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(54) ORGANIC COMPOUND

(54) COMPOSE ORGANIQUE

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We have found that by the reduction of oxides of carbon organic compounds containing or not containing oxygen are produced in a highly advantageous manner when employing catalysts which, in addition to iron, nickel or cobalt, or compounds of the same, in any convenient form, contain cadmium, or thallium, or their compounds or mixtures of these substances. Such catalysts are effective at comparatively low temperatures as for example even at from 180° to 200° Centigrade, but preferably the process is carried out between about 250° and 350° Centigrade, though the catalysts may also be used at temperatures of about 400° Centigrade. Again the admixture of cadmium or thallium or their compounds or mixtures completely prevents any deposition of soot on the catalyst, even if the gas under treatment be rich in carbon monoxide. The other elements of the 8th group of the periodic system, such as platinum, palladium and the like, also give in combination with cadmium or thallium, a similar action to that given by iron, nickel or cobalt. The activity of the catalysts is assisted by still further additions of substances such as copper, gold, silver, cerium, zirconium, aluminium, vanadium, uranium, chromium, molybdenum, tungsten, manganese, alkali metals and alkaline earth metals or compounds of the same, provided that metal compounds containing oxygen which are difficultly reducible to the metal should not be present at all or only in an amount not exceeding 5 per cent, preferably less than 1 per cent of the catalytically active material.

A typical catalyst suitable according to the present invention is one containing iron, copper and cadmium, employed either as such or with still further additions, especially of oxides or carbonates of the alkali metals in the abovementioned amounts. Thus for instance, a catalyst containing more than

50 per cent of iron besides copper and cadmium and less than one per cent of an alkali metal in any convenient form is particularly effective. The catalysts may be used in a great variety of forms, such for example as alloys, and may also be prepared in a great variety of ways. For example, the oxides prepared by carefully heating the nitrates, may be used as initial materials or the components concerned may be precipitated jointly, from solution, as hydroxids, in some cases in the presence of a carrier such as pumice, asbestos and the like, or the oxides or hydroxids and the like may be intimately mixed together, and then charged into the reaction chamber after having been reduced. Highly effective catalysts may also be obtained by using the corresponding salts of ferro- or ferri-cyanic acid as initial substances.

The process may be carried out at ordinary or elevated pressure, e.g. up to about 200 atmospheres, though even very high pressures up to 1000 atmospheres and higher may also be used. According to the conditions of working as regards temperature, pressure, composition of the gases, catalysts and the like, solid, liquid or gaseous hydrocarbons may be produced, or also products containing oxygen or mixtures of these. In some cases the products containing oxygen consist to a large extent of ethyl alcohol or of esters, which boil between 150° and 300° Centigrade, and are highly suitable as solvents and softening agents for lacquers and the like. Highly valuable fats and waxes may be recovered from the fractions of high boiling point. The liquid hydrocarbons may be used as motor fuel. The gaseous products may be separated, for example by strongly cooling, adsorption with active carbon, extraction with washing oils and, if desired, may be converted, by further treatment, into alcohols, or also into liquid hydrocarbons. They may also be employed as lighting or heating gas or the like. The working operations

already known in the catalytic production of compounds from gases, especially from carbon monoxid and hydrogen, namely operating in a continuous cycle, previously purifying the gases, separating the reaction products by cooling and / or by adsorption masses, purifying the circulating gases and so on, and also the devices for carrying off the heat of the reaction and the like may also be applied, in a suitable manner, to the present process.

A great variety of gaseous mixtures may be employed such as water gas, coke-oven gas, coal gas, producer gas, or mixtures of the same, with or without the addition of carbon monoxid and other gases. Moreover, coals or hydrocarbons such as natural gas and the like, or tar oils and the like, may be incompletely burned with oxygen, or mixtures of oxygen and steam, the resulting gases being employed after purification and, if necessary, after catalytic conversion of a portion of the carbon monoxid by means of steam into carbon dioxid and hydrogen.

The hydrogen needed for the reaction may also be generated by the aid of the catalyst itself, by wholly or partially replacing the hydrogen in the initial gases by steam, light hydrocarbons such as methane, or mixtures of the same.

Such parts of the apparatus as grow hot and are exposed to the contact with the reaction gases, are preferably constructed, with a view to prevent the deposition of carbon, in place of iron, of silver, copper, manganese bronze, chromium-nickel, aluminium, iron alloys, such as chromium-, tungsten-, or manganese steels, ferrosilicon or the like, or the said parts may be provided with a suitable coating.

The following examples will further illustrate the nature of the said invention which however is not limited thereto. The parts are by weight.

Example 1.

A solution containing 12 parts of iron, 2 parts of cadmium and 3 parts of copper, in the form of nitrates, is precipitated with potassium carbonate, and the resulting precipitate is thoroughly washed and dried. Over the resulting catalyst placed in a copper-lined contact tube, water gas, freed from carbon dioxide and sulfur compounds in the usual manner is passed at a rate of 60 litres per each 8 cubic centimetres of the catalyst, per hour, at from 300° to 350° Centigrade and at 100 atmospheres pressure. From the separator, which is cooled to the ordinary temperature, but maintained under pressure, a liquid is drawn off composed of two layers, the upper one consisting mainly of hydrocarbons, higher alcohols and esters, while the lower one consists mainly of water with about 40 per cent of lower alcohols, especially ethyl alcohol and organic acids. The catalyst may be kept in use for a long period without losing its efficiency.

By attaching a vessel, charged with active carbon, in rear of the separator, benzines and hydrocarbons boiling below 20° Centigrade, may also be recovered.

Example 2.

Water gas is passed, at the rate of flow described in Example 1, at from 250° to 300° Centigrade and at from 50 to 100 atmospheres pressure over cadmium-copper-ferrocyanide prepared by precipitating a solution of potassium ferrocyanide with a solution of cadmium and copper salts. The yield is still higher than in Example 1.

If the operation be conducted at ordinary pressure, considerably larger quantities of catalysts must be employed in order to obtain the same yield.

Example 3.

A solution of 2 molecular proportions of iron nitrate and 1 molecular proportion of thallous nitrate and copper nitrate the carbonates of the metal are precipitated with sodium carbonate, the deposit is worked and dried. Under the conditions of working specified in Example 1 ample quantities of hydrocarbons and organic compounds containing oxygen are obtained with the above catalyst which does not gradually lose its effectiveness by deposition of carbon as it is mostly the case when employing iron catalysts free from cadmium or thallium.

A similar action is produced by catalysts prepared by precipitating the heavy metal components from solutions or their salts by means of the calculated amount of ammonia, intimately mixing the precipitates and impregnating them, after drying, with such small amounts of sodium, potassium, or lithium carbonate, that the alkali carbonate content of the catalyst does preferably not exceed 1 per cent calculated as metal.

What we claim is:

1) The process of producing organic compounds which consists in exposing at a temperature between about 180 and 400° Centigrade a gas mixture comprising an oxid of carbon and hydrogen to the action of a catalyst comprising a component selected from the group of metals of the 8th group of the periodic system, their compounds and mixtures thereof, and a component selected from the group of cadmium and thallium, their compounds and mixtures thereof, but containing at the most 5 per cent of a metal compound containing oxygen difficultly reducible to the metal.

2) The process of producing organic compounds which consists in exposing at a temperature between about 180 and 400° Centigrade a gas mixture comprising an oxid of carbon and hydrogen to the action of a catalyst comprising a component selected from the group of metals of the iron group, their compounds and mixtures thereof, and a component selected from the group of cadmium and thallium, their compounds and mixtures thereof, but containing at the most 5 per cent of a metal compound containing oxygen difficultly reducible to the metal.

3) The process of producing organic compounds which consists in exposing at temperatures between 180 and 400° Centigrade a gas mixture comprising an oxid of carbon and hydrogen under a pressure up to about 1000 atmospheres to the action of a catalyst comprising a component selected from the group of metals of the iron group, their compounds and mixtures thereof, and a component selected from the group of cadmium and thallium, their compounds and mixtures thereof, but containing at the most 5 per cent of a metal compound containing oxygen difficultly reducible to the metal.

4) The process of producing organic compounds which consists in exposing at temperatures between 250 and 350° Centigrade a gas mixture containing an oxid of carbon and hydrogen under a pressure up to about 200 atmospheres to the action of a catalyst containing iron, copper and cadmium in the elementary form and less than 1 per cent of an alkali metal in the form of an oxygen containing compound, not more than 5 per cent of metal compounds containing oxygen difficultly reducible being present in total.

5) The process of producing organic compounds which consists in exposing at temperatures between 250 and 350° Centigrade a gas mixture containing an oxid of carbon and hydrogen under a pressure up to about 200 atmospheres to the action of a catalyst consisting of at least 50 per cent of iron, copper and cadmium and less than 1 per cent of an alkali metal in the form of an oxygen containing compound.

6) The process of producing organic compounds which consists in exposing at a temperature between about 180 and 400° Centigrade a gas mixture comprising an oxid of carbon and hydrogen to the action of a catalyst comprising a component selected from the group of metals of the 8th group of the periodic system, their compounds and mixtures thereof, and a component selected from the group of cadmium and thallium, their compounds and mixtures thereof, but containing at the most 5 per cent of a metal compound containing oxygen difficultly reducible to the metal, the catalyst being deposited on a carrier.