalysts. Thus, for example, the weakened catalysts may be treated with hydrogen or a suitable liquid at synthesis temperature. Such measures for the regeneration of catalysts can be frequently repeated, but in the end the high-molecular organic substances adhere so tenaciously to the catalysts that further use of the material is possible only by dissolving the contact substance and restoring it by again precipitating the catalytic metals. It has been found that if the spent contact substances be thus treated the deposits remaining in the contact substance increase the difficulty of restoring the catalysts, the high-molecular paraffins contained in the spent catalysts prejudicially affecting the filtration of the metal salt solution produced by decomposition by the use of acids.

Furthermore the deposition again of the catalytic metals is disturbed by the fatty acids and other oxygen-containing compounds that are produced from the paraffins when the catalysts are dissolved in nitric acid, these preventing precipitation by the formation of complexes. It has been found that all these difficulties can be avoided by passing over the spent catalyst before it is dissolved in acids, hydrogen or inert gases and vapours such as nitrogen, carbon dioxide, or steam, at temperatures lying above 300° C., and preferably at temperatures of from 350 to 400° C. This preliminary treatment removes the organic substances from the catalysts completely.

If it is desired first to recover by itself the high-melting hard paraffin that is to be separated from the spent catalysts, it is advisable before carrying out the preliminary treatment to extract the catalysts with a suitable solvent or scouring agent. It is preferable to employ during this treatment temperatures that lie above the melting points of the hard paraffins. The preliminary extraction of the catalysts whose efficiency has been impaired, has the further advantage that during the subsequent treatment of the catalysts with hydrogen there is a substantial saving of this gas.

The time needed for removing the organic deposits from the catalysts is dependent on the temperature employed and the velocity of the gases or vapours passing over by reason of the fact that when the velocity is raised and the temperature of treatment increased the time needed for the preliminary treatment of the catalysts is reduced. It is possible to remove the deposited organic substances from the contact substance within half an hour when the gases are passed over at temperatures of from 350 to 400° C. at a velocity of from 50 to 60 litres per hour per sq. cm. This time can be shortened by further raising the velocity and by further increasing the temperature accordingly.

The organic compounds are removed from the contact substance under otherwise like conditions more rapidly with hydrogen than with nitrogen or carbon dioxide. The working conditions may, however, be such that it is preferable to employ the inert gases referred to or steam in place of hydrogen. Further experiments have shown that instead of generally employing inert gases or steam, mixtures of these with hydrogen may be

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employed. The use of steam or the addition of steam to the gases has the further advantage that the contact substances loses its pyrophoric properties under the oxidising action of steam, so that it can be brought into contact with air without the danger of spontaneous combustion.

The process of the invention may be employed in many different ways, and the working conditions can always be adjusted to the prevailing conditions. The contact substance may for example be first treated with hydrogen and then with inert gases; finally the treatment with hydrogen or inert gases may be supplemented by a treatment with steam in order to remove the pyrophoric properties from the the catalysts from which the organic substances have been removed.

The following is an example of the carrying out of the invention:

A catalyst consisting of 100 parts Co, 15 parts ThO_2 and 200 parts Kieselguhr, that has been used for benzine synthesis by reduction of carbon monoxide with hydrogen and which has been regenerated in the synthesis furnace very many times by treatment with hydrogen at synthesis temperature until its efficiency has finally departed, is treated with a rapidly flowing stream of hydrogen at 350° C. After two hours have elapsed the mass of catalyst has lost about 40% of its weight, paraffin and other organic substances having been withdrawn from the catalyst with the hydrogen, 80 to 85% of the organic substances removed from the catalyst being obtained in the form of a paraffin with a melting point of from 60 to 65° C. The catalyst mass which under the hydrogen treatment has lost practically all organic substances is moistened with water after cooling, and dissolved in nitric acid. The catalytically active metals are precipitated by means of a soda solution from the metal salt solution thus obtained, after purification by filtration. The washer and dried catalyst is then reduced with hydrogen and is again suitable for effecting synthesis.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—

1. A process for regenerating catalysts that have been used for benzine synthesis from mixtures of carbon monoxide and hydrogen, by dissolving the catalytically active metals in acids and then again precipitating, characterised in that the catalysts whose efficiency has been impaired are treated at temperature of 300° C. or over with flowing hydrogen and/or inert gases before being dissolved in acids.

2. A process according to claim 1 characterised in that steam is mixed with the gases to be used for the preliminary treatment.

3. A process according to claim 1 characterised in that steam is used alone for the preliminary treatment.

4. A process according to claim 1, 2 or 3, characterised in that the preliminary treatment of the catalysts is carried out in stages in such manner that the treatment with different gases is effected in the different stages, the catalyst in the last stage being advantageously treated with steam or gases mixed with steam.

5. A process according to claim 1, 2, 3 or 4, characterised in that before the treatment with hydrogen or inert gases the catalysts are extracted with a suitable solvent for the partial removal of the organic substances.

Dated this 20th day of September, 1937.

EDWARD EVANS & CO.,
40/43, Chancery Lane, London, W.C.2,
Agents for the Applicants.

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ERRATA

SPECIFICATION No. 484,962.

Page 2, line 3, *for* "substances" *read* "substance"

Page 2, line 17, *after* "from" *delete* "the"

Page 2, line 51, *for* "washer" *read* "washed"

Page 2, line 67, *for* "temperature" *read* "temperatures"

THE PATENT OFFICE,
January 21st, 1939.