UNITED STATES PATENT OFFICE.

CARLETON ELLIS, OF MONTCLAIR, NEW JERSEY.

MAKING POWDERED METALS AND REDUCED METALLIC COMPOUNDS.

1,329,322. Specification of Letters Patent. Patented Jan. 27, 1920.

No Drawing. Application filed August 1, 1919. Serial No. 314,738.

To all whom it may concern:

Be it known that I, CARLETON ELLIS, a citizen of the United States, and a resident of Montclair, in the county of Essex and State of New Jersey, have invented certain new and useful Improvements in Making Powdered Metals and Reduced Metallic Compounds, of which the following is a specification.

This invention relates to a method of producing metals or metal-containing substances in a finely-divided condition and relates especially to substances which are capable of being used as catalytic material as for example nickel in a finely-divided form adapted for use in the hydrogenation of fatty oils.

Various methods have been proposed for reducing various oxids and salts to yield finely divided metals or sub-oxids having various uses in the arts. For example in the catalytic field it is proposed to reduce nickel oxid by heating in a current of hydrogen to form metallic nickel in a finely divided state. Also it has been proposed to suspend an oxid, hydrate, carbonate or other compound of nickel in a fatty or mineral oil vehicle and to heat while passing a reducing gas through the oil vehicle. Also it has been proposed to heat organic salts of nickel in an oil vehicle to bring about the formation of finely divided nickel.

In the present invention a series of compounds which are not easily reduced by simple heating in an oil vehicle without the addition of a reducing gas are readily converted into catalytic material by heating in a vehicle which breaks down or cracks at an elevated temperature while at the same time reduction of the nickel compound occurs without the formation of objectionable quantities of tars or deposits which would interfere with the catalytic activity of the material or affect the substances in which the catalytic material is to be used.

In making finely divided metals or metal-containing compounds suitable for use as catalysts in hydrogenation in accordance with my invention I preferably employ a vehicle which may be termed neutral or inert to nickel compounds at ordinary temperatures and especially prefer to employ hydrocarbons such as petroleum oils boiling at an elevated temperature and preferably capable of cracking at a temperature somewhat below the normal boiling point. In addition to such oils I may also employ paraffin or ceresin wax or petrolatum (petroleum jelly), as the vehicle with or without the addition of normally liquid hydrocarbon oils. The raw material employed for making the catalyzer is simply heated in this vehicle preferably to a temperature substantially above 300° C., the best temperature in most cases being from 340° to 370° C. but may be higher if desired and in some cases even lower. Preferably no hydrogen or other reducing gas is introduced into the vehicle during the reduction as the latter takes place due to reaction between the nickel compound and the vehicle or the gases or vapors formed by its decomposition during cracking, yielding sub-oxids and metal compounds of a highly useful character. The process has the important advantage, as compared with many methods proposed, of yielding for example catalytic nickel very cheaply.

By using such an inert or neutral vehicle a product is obtained consisting of nickel or reduced nickel compounds incorporated with hydrocarbon oil or wax which can be preserved indefinitely. When finely-divided nickel is kept in fatty oils for any considerable period reaction may take place to some extent with a change in the surface form or spongy condition of the nickel which is not desirable. Hydrocarbons or waxes overcome this objection as in such mediums the catalyzer may be preserved for an indefinitely long time. Hydrogenated fatty oils in many cases are better than unhydrogenated fatty oils for preserving the nickel powder but like the latter are also open to the objection of possible changes including formation of rancid bodies. Finely-divided nickel such as that obtained by reducing a nickel compound, *e. g.*, nickel formate in a liquid or molten vehicle is very finely divided, even colloidal. Because of its degree of attenuation it is easily affected by fatty acids. When the surface of such a colloidal catalyzer is acted upon by a rancid fat it may lose its activity wholly or partially, according to the changes in the porosity, etc., and consequent capacity for absorbing hydrogen gas, is modified.

A hydrocarbon vehicle such as paraffin

wax is a truly inert substance under ordinary condition of storage and shipping. It does not react with the surface film of the particles of finely divided nickel. The wax may be obtained of a high degree of purity at a comparatively low cost. The reduced product solidified and embedded in paraffin constitutes a desirable form in which to make shipment. Besides paraffin or ceresin, compounds such as petrolatum or petroleum jelly, Russian petroleum oil (which is sulfur-free) and the like may be used under certain conditions in modifications of the invention.

The employment of paraffin wax will however be described as representing the preferred form or embodiment. For hydrogenation purposes a wax free from or substantially devoid of sulfur is desired, that is the amount of sulfur, if any be present, should not be sufficient to react with the catalyzer in an objectionable manner to poison and render it inoperative.

The use of paraffin wax or similar inert sealing vehicle is desirable on account of the fact that the water liberated in the reduction of many nickel compounds, which ordinarily at the high temperature required would hydrolyze and decompose fatty oils producing acids, etc., in turn perhaps reacting on the nickel in a detrimental manner, does not have this effect on paraffin. Such decomposition as occurs is more of the nature of oil cracking with liberation of various gases or vapors which in a nascent condition (as liberated in the cracking reaction) may be expected to react vigorously with the nickel material.

Not all hydrocarbon oils are suitable for the purpose of the present invention. Those cracking or boiling off at too low temperatures, or those forming great amounts of tar, gummy masses, resins and the like are, generally speaking, objectionable. In some cases the carbon which may be formed is not objectionable as a component of the catalytic material and in fact may be employed therewith to augment catalytic action. Some hydrocarbon oils contain large amounts of free sulfur or sulfids (or other sulfur compounds) and oils of this character should not be used when producing catalytic material that is poisoned by sulfur.

The action which takes place between, for example, paraffin and nickel carbonate may result directly in the production of nickel or in an intermediate oxid with the liberation of gases such as carbon monoxid, and water vapor.

As stated in Serial No. 236,669, filed May 27, 1918, a catalyzer may also be made by placing nickel hydrate in paraffin wax and heating the mixture to or near the boiling point of the wax. For example in one case the heating was carried out in a deep vessel closely covered (but not sealed in a pressure-tight manner) so that air had practically no access as the vapors of the boiling wax filled the space above the liquid and expelled the air. In the first part of the operation water was evolved with some foaming and later the wax was brought to boiling gently. In the course of twenty minutes the wax became black in color and shortly afterward on examining the material it was found to be filled with fine particles of magnetic nickel. No hydrogen or other reducing gas was introduced into the paraffin during the operation, the reduction taking place spontaneously in the presence of the boiling paraffin or whatever decomposition products may have been produced by the heating. In like manner nickel carbonate or other form of inorganic or organic nickel compound may be employed. In some cases the presence of sulfates or phosphates such for example as sodium sulfate is not objectionable. If however the temperature of the boiling wax is too high sulfates may be reduced to sulfids which would poison the catalyzer. Hence under these conditions the nickel hydrate, nickel carbonate or other form of nickel compound employed should be washed free from sulfates, phosphates and the like. In some cases low boiling waxes may be used in place of the hard paraffin wax which was employed in the above test.

In another case a spent catalyzer containing nickel was ignited to burn out the oil and was suspended in paraffin wax and heated to 340° C., for three hours. Some of this product, when added to cottonseed oil and hydrogen introduced, caused the oil to harden. In another case some of the spent catalyzer was extracted with gasolene and the residual matter heated in paraffin wax for five hours at 340° C. producing catalytic material.

In another case approximately equivalent parts of a mixture of nickel carbonate and sodium sulfate obtained by reacting on nickel sulfate with sodium carbonate was heated in white Russian mineral oil to 340° for one hour affording a black product containing reduced nickel. When vaseline was used as a vehicle in place of the Russian oil a reducing effect was likewise secured.

One part by weight of copper carbonate was heated with two parts by weight of paraffin wax to a cracking temperature for two hours when a reddish product was obtained containing reduced material.

A desirable form of catalyzer is made from nickel carbonate. This may be obtained at a comparatively low cost in the form of a fine powder (*e. g.* like confectioners' sugar) which is free or nearly free from sulfates. This may be added to a vehicle such as paraffin wax, adding a quantity of the carbonate sufficient to make a thin paste when the wax is highly heated.

This amount will vary somewhat depending upon the physical condition of the carbonate but it is possible to use say one part by weight of the carbonate to two or three parts of the wax. The latter is preferably a clean white wax melting between 50 and 60° C. This mixture is contained in a closed receptacle fitted with a stirrer to give a rapid agitation and is heated by direct fire, electrical heating devices, or in any other suitable manner to a temperature preferably as stated above 300° C. to the point where decomposition of the wax takes place to some extent. By keeping the temperature around 340 to 350° C. for four or five hours a good catalytic product is obtained. On cooling, the mass solidifies to a cake which is black in color and contains the nickel in a state of fine sub-division. It is quite active as a catalyzer only .2 of 1% being required for hardening a good grade of cottonseed oil.

The proportion of nickel may vary from say 20 or 25% of the latter for an ordinary factory concentrate to one which may contain 60 or 80% or more of nickel which is better as a shipping concentrate. For example the molten wax and catalyzer may be allowed to settle or be centrifuged to concentrate the nickel. The molten wax thus separated may be removed in a liquid state or the mass may be allowed to cool and the nickel concentrate then separated from the solid waxy portion which is free from the catalyzer. By proceeding in this manner a concentrate containing only 18.7% of paraffin can readily be obtained.

When the wax is heated to a temperature of 340° some loss occurs by cracking, oily material resembling kerosene, etc., distilling off. The closed receptacle referred to above should be provided with a vent preferably a pipe leading into a condenser to collect the oils formed.

In Serial No. 274,357, filed January 31, 1919, the preparation of a catalyzer from nickel nitrate is described in which molten paraffin wax is used as the suspending and decomposing vehicle. Nickel nitrate is placed in a receptacle having a cover and opening through which steam and vapors can escape.

At about 200° fumes of oxids of nitrogen appear and the rise in temperature should then be checked and then heating rendered quite gradual for a period as the fumes of oxids of nitrogen are evolved. Unless this precaution is taken there is danger of violent reaction leading to an explosion. Gradually the temperature is raised while the mass is stirred until a final temperature of about 340° C. is reached. It is held at this point for a half hour or more and a deep black powdered product is obtained. This may be separated from the paraffin by settling and washing the settled out powder with benzol or gasolene. The product obtained is a peculiar one, it is magnetic and has a deep black, mossy appearance in its preferred form being substantially free from metallic luster. The product is in very fine particles and this deep velvety black, amorphous nickel material produced in this manner constitutes a product new in the art and is one of the features of the present invention.

In heating up paraffin to the temperature referred to above it boils and the vapors are condensed on the sides of the vessel and run back. The shape of the receptacle and the condensing arrangements employed should be such as to retain the paraffin oil permitting the escape of the nitrous fumes and other gases. No hydrogen or other reducing gas is introduced into the paraffin wax during this operation and it is believed the physical qualities of the catalyzer are due to the absence of such extraneous reducing gas. Of course it is possible to introduce a reducing gas if desired but this is not advisable in the preferred form of the invention.

The catalyzer obtained in the foregoing manner by treatment in paraffin is non-pyrophoric and is not readily oxidized. Specimens exposed to the air for a considerable period of time showed little or no deterioration.

In one case one part by weight of nickel nitrate and two parts of paraffin wax were heated to a temperature of somewhat above the melting point of the wax when foaming occurred and the volume increased to four times the original volume due to the evolution of water vapor. On continuing the heating fumes of oxids of nitrogen were evolved which was especially marked at 200°. The temperature was raised and maintained at 340° for a half hour. When reducing nickel nitrate in this manner it is usually necessary or at least advisable to agitate the wax vehicle violently as the nickel nitrate may otherwise, during the reduction, form compact masses which may not be suitable for catalytic purposes although they may have other uses.

In another case the nickel nitrate was heated eventually up to 370° for a half hour giving a finely-divided product.

In a similar manner cobalt compounds may be reduced. For example one part of cobalt nitrate and two parts of paraffin wax were heated to 200° C. at which temperature a violent action occurred, after holding at this point until the reaction had subsided somewhat, the temperature was raised to 340° and was held at this point for about an hour. A very finely divided black catalyst was obtained. While I prefer to prepare, for example, catalytic material with-

out the use of any carrier or supporting material it is also possible to impregnate porous material such as pumice or kieselguhr with a solution of nickel nitrate or to precipitate nickel carbonate in and on the particles of this porous mineral matter, dry and heat in an oil to a cracking temperature to bring about reduction. While I prefer not to introduce a reducing gas such as hydrogen and the like owing to the cost of the latter, the possibility of introducing impurities and to danger from explosions, etc., I do not definitely exclude the introduction of a reducing gas at a temperature of 300° and higher more especially at a cracking temperature of the wax or oil while employed.

The containers which may be used for carrying out the reduction are simple iron or steel tanks and if desired the operation may be carried out at sub- or super-atmospheric pressure instead of at ordinary atmospheric pressure. While I have indicated that it is preferable to use a hydrocarbon solvent in place of a fatty oil it is possible under certain conditions to use fatty oils such for example as hydrogenated oils, heating this to a cracking temperature or higher but in such cases the product is ordinarily not so well suited for catalytic purposes as when paraffin is used, but may be used for other purposes.

Nickel obtained by reduction of for example nickel carbonate, hydrate or nitrate in paraffin wax may be freed of wax by extraction with a suitable solvent such as benzol. The nickel powder obtained in this manner is more air-stable than nickel reduced by hydrogen as for example by passing hydrogen over nickel oxid at a temperature of 400 or 500° C. The nickel obtained in the latter manner oxidizes and takes fire quickly in the air while my product may be exposed to the air for an indefinite period without heating and the loss in activity on exposure is relatively slight in comparison with the dry reduced product referred to. In the preferred form of the invention the catalyzer is non-pyrophoric or substantially non-pyrophoric and is of a relatively air-stable nature. When consisting or containing mainly nickel material its preferred form is amorphous finely-divided having a deep black color. When incorporated in a medium such as cracked paraffin wax a black waxy solid mass or cake is produced which when cut or broken does not show any gritty qualities owing to the fineness of the nickel material. The present invention contemplates as one feature a reduced metal or mixtures of metals or their sub-oxids, or mixtures of a metal and a sub-oxid, incorporated in a hydrocarbon material or medium such as petroleum jelly, paraffin or ceresin wax or other wax, mineral oils and the like, such hydrocarbon being in a form substantially as obtained by cracking but being substantially free from tars, resins, pitches and other products which would impair catalytic activity. As stated paraffin wax is especially suited for the purpose because on cracking the latter little or no tar is obtained, in fact only a discoloration with the production of very slight carbonaceous residue in the finished product. It may be added, however, that the formation of clean finely-divided carbon in the material is not objectionable in many cases, the carbon forming a co-catalyzer, useful in the hydrogenation of fatty oils. It is possible, however, to use an oil medium which produces little or no free carbon, at least no amount which is perceptible to any degree in the finished product.

When using the catalyzer it may be melted into the oil which is to be hydrogenated. As only .2 or .3% of the catalyzer may be used the amount of paraffin wax introduced in the oil in many cases being so small as not to be detectable by ordinary analytical methods. Such a quantity of wax is, moreover, used only in starting off the first hydrogenation run as the catalyzer is used repeatedly and if the presence of wax in the oil is objectionable in the first run, as for example assuming its presence to be criticised in an edible product, the first run may be employed for making, for example, soap and the catalyzer filtered out from the first run being thus substantially freed of all but a mere trace of wax, may then be used to hydrogenate an oil for edible purposes. When employing a refined oil such as high grade cottonseed oil the catalyzer may be used over and over for a long period of time with the addition of say 3 to 5 one hundredths of the normal quantity of fresh catalyzer after each run in order to make up for losses in handling, etc.

Using about 40 pounds of the nickel powder (which may be used alone or suspended in about 150 pounds of slightly cracked paraffin wax) as a catalyzer, a quantity of refined oil ranging between 15,000 and 20,000 pounds may be hydrogenated, and the catalyzer recovered by filtration may amount to about 38 pounds. This quantity of catalyzer is added to a fresh batch of oil similar in amount as the first but with the addition of another 2 pounds of fresh nickel powder (in wax if desired) to make up for the loss incurred. In the same manner after hydrogenating the next batch of oil fresh catalyzer is added in like quantity to replace the nickel loss in handling, dissolved in the oil, or otherwise lost. The catalyzer used under these conditions is best made from nickel carbonate.

Nickel formate referred to above decomposes (by itself without reference to any reduction by the oil vehicle) when heated in

oil or wax at 210° or slightly higher. It is therefore not necessary when using nickel formate to heat the mixture to a temperature sufficient to crack the oil. Nickel formate alone reduces spontaneously on heating. Nickel acetate and oxalate decompose less readily than the formate and the latter on heating alone tends to give the oxid of nickel. The acetate and oxalate may be heated in petroleum hydrocarbon to a cracking temperature to give catalytic material. However the present invention is not primarily directed to the reduction of organic salts of nickel or metallo organic compounds thereof such for example as the salts of organic nitrocompounds, etc., which contain agencies for their own reduction, namely, such quantities of carbon and hydrogen in their structure in excess of the oxygen present that heating, the compound itself in the absence of other material, causes decomposition and leaves a residue of nickel. The present invention is intended more especially for the production of finely divided metals *e. g.*, nickel or cobalt from inorganic compounds of these metals which compounds do not give the metal on simple heating by themselves. In this group fall for example nickel hydrate and carbonate, cobalt nitrate, nickel nitrate, nitrite and the like which on heating give nickel oxid and no further heating, certainly at least at a temperature which would not cause sintering will bring about reduction to the metal or to material which is catalytically active. Compounds such as nickel carbonate or green hydrated nickel oxid are very cheap in comparison with nickel formate, they are even found in nature in a state of fair purity. Such compounds have heretofore been reduced only by the aid of reducing gases brought from some outside source and on account of their cheapness it has been desired to bring about their reduction in a simple manner which would secure the conversion of the bulk of the raw catalytic material into active catalyzer. The present invention thus embraces the reduction by heating in a hydrocarbon oil or in a wax, etc., of an inorganic compound of nickel, cobalt, and the like *e. g.*, (the oxid, hydrate or carbonate), which is not reducible to a catalytic material by simple exposure to an elevated temperature. The heating with the hydrocarbon oil or wax, etc., being carried to the point at which reaction on the nickel or other compound by the hydrocarbon material (or products of decomposition of the latter) is brought about, and a metallic powder or a precipitate containing metallic material or at least material of a highly reduced character, is formed. The decomposition of the wax or hydrocarbon oil doubtless evolves compounds whose chemical activity when freshly liberated is especially marked in forming catalytic material.

Attention has been called above to the undesirability of using hydrocarbons which form tarry matter on heating to a cracking temperature. These tars dissolve in the fatty oil and discolor it. Hence any hydrocarbon forming an objectionable amount of this material is preferably not used, or if employed, it is preferable to wash out the cracked organic matter and tar from the catalyzer, by means of, for example, benzol or any other suitable washing medium. Further as noted, paraffin wax or similar mineral wax is well suited as a medium in which the catalyzer is formed and the use of a hard brittle wax free from oleaginous material including freedom from fatty material reactive with nickel on standing (thus changing the physical structure of the catalyzer) affords a product which can be shipped in ordinary cloth bags or slack cooperage. No expensive shipping container is required. The catalyzer may be shipped in the form of cakes or rough fragments which for use are simply added to the oil to be hardened or if desired may be melted in a separate container with some oil and pumped to the point of use. By employing a hard wax so that the fragments do not fuse together in shipping, the product is more readily handled for some purposes.

With regard to temperature limitations of the process it may be stated that the lower limit is the temperature at which reduction takes place at a sufficient degree of speed. Ordinarily little or no reduction takes place at 300° but the effect increases until at 340 to 350° the action is quite rapid in the case, for example, of paraffin wax. The upper limits of temperature (preferably but not necessarily) are those prescribed by tar formation and the like, and preferably with salts of the nature of nickel hydrate and nickel carbonate, I prefer a temperature between 300 and 400° C.

The metals nickel, cobalt and copper being in many respects similar, I use the expression metal having an atomic weight between approximately 58.7 and 63.6 to cover any of this group of metals.

I have claimed the treatment of nickel nitrate, as herein described, in a copending application Serial No. 274,357, filed January 31, 1919.

What I claim is:—

1. The process of making a powdered metal which comprises heating a reducible metal compound in a hydrocarbon vehicle, which is substantially non-volatile at the temperature employed to a cracking temperature whereby reduced metallic material is obtained.

2. The process of making a catalyzer which comprises heating a reducible nickel

material in paraffin wax to the cracking temperature of the latter whereby finely divided nickel material is obtained.

3. The process of making a catalyzer which comprises heating a reducible compound of nickel in oil to a temperature sufficient to produce cracking of the oil.

4. The process of making a catalyzer adapted for use in the hydrogenation of fatty oils which comprises agitating nickel carbonate with paraffin wax at a temperature of over 300° C. without the addition of any extraneous reducing gas whereby nickel catalyzer is produced.

5. The process of making a catalyzer adapted for use in the hydrogenation of fatty oils which comprises incorporating a nickel material comprising nickel carbonate with a hydrocarbon vehicle comprising paraffin wax, in heating the mixture to a temperature above 300° C. whereby cracking of the wax occurs, agitating the mixture during the progress of the cracking, withdrawing moisture, gases and vapors, and in settling the resultant product to form a concentrate of nickle powder in a waxy medium.

6. The process of making a catalyzer adapted for use in the hydrogenation of fatty oils which comprises incorporating a basic compound of nickel with a hydrocarbon vehicle comprising paraffin wax, in heating the mixture to a temperature above 300° C. whereby cracking of the wax occurs, agitating the mixture during the progress of the cracking, withdrawing moisture, gases and vapors, and in settling the resultant product to form a concentrate of nickel powder in a waxy medium.

7. The process of making a catalyzer adapted for use in the hydrogenation of fatty oils which comprises incorporating a basic compound of nickel comprising nickel carbonate with a hydrocarbon vehicle, in heating the mixture to a temperature above 300° C. whereby cracking of the hydrocarbon vehicle occurs, agitating the mixture during the progress of the cracking, and withdrawing moisture, gases and vapors formed during the heating operation.

8. The process of making a catalyzer adapted for use in the hydrogenation of fatty oils which comprises incorporating a basic compound of nickel with a wax vehicle, in heating the mixture to a temperature above 300° C. whereby cracking of the wax occurs, and agitating the mixture during the progress of the cracking.

9. The process of making a catalyzer adapted for use in the hydrogenation of fatty oils which comprises incorporating a compound of nickel with a hydrocarbon wax, vehicle not volatile at 300° C. in heating the mixture to a temperature above 300° C. whereby cracking of the wax occurs, and agitating the mixture during the progress of the cracking.

10. The process of making a catalyzer adapted for use in the hydrogenation of fatty oils which comprises incorporating a basic compound of nickel comprising nickel carbonate with a vehicle comprising wax, in heating the mixture to a temperature above 300° C. whereby cracking of the wax occurs, agitating the mixture during the progress of the cracking, withdrawing moisture and vapors, and in settling the resultant product to form a concentrate of nickel powder in a waxy medium.

11. As a catalytic material, a reduced nickel powder of a black amorphous appearance suspended in a mass of cracked paraffin wax.

12. As a catalytic material, a reduced nickel powder of a black amorphous appearance suspended in a mass of cracked wax.

13. As a catalytic material, a reduced nickel powder of a black amorphous appearance in cracked petroleum.

14. As a catalytic material, a reduced nickel powder of a black amorphous appearance suspended in a mass of cracked solid hydrocarbon.

15. As a catalytic material, a reduced nickel powder suspended in a mass of cracked paraffin wax.

16. As a catalytic material, a reduced nickel powder of a black appearance suspended in a cracked hydrocarbon.

17. A process of preparing a nickel catalyzer by reduction, which comprises heating to 340° C. a bath consisting essentially of paraffin wax carrying an inorganic nickel compound which, when heated alone, is incapable of producing metal, without introducing extraneous reducing gas thereto.

18. A process of preparing nickel catalyzer by reduction which comprises heating to a temperature between 300 and 400° C. a bath comprising a hydrocarbon capable of cracking within the temperature limits mentioned, and carrying an inorganic nickel compound which, when heated alone, is incapable of producing metal, without introducing extraneous reducing gas thereto.

19. The process of preparing a nickel catalyzer by reduction which comprises heating an inorganic nickel compound which, when heated alone, is incapable of producing metal, in an organic vehicle containing hydrocarbon material, to a cracking temperature but not to a temperature at which tarry material is formed in any substantial amount; said operation being carried on without introducing extraneous reducing gas thereto.

20. A process of preparing nickel catalyzer by reduction which comprises heating to a temperature between 300 and 400° C., a bath comprising a hydrocarbon capable of cracking within the temperature limits mentioned, and carrying an inorganic compound of a metal having an atomic weight between approximately 58.7 and 63.6, which compound, when heated alone, is incapable of producing metal, without introducing extraneous reducing gas thereto.

21. As a catalytic material, a reduced metal having an atomic weight between approximately 58.7 and 63.6, carried in a vehicle of cracked hydrocarbon which is essentially non-volatile at 300° C.

CARLETON ELLIS.