

UNITED STATES PATENT OFFICE

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PRODUCTION OF CATALYSTS

No Drawing.

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This invention relates to methods of preparing catalysts, and more particularly to methods of preparing the so-called pellet catalysts which are produced by compacting granular material.

Many catalysts, and especially many vanadium catalysts, such as those used for the contact sulfuric acid process and for various catalytic oxidations of organic compounds, such as naphthalene to phthalic anhydride, anthracene to anthraquinone, acenaphthene to naphthalic anhydride, etc. are prepared in the form of pellets by compacting more or less finely granular material which has been mixed or impregnated with the catalytically active compounds into small cylindrical pellets, in which form it is then used in the catalytic converter.

Large scale production of such catalysts presents difficulty, for many of the materials with which the catalyst is associated are relatively abrasive and the ordinary types of tablet machines cannot be used as they seize after a few moments' operation. It has been necessary, therefore, in the past either to make the pellets by hand or to use special machines with large numbers of expensive plates and dies such as, for example, the type of machine described in Patent No. 1,741,312 dated December 31, 1929, to John, Coslow & Schwindt. This type of machine permits very large outputs but requires a relatively large amount of hand labor, as machines of this type require removal of the pellets from the die plates by hand. Despite the amount of hand labor required, which is one of the large factors of cost of the process, machines of the type described in the patent above referred to have been practically the only commercially useful machines for catalyst pellet manufacture.

The present invention avoids the difficulties of the prior manufacture and permits the use of automatic tablet machines such as, for example, the well-known Stokes rotary ma-

chine in which a plate or die is rotated and is filled at one point, the filler material compressed to a pellet at another by means of a punch, and discharged at a further point. Single punch machines can also be used in the present invention. These automatic tablet machines can be continuously and effectively operated by means of the present invention, which consists in adding to the granular pellet-forming material a suitable lubricant which prevents the machine from seizing, reduces wear due to the abrasive nature of the material, and produces pellets of more uniform appearance and enhanced mechanical strength. Any suitable lubricant may be used, such as, for instance, greases, heavy oils such as American oil petrolatum, paraffin wax and other waxes, soaps—both of the alkali metals and other metals—such as aluminum soaps, calcium soaps, etc., and in fact any suitable lubricant which does not introduce undesired constituents into the catalyst. For practical results I have found that higher fatty acids such as palmitic acid, and particularly stearic acid are especially desirable. These products give a very high degree of lubrication, they are cheap, readily available, and at the temperatures under which the catalysts are used they are completely burned out, thus not only assuring that no undesired impurities are added to the catalyst but actually enhancing its porosity and therewith its catalytic effectiveness since the burning out of the acid leaves additional fine pores.

Other stearates such as sodium and potassium stearates (ordinary hard and soft soaps) can be used where the inorganic constituent does not harm the catalyst. Thus, for example, when used in conjunction with a vanadium catalyst for the contact sulfuric acid process or for the catalytic oxidation of organic compounds, sodium is less desirable, but since the amount of the lubricant used is small, potassium is entirely harmless and in some cases actually exerts a desirable action.

In general, for oxidation catalysts aluminum stearates or other aluminum soaps are also harmless, as aluminum is a desirable component of many oxidation catalysts and of vanadium catalysts for the contact sulfuric acid process. The organic lubricants which do not contain any inorganic material are, of course, applicable to all catalysts as they are completely burned out during catalysis, and this is one of the reasons why I prefer to use stearic or similar fatty acids, although the invention is in no sense limited thereto and other lubricants can be used with great effect.

The amount of the lubricant added will depend to some extent on the characteristics of the pellet-forming material and on the exact design of machine. I have found that in making up vanadium catalysts for the contact sulfuric acid process in which complex potassium aluminum polyvanadate gels are admixed with potassium aluminum zeolites diluted with kieselguhr or other porous material, it is desirable to use from one-half to 3% of stearic acid or corresponding amounts of similar lubricants. If aluminum stearate is used, a somewhat larger quantity is ordinarily desirable, for example from one to 5%. With some compositions it is possible to use even less than 1/2%, but generally it is desirable to use at least this much as the lubricant is inexpensive and harmless and a small excess of lubricant does no harm.

While it is a great advantage of the present invention that it permits the use of automatic tablet machines, it should be understood that the invention in its broader aspects is in no sense limited thereto and can be used in any pellet-forming machine. Thus, for example, the addition of from 1/2 to 3% of stearic acid or vaseline to a contact sulfuric acid catalyst material such as that indicated above renders the product more suitable for use in a tablet machine such as that described in the John, Coslow and Schwindt patent referred to above. It is true that the lubricant is not absolutely essential to the functioning of this type of machine but it permits a smoother operation, the pellets obtained have a glossier finish and better mechanical strength. In use, of course, the pellets, by reason of the enhanced porosity resulting from the burning out of the lubricant, show improved activity, and the wear on the dies, always a serious item of expense, is very greatly reduced. Therefore, the advantages of the invention are applicable to machines which can be run without the use of a lubricant. In the preferred modification of the present invention, however, its use in conjunction with automatic tablet machines in this embodiment opens up a new field of pellet catalyst production which was hitherto closed because the automatic machines could not be operated with unlubricated catalyst material.

The present invention not only results in a

greatly improved manufacturing process but it produces a pellet of better physical characteristics, and these improved pellets are included within the scope of the present invention as new products.

The reduction of wear on the machine and the smoother finish of the pellets is no doubt due to a coating of many of the granules of the catalyst material of a thin film of the lubricant and when this film is burned out the catalyst possesses a more open and more porous surface.

It should be noted that the particular catalyst material described above by way of example is not claimed per se, this catalyst being covered by two patents of A. O. Jaeger, No. 1,675,308 and No. 1,694,123, and other similar catalysts described in these patents or otherwise known to the art may be used in conjunction with the present invention, which is directed solely to the feature of the addition of the lubricant and porosity-increasing ingredient and is not concerned with any particular specific pellet catalyst material, being generally applicable to all granular catalyst material which is to be formed into pellets.

The incorporation of the lubricant material may take place by any suitable means, such as, for example, mechanical mixing of finely divided lubricant with the granular material, incorporation by melting the lubricant, or impregnating in the case of liquid lubricants, or by any other suitable means. If desired, in the case of some relatively hard lubricants such as aluminum soaps, they may be dissolved in volatile solvents and incorporated in solution form, the solvent then being permitted to evaporate off. It is an advantage of stearic acid, the preferred type of lubricant, that it can be incorporated without the use of heat or special solvents and is one of the reasons why it constitutes the preferred embodiment of the present invention.

What is claimed as new is:

1. A method of producing catalyst pellets, which comprises incorporating with a catalyst composition 1/2 to 5% of stearic acid, which under the conditions of high temperature vapor phase catalysis does not leave an injurious material in the catalyst, and forming said catalyst material into pellets in an automatic tablet-forming machine.

2. A method of producing compacted catalyst pellets which comprises mixing an active catalyst with abrasive carrier material and an innocuous lubricant in amounts sufficient to offset the abrasive action of the carrier material during compacting, and mechanically compacting the product to form an autogenous bond, independent of any cohesive action of the lubricant.

3. A process according to claim 2 in which the lubricant is stearic acid.

4. A process according to claim 2 in which silica is included in the carrier material.

5. A method of producing a compacted catalyst in an automatic tablet-forming machine which comprises intimately associating an active catalyst with abrasive carrier material, mixing the product with an innocuous lubricant in amounts sufficient to offset the abrasive action of the abrasive carrier material but insufficient to substantially affect the mechanical strength of the pellet and subjecting the product to mechanical compacting.

6. A method of producing catalyst pellets for vapor phase oxidation reactions which comprises admixing an innocuous lubricant with a catalyst composition, compacting the mixture into pellets, and removing the lubricant by heating the pellets in the presence of air.

7. A compressed catalyst pellet comprising mechanically compacted, finely granular abrasive material carrying catalytically active compounds impregnated therein, said pellet containing an innocuous lubricant in amounts sufficient to offset the abrasive action of the granular material but insufficient to substantially affect the mechanical strength of the pellet.

8. A compressed catalyst pellet comprising mechanically compacted, hard granular silicious material having catalytically active compounds impregnated therein, said pellet containing an innocuous lubricant in amounts sufficient to offset the abrasive action of the granular material but insufficient to substantially affect the mechanical strength of the pellet.

9. A compressed catalyst pellet comprising mechanically compacted finely granular abrasive material carrying catalytically active compounds impregnated therein, said pellet containing an organic lubricant free from inorganic constituents in amounts sufficient to offset the abrasive action of the granular material but insufficient to substantially affect the mechanical strength of the pellet.

10. A compressed catalyst pellet comprising mechanically compacted finely granular abrasive material carrying catalytically active compounds impregnated therein, said pellet containing stearic acid in amounts sufficient to offset the abrasive action of the granular material but insufficient to substantially affect the mechanical strength of the pellet.

11. A compressed catalyst pellet comprising mechanically compacted finely granular abrasive material carrying a vanadium compound impregnated therein, said pellet containing an innocuous lubricant in amounts sufficient to offset the abrasive action of the granular material but insufficient to sub-

stantially affect the mechanical strength of the pellet.

12. A compressed catalyst pellet comprising a mechanically compacted permutogenetic mass having associated therewith a vanadium compound and containing an abrasive diluent, said pellet containing an innocuous lubricant in amounts sufficient to offset the abrasive action of the mass but insufficient to substantially affect the mechanical strength of the pellet.

Signed at Pittsburgh, Pennsylvania, this 26th day of May 1930.

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