

UNITED STATES PATENT OFFICE.

FOORD VON BICHOWSKY, OF GLENDALE, CALIFORNIA.

METHOD FOR THE PRODUCTION OF COMPOUNDS CONTAINING SILICON AND NITROGEN.

1,415,280.

Specification of Letters Patent.

Patented May 9, 1922.

No Drawing.

Application filed February 23, 1921. Serial No. 447,281.

To all whom it may concern:

Be it known that I, FOORD VON BICHOWSKY, a citizen of the United States of America, residing in the city of Glendale, in the county of Los Angeles and State of California, have invented certain new and useful Improvements in Methods for the Production of Compounds Containing Silicon and Nitrogen, of which the following is a specification.

My invention relates to the production of compounds containing principally silicon and nitrogen from crude materials, mainly composed of the oxygen containing compounds of silicon and free from carbon as such, through the agency of catalytic substances so that temperatures, lower than previously deemed practical, may be employed, thereby avoiding the loss of any appreciable quantity of nitrogen containing material through volatilization.

I have discovered that this result may be easily obtained by using as a reducing agent carbon, solely in the form of hydrocarbon gases and in particular the impure mixtures of such hydrocarbon gases, known as natural gas, consisting principally of methane and practically free from oxygen, which mixture of hydrocarbon shall hereinafter be referred to as methane. By the use of this gas mixed with nitrogen and in the absence of free carbon, as a reactive agent, but in the presence of certain activating metallic oxides or combinations of metallic oxides, or metals and their alloys, as supplementary catalysts, and certain alkali metal salts, hereinafter described the methane is split, at the temperatures employed very completely into carbon and hydrogen, both of which being in the nascent state have an exceptionally high reducing power. By these means the silicon nitrogen compounds may be prepared in an indirect fired pottery tunnel kiln without the use of saggers, provided the material be briquetted in a very efficient and cheap manner, and the employment of the electric current for either the supplementary or the direct heating of the reactive mass is therefore avoided. Prepared in this way the silicon nitrogen compounds are enabled to compete with ammonia as fertilizing agents, because of their high nitrogen content. These compounds may be used as a source of other nitrogen compounds such as the cyanides or ammonia

and from them pure silica, alumina and the like may also be recovered.

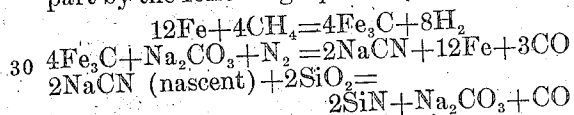
Having now outlined the purpose of my invention, I will go in greater detail into the method of carrying it out. The activating metallic oxides, or combination of metallic oxides hereinbefore referred to, may be small amounts of the oxides of such metals as vanadium, molybdenum, manganese, aluminium, titanium, zirconium, tungsten, niobium, chromium, iron and in general those metals which will directly combine with nitrogen to form nitrides. These oxides may occur as natural impurities in the siliceous material treated or they may be added, singly or in mixtures, to increase the activity of the siliceous material towards nitrogen. Most valuable for this purpose, however, are the compounds of the iron group of metals or their alloys which exert a most marked effect in splitting the methane into active carbon and hydrogen. Under this grouping of elements come the metals, iron, nickel and cobalt, which, alone or alloyed, or as their reduced oxy compounds or mixture of same, with the addition of small amounts of the nitride forming metals already referred to, constitute the activating or catalytic agents which I most frequently use in carrying out this invention. The alkali metal oxygen containing salts may be either the carbonates, oxides or hydroxides of sodium or potassium or mixtures of these salts with or without the use of similar compounds of the alkaline earth metals, or they may be the alkali silicates, aluminates, borates, vanadates, titanates, molybdates, zirconates and the like. By the use of these substances the absorption of nitrogen by the impure silicon oxygen containing compounds begins in the neighborhood of 1100° C. and is quite rapid at 1300° centigrade. Above that temperature many of the metallic alloys begin to melt and if that occurs the temperature must be lowered lest the reactive mass fuse together, making the separation of the metallic particles from the nitrides a difficult mechanical operation. With this point in mind it is best to avoid, as far as possible, any sulphur containing material, as this element rapidly lowers the resistance of the metals towards high temperature by forming easily fusible sulphides with many of them.

The following examples, in which the parts are given in weight, and the temperature in degrees centigrade, will serve to make clear the exact nature of my invention, which, however, is not limited to the following:—

Example No. 1.

- Mix together very thoroughly
 100 parts of finely ground titaniferous
 beach sand containing 3 per cent of
 titanium oxide
 93 parts of iron filings containing 30 per
 cent of nickel
 2 parts of soda ash
 3 parts of water glass 42° Bé. as a
 binder

and press the mixture into briquettes. Stack the latter on cars and heat them in an indirect fired tunnel kiln for 12 hours at a temperature of 1300°. Allow the briquettes to cool in the cooler part of the kiln in a neutral atmosphere. Upon grinding the briquettes, the iron alloy may be recovered by magnetic means. The residue consists mainly of titanium nitrides and silicon nitride. The reaction that takes place may be represented in part by the following equations—



The gaseous products escape and may be burned.

Example No. 2.

- Mix together most thoroughly
 50 parts of fire clay
 46 parts of finely ground iron oxide
 1 part of sodium vanadate
 3 parts of soda ash

and moisten and press the mixture into briquettes. Place on cars in the tunnel kiln and while the briquettes are being slowly heated, pass in pure methane, or other reducing gas to reduce the iron oxide used to the metal. When the temperature of the briquettes reaches 1200°, pass in a dry mixture of methane and nitrogen in equal volumes

and continue the heating to just below 1300° for 15 hours. When cold the magnetic iron may be separated out mechanically, the vanadium nitride and unacted upon vanadate may be recovered by heating with sulphuric acid or the mass, freed from iron, may be fused with caustic soda whereby ammonia is evolved in large quantity. The silica, alumina and vanadium may be recovered from the melt by treatment with water, precipitating the alumina from the solution with CO_2 and removing the precipitate, then precipitating the silica by acids from the heated solution. The vanadium will be found in the filtrate from this reaction.

I claim:

1. The process of producing compounds containing silicon and nitrogen by heating a silicon oxygen compound containing reactive mass and comprising a metallic oxide and a metal of the iron group together with an alkali metal salt, in the absence of carbon, as a chemical reactive, but in the presence of methane and nitrogen.

2. The process of producing compounds containing silicon and nitrogen by heating a reactive mass containing silica, a metallic oxide and a metal of the iron group together with sodium carbonate, in the absence of carbon as such but in the presence of methane and nitrogen.

3. The process of producing compounds of silicon and nitrogen by heating a reactive mass composed of silica and a body containing metallic oxide and particles of iron together with sodium carbonate, but free from carbon as a chemical reactive, at a temperature not exceeding the fusion point of the mass, and passing over it a mixture of nitrogen and methane.

4. The process of producing compounds containing silicon and nitrogen by heating a reactive mass composed of silica and aluminium oxide and particles of metallic iron, and sodium carbonate, but free from carbon as a chemical reactive at a temperature not exceeding 1300° C., while passing over it a mixture of nitrogen and methane.

FOORD VON BICHOWSKY.