UNITED STATES PATENT OFFICE.

OTTO CHRISTIAN HAGEMANN, OF YONKERS, AND CHARLES BASKERVILLE, OF NEW YORK, N. Y.

PROCESS OF EFFECTING CATALYTIC REACTIONS.

1,238,137. Specification of Letters Patent. Patented Aug. 28, 1917.

No Drawing. Application filed January 12, 1914. Serial No. 811,734.

To all whom it may concern:

Be it known that we, OTTO CHRISTIAN HAGEMANN, a subject of the King of Great Britain, residing at Yonkers, N. Y., and Dr. CHARLES BASKERVILLE, a citizen of the United States, residing at New York, N. Y., have invented new and useful Improvements in Processes of Effecting Catalytic Reactions, of which the following is a specification.

The present invention relates to catalytic processes, and has special application in the art of hydrogenating oils, such as cotton seed or the like, also oxidation of oils, such as linseed or other drying oils, either with oxygen or ozone, and also is applicable in the effecting of various gas reactions, and other reactions depending upon the presence of a catalytic body.

In our copending application, Serial No. 747,181, (Patent No. 1,083,930, Jan. 13, 1914) we have described and claimed, the use of nickel or similarly acting catalytic metal as a catalyst in various reactions, especially the reaction of hydrogen gas upon liquid oils, for the production of hardened oils or fats, said nickel being in a particular mechanical condition.

In the present invention the catalyst which we preferably employ is nickel, or an alloy of nickel with a small amount of cobalt, having all or a part of its surface oxidized. Since the use of this metal, in the form of extremely thin leaf of a thickness of one twenty-thousandth to one forty-thousandth of an inch, of which the surface portions are oxidized is already covered in our patent above referred to, the present case is restricted to metal forms, having an oxidized surface, (all or part only) said metal being in more massive forms, that is to say the catalytic body is in the form of pieces, of which the smallest dimension is materially greater than one twenty-thousandth of an inch. In other words the present case relates to the use of the metal in forms other than leaves, and with portions of the surface oxidized, as more fully described farther on.

We have discovered that more uniform reliability of catalytic action by such metallic nickel shapes can be secured when previously applying to such nickel shapes, such as wire or sheets, etc., a very slight surface oxidation.

As in our prior application above referred to, the revivification or regeneration of the catalyzer after use, can be effected by further oxidizing, and then reducing the oxidized surface of said nickel shapes or bodies, after which in the present process a slight surface oxidation is again effected prior to the further use of the catalytic body or material. Or we may, in revivifying the catalyzer after use, dispense with the step of reduction, and re-use the metal shapes aforesaid after having previously applied to them slight surface oxidation.

In carrying out the process, we may employ a stationary catalyzer, and move the oil to be treated and the gas for effecting the desired reaction, in opposite directions over said catalyzer, or the oil may be placed in a receptacle, the gas introduced near the bottom of the receptacle, and the mass of oil and catalyzer agitated, in order to bring about complete reaction between various portions of the oil and the reactive material. In this mode of treatment the catalyzer may be made in the form of a stirring or agitating member, for example of wire gauze, or thin plates of nickel or similarly acting metal, the surface portions of which are previously oxidized. The employment of nickel shapes having the surface portion oxidized is believed to be broadly new, and it could not be foreseen that the effectiveness of the catalyzer would be increased, by previously partially oxidizing the surface portions.

We disclaim the use of nickel oxid alone, as a catalyzer, since the same is old and well known.

While we have referred particularly to nickel as the metal to be employed, we desire to call attention to the fact that other similarly acting metals may be employed, such as cobalt, and the like, and we call particular attention to the fact that mixtures or alloys of nickel containing small quantities of cobalt, when oxidized on the surface, act much more vigorously as catalyzers, than either pure nickel or cobalt.

In preparing the sheets, wire and other suitable nickel shapes, for catalytic use, we prefer to heat them in air, oxygen or ozone

until the metal surface ceases to be shiny and bright, or in other words until tarnished or oxidized on its surface, although such oxidation may be very superficial.

In the revivification of the catalyzer we find that a convenient method of procedure is first the separation of all reaction products, such as hydrogenized oil or the like, from the plates, wires or other shapes of nickel, which may be effected by treating the same with a suitable oil solvent, then drying, and heating in an atmosphere of oxygen, ozone or air to a temperature of 300° C. or higher, until the nickel or other metal has been oxidized to a slight depth, after which the catalyzer will be subjected to the action of a reducing gas, such as hydrogen, at a temperature of 300° C. for the purpose of reducing the nickel oxid, after which the catalyzer is again subjected to the action of air or the like, at about 300° C. for a short time, until the surface becomes coated over more or less of its entire surface, with a thin film of oxid.

We call attention to the fact that the method of revivifying nickel and similar catalyzers, as herein described, is claimed in our copending application, Serial No. 801,641.

In regard to the treatment of oils with the reactive gases, such as hydrogen, we may state that the ordinary temperature employed in the art may be employed, and also that the reaction may be facilitated and accelerated by the action of the gases under considerable pressure.

We find in using these oxid-tarnished metal shapes for hydrogenation of oils, that the hardened fats are obtained in a remarkably clean state, and not in any phase of the hydrogenation have we observed any discoloration of the product, as is the case with many other catalyzers heretofore suggested.

As an example of a particular use of the nickel shapes having oxidized surfaces, we call attention to the fact that nickel wire gauze, ordinarily does not operate to produce any appreciable amount of reaction between hydrogen and unsaturated oils, since said metal is not in sufficiently finely divided condition to come into contact with any appreciable amount of oil and hydrogen, but if this nickel wire gauze is somewhat oxidized on its surface, as above described, it operates comparatively rapidly, and produces a comparatively rapid hydrogenation of the oil. This result was not to be foreseen from any of the literature previously published on the subject.

The term "nickel shapes" as used in the specification, and the claims is intended to apply to a body composed essentially of nickel, or nickel containing small amounts of other metals, and is not intended to cover such a material, as pumice stone, or the like, which is impregnated or coated with metallic nickel.

We call attention to the fact that the nickel oxid forming the surface of the catalyzers referred to in this application, is in the form of a coherent layer, closely adhering to the metal, and is not in the form of finely divided nickel oxid, which has heretofore been suggested, as a catalyzer for use in hydrogenating oils. The nickel oxid in the form which we employ it, is very readily separated from the products of the reaction, while in the form of a fine powder this separation is rather difficult.

What we claim is:—

1. A process of effecting catalytic reactions which comprises subjecting the materials entering the reaction, to the action of a catalytic body comprising a metal of the nickel group, in a form other than that of extremely thin leaves, having at least a portion of the surface thereof oxidized.

2. A catalytic process of treating a fluid, organic material which comprises subjecting such a material, and a substance designed to react therewith, to the influence of a catalytic body consisting of a metal in a form other than that of extremely thin leaves, of which at least portions of the surface are oxidized.

3. A process of treating oils which comprises bringing said oil and a medium capable of reacting therewith, into contact with a catalytic comprising a metal in a form other than that of extremely thin leaves, of which at least portions of the surface are coated with a catalytic metal oxid.

4. A process of hydrogenating oil containing unsaturated organic bodies, said process comprising the step of treating said oil with hydrogen in the presence of a catalytic body comprising metal in a form other than that of extremely thin leaves, having at least portions of its surface oxidized.

5. In the art of treating oils, the step of subjecting the oil to the action of a gas capable of combining therewith, in the presence of a catalyzer comprising a nickel shape, in a form other than that of extremely thin leaves, portions of the surface of said nickel shape being oxidized.

6. A process of treating oils and other unsaturated organic bodies, which comprises subjecting said oil, in the presence of a material capable of reacting therewith to the action of an agitating device, including a member made of a catalytic metal, portions of the surface of which are coated with a layer of a catalytic oxid.

7. A process of effecting catalytic reactions which comprises subjecting the materials entering said reaction, to the action of a catalytic body comprising a metal of the nickel group in a form other than very thin

leaves, said body having portions of its surface oxidized.

8. A process of hydrogenating oil, which comprises treating said oil with hydrogen in the presence of a catalytic body comprising nickel in a form other than very thin leaves, having portions of its surface oxidized.

9. A process of treating an unsaturated organic body, in the fluid state, which comprises subjecting said body to the action of a substance capable of reacting therewith, in the presence of a catalyzer containing nickel, and a small amount of cobalt, said catalyzer being in a form other than very thin leaves, the surface of said catalyzer being oxidized.

10. A process of effecting catalytic reactions, which comprises subjecting fluid materials to the action of a catalytic body comprising nickel in a form other than thin leaves, a portion of the surface thereof being lightly oxidized.

11. A process of effecting catalytic reactions, which comprises subjecting fluid substances to the action of a catalytic body comprising nickel in a form other than thin leaves of which the surface has been oxidized to a sufficient extent only to remove the shiny and bright surface appearance thereof.

In testimony whereof we have affixed our signatures in the presence of two witnesses.

OTTO CHRISTIAN HAGEMANN.
CHARLES BASKERVILLE.

Witnesses:
CHARLES WEISMAN,
THOMAS F. O'KEEFFE.