To all whom it may concern:

Be it known that I, Heinrich Specketer, a citizen of Germany, and resident of Griesheim-on-the-Main, Germany, have invented certain new and useful Improvements in Methods of Producing Nitrogen Compounds, (for which I filed an application in Germany Aug. 18, 1916,) of which the following is a specification.

This invention relates to a method of fixing nitrogen by heating metallic oxygen compounds with coal in the presence of nitrogen. According to this invention, nitrogen compounds are produced by heating mixtures of metallic oxygen compounds (oxides or carbonates) with coal in an atmosphere containing nitrogen, the materials being intimately associated by being mixed together in pulverized form and the nitrogen being provided free access to all particles of the material by continuously agitating the material during the reaction. The mixed material is brought to reaction temperature in the method according to this invention by passing through the material an electric current of sufficient amperage for this purpose, after said material has been raised to an electrically conducting temperature by a preliminary heating operation.

Prior methods of producing nitrogen compounds by heating in the electric furnace briquettes composed of metallic oxides, or carbonates, and coal have not given favorable results. The present method differs from these prior methods, in that, in the present method the materials are treated in a finely divided or pulverulent condition, and continuously agitated during the reaction, whereby a closer association of the solid material with the nitrogen and a more thorough and rapid reaction is obtained than is possible where the material is treated in the form of briquettes. It has been demonstrated in the working of the present method that lignite is a particularly good reduction coal, and especially lignite which has been freed as much as possible from ash forming constituents. To obtain coal of this character, pulverized lignite is treated with acids, preferably hydrochloric acid.

The accompanying drawings illustrate in a more or less diagrammatic way an apparatus which may be used in carrying out my invention. As the apparatus is not claimed in this application, I have deemed it sufficient merely to show the principal features thereof without attempting to show structural details which those skilled in the art can readily supply.

Fig. 1 shows the apparatus partly in vertical section and partly in elevation.

Fig. 2 is a section on line A—B of Fig. 1. A refractory tube a is adapted to be revolvable mounted and rotated as indicated on the arrows on Fig. 2 by any suitable motor in a manner which will be obvious to those skilled in the rotary furnace art. Tube a is subdivided into an electrically heated chamber b and preliminary heating chambers c and d, the chamber c being located axially of the furnace and the chamber d being an annular chamber surrounding e. The reaction chamber b has mixing devices designed to distribute the material uniformly over the entire cross section of the chamber. These mixing devices consist preferably of a diametrically arranged perforated partition m which rotates with tube a during the reaction, and thereby operate to mix and agitate the material.

The method constituting my invention is carried out as follows:

The furnace is first heated by any suitable exterior source of heat, as by locating a combustion chamber for coal under that part of the furnace constituted by chambers c and d. This heating is continued until the furnace reaches a temperature of about 1200° C., whereupon the pulverized mixed material, for example, a finely pulverized mixture of barium carbonate and lignite, is charged at e into the preliminary heating chamber c. As indicated by the drawing, the furnace may be supported in an inclined position, whereby a continuous longitudinal feeding movement is imparted to the material consequent upon rotation of the furnace. In passing through chamber c the material attains a temperature of about 1000° to 800°, at which temperatures it is electrically conducting. The material now passes to the reaction chamber b, where it forms a part of the circuit between electrodes f, and is electrically heated to the reacting temperature in the presence of ni-
trogen. The reaction may be expressed by the following equation:

$$\text{BaCO}_3 + 4\text{C} + 2\text{N} = \text{Ba(CN)}_2 + 3\text{CO}$$

The nitrogen is supplied to the reaction chamber through tube \( g \) from a collector \( h \) into which it enters from \( k \).

The outflowing hot gases, consisting of carbon monoxide and nitrogen flow through the chamber \( c \) where they give up heat to the incoming pulverized mixture and may be conducted by any suitable means to the exterior combustion chamber to contribute to the exterior heating of tube \( a \).

10 The solid product from the reaction is conducted from chamber \( b \) through tube \( g \) and collector \( h \), and coming in contact with the inflowing current of nitrogen serves to preliminarily heat the latter. The solid product is taken from collector \( h \) through the outlet \( i \) which has a suitable closure.

15 This improved method presents, in comparison with the above mentioned method of heating briquettes, great advantages, which consist in that the absorption of nitrogen takes place much more rapidly and that products can be obtained which have a considerably greater percentage of nitrogen than by the prior method.

According to the method of treating the material in briquette form a mixture of 100 parts of barium carbonate and 50 parts of coal yielded after three hours heating in the electric furnace to 200° C. a final product which contained only 3.2% of nitrogen. Whereas the same mixture treated according to the improved method, in pulverized state, yielded after 3 hours heating at 1200° C. a final product of 7.8% nitrogen.

10 I claim:

1. The method of producing nitrogen compounds, which consists in heating a finely divided mixture of a metallic oxygen compound and coal to a temperature at which the mixture is electrically conducting, and then bringing the mixture to reaction with nitrogen by passing an electric current through the mixture while agitating the same in the presence of nitrogen.

2. The method of producing nitrogen compounds, which consists in heating a finely divided mixture of metallic oxides and coal to a temperature at which the mixture is electrically conducting and then causing the mixture to react with nitrogen by passing an electric current through the mixture while agitating the same in the presence of nitrogen.

3. The method of producing nitrogen compounds, which consists in preliminarily heating a pulverized mixture of a metallic oxygen compound and coal to a temperature at which the mixture is electrically conducting, and then conducting the mixture into a reaction chamber and passing an electric current through the mixture while agitating the same in the presence of nitrogen.

4. The method of producing nitrogen compounds, which consists in preliminarily heating a finely divided mixture of metallic oxygen compounds with lignite which has been substantially freed of ash forming constituents, and then heating the mixture in the presence of nitrogen to reaction temperature while continuously agitating the same.

In testimony whereof I have signed my name to this specification in the presence of two subscribing witnesses.

HEINRICH SPECKETER.

Witnesses:

HANS FLESCHE, GERMER LOTTERHOS.