

UNITED STATES PATENT OFFICE.

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PROCESS OF PRODUCING CYANOGEN COMPOUNDS.

1,329,652.

Specification of Letters Patent.

Patented Feb. 3, 1920.

No Drawing.

Application filed June 12, 1917. Serial No. 174,377.

To all whom it may concern:

Be it known that I, JOHN COLLINS CLANCY, a citizen of the United States, residing at Freeport, in the county of Nassau and State of New York, have invented certain new and useful Improvements in Processes of Producing Cyanogen Compounds, of which the following is a specification.

This invention relates to an improved process of producing substantially pure cyanogen compounds and derivatives thereof; and more especially concerns a cyclic method of fixing nitrogen preferably in the form of an alkali metal cyanid.

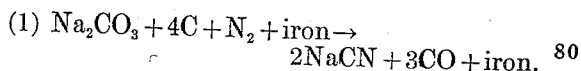
As is well known, attempts, extending over a period of more than seventy years, have been made to effect the fixation of nitrogen economically in the form of cyanogen compounds, and while it was for a long time suspected that it might be possible to fix free nitrogen in the form of alkali cyanid in such a manner as to warrant the commercial development of such a process,—owing to a non-recognition of important factors involving the provision of a suitable catalytic agent in the solid phase and the disposition of the latter with respect to other essential ingredients in the charge in such fashion as to permit of a satisfactory yield of the product sought, together with the vital maintenance of the essential conditions after they were once established, to effect a really satisfactory and copious formation of such product at a reasonably low temperature,—there followed, one after another during this long term of years, a series of practically abortive attempts to solve the problem. The practical failure of these attempts is evidenced by their failure of commercial development, despite the great need of a nitrogen fixation process which could very cheaply convert free nitrogen into such a product as would be substantially at once available for use without further costly treatment after its formation.

The first recognition of the factors aforesaid,—certainly at least in their entirety and in such fashion as to make them apparent to those skilled in the art,—occurs in the patents to John E. Bucher, and notably those numbered 1,091,425 and 1,120,682 and respectively dated March 24, 1914 and December 15, 1914. These Bucher processes have

come into extensive commercial use and in one of its principal aspects, the hereindescribed process or method may be regarded as a further development of this art thus made practicable by said Bucher.

As set forth in the aforesaid Patent No. 1,091,425, cyanid of sodium, for example, may be made at a comparatively low temperature by thoroughly mixing together, in the manner described in said patent, sodium carbonate, pulverulent coke or charcoal and preferably very finely divided iron. The mixture may be used in powdered form or it may be formed into porous briquets; preferably in accordance with the disclosure in the Bucher Patent No. 1,120,682.

Upon passing hot nitrogen gas or even producer gas through this mixture, in a suitable retort, heated to, for example, a red heat or bright red heat, the alkali metal, which forms the base of the carbonate or its equivalent, is combined with the nitrogen and with carbon from the catalyzer, to form alkali metal cyanid; the reaction being represented thus:



Obviously, other alkali-base supplying compounds than the carbonates are available; as for example the hydrates, bicarbonates, etc. Also, as is pointed out in the said Bucher patents, various catalyzers are adapted for use in the process; but iron is to be preferred, principally, on account of its cheapness.

In view of the very full disclosures in said patents to John E. Bucher, for brevity herein, it is obvious that the various equivalents of substances,—such as graphite, carbon-supplying gases, *e. g.*, various hydrocarbons, etc., for the above mentioned powdered coke or charcoal; producer gas, etc., as a source of free nitrogen; not alone iron, but manganese, nickel, etc., or mixtures or compounds of these, as catalyzers;—need not be further referred to herein.

The reactive mass, either in pulverulent form, or, preferably, in the form of porous catalytic briquets, as aforesaid,—after subsection to nitrogen during the proper heat treatment in which the temperature throughout said mass is carefully controlled

to prevent sintering of the catalyzer, or flooding of the reactive carbon-supplying catalytic "surface" in the pores of said briquets,—is, as described in the Bucher patents, efficiently cyanized; a very considerable proportion of the carbonate, or other source of the alkali metal, being converted to alkali metal cyanid; or, by regulation of the supply of carbon, in a known manner,—to, for example, alkali metal cyanamid.

After the so cyanized charge has been cooled, it has been customary in practice to lixiviate it with water to dissolve out the cyanid; but right here was encountered a difficulty which has been the source of trouble and expense. In operating the Bucher processes, while in theory, or on a comparatively small scale, substantially a quantitative conversion of carbonate, for example, to cyanid was possible,—in practice and on a large scale the conversion could be by no means complete; so that there is almost inevitably, under such conditions, a remnant of the carbonate intimately associated with cyanid in the mass, and when the latter is lixivated with water this residual carbonate or the like goes into solution along with the cyanid and contaminates the latter.

What is still worse, from several points of view, is that the metal catalyzer is attacked and if this be iron, as preferred, ferrocyanid of the alkali metal is formed and this also goes into the solution.

Thus impure cyanid is produced, when operating commercially, and, further, there is a loss of catalytic material; not to mention an impairment of that which is left when the residues have been re-pulverized preparatory to re-use in the process.

Furthermore, the lixiviation has to be long continued to effect a satisfactory recovery of cyanid from the mass, which itself is costly; while an aqueous solution of the impure product is formed, involving a prolonged and costly drying operation.

My problem then was to further develop the Bucher processes in question, so as to render the whole, in effect, cyclic,—by winning the cyanid from the cyanized charge or mass without destroying or impairing the catalyzer, and preferably without dissolving out the carbonate, or its equivalent, or forming or dissolving out already formed contaminating by-products. It was also aimed to eliminate the costly lixiviation with water and the subsequent tedious treatment of the aqueous solution.

This problem I have solved and the various difficulties aforesaid have been completely overcome by my discovery of a solvent for alkali metal cyanids which is wholly incapable of attacking the iron, nickel, manganese or equivalent catalyzer, either to dissolve the same, or to oxidize,

or otherwise form a compound or compounds therewith; while, furthermore, said solvent possesses practically no solvent action upon the alkali metal carbonates, or other preferred sources of the base of the cyanogen compound to be formed as per said Bucher processes, or upon any of the solid by-products incident thereto.

This action is the more peculiar since while an alkali metal cyanid may be dissolved in liquid ammonia, which is the solvent preferred, the alkaline-earth metal cyanids cannot. On the other hand the alkali metal cyanamids and cyanates are thus soluble in this reagent; as are also, for example, sodium chlorid, sulfid, hypochlorite and chlorate. Further, it is somewhat remarkable that while barium cyanid, for example, is insoluble in liquid ammonia,—barium chlorid, contrawise, is soluble; and while nickel in the metallic state is unaffected by this agent,—nickel chlorid, nickel acetate, etc., are soluble. Similarly, cadmium chlorid, for example, is also soluble; as are, too, zinc chlorid, manganese chlorid and many other salts.

When the Bucher processes described in said patents numbered 1,091,425 and 1,120,682 are followed up to the stage where the charge has become well cyanized,—not only will there be sodium carbonate, for example, present in the reaction residues; but there also may be present, when operating commercially, sodium oxid and sodium hydrate, the latter probably as a result of the presence of some water vapor, either formed during the course of the operation or initially present either in the charge or in the gases supplied to the latter.

Here again the ammoniacal solvent closes its peculiar value in connection with said processes, since even these impurities are all substantially insoluble in it; with the result that when the said reaction residues are treated with ammonia in the manner hereinafter described,—an alkali metal cyanid is obtained which is indeed remarkable for its purity; being almost chemically pure. Carbon, it may be added, is of course, insoluble in liquid ammonia.

While the solvent action of ammonia upon the alkali metal cyanids has been known, or suspected, for nearly twenty years, this compound seems not to have been available for commercial production of the product sought probably on account of the fact that chlorids and sulfids are, as above pointed out, so soluble in it that the obtaining of an uncontaminated product by means thereof was rendered prohibitively costly.

Thus, in the British patent to Craig, No. 11,912 of 1900, there is a disclosure of the use of liquid ammonia in the obtainment of cyanid; but the reaction residues, which are

described in this patent as being treated with this reagent, are not those resulting from a cyanid forming reaction; but on the contrary, the "crude cake" referred to in said patent, must first be treated to, at least in theory, render the injurious soluble impurities, insoluble in liquefied ammonia.

That this treatment appears to have been successful in practice does not satisfactorily appear, while from the fact that it is not today in commercial use, in spite of the demand for pure cyanid, raises a strong presumption, in view of the lapse of time, that this process was not a practicable one.

On the other hand, in the present process, there is the special relationship between the cyanid producing step of the process, *per se*, and the step of at once recovering pure product from the reaction mass, which has enabled the production of alkali cyanid cheaply and in large quantities; while said present process owes much of its efficiency to its cyclic character; there being no waste of material, catalytic or otherwise; nor prolonged lixiviation of the cyanized reaction products required.

In brief the present process preferably comprises the formation of the cyanid of an alkali metal, such as sodium, from a suitable compound of such metal, to wit, sodium carbonate,—which compound and all reaction products thereof, save the product sought, are wholly or very nearly completely insoluble in the ammoniacal reagent subsequently used. The cyanogen compound forming reaction is preferably effected at such a temperature as will afford a copious yield of said cyanogen compound without wastefully vaporizing the carbonate or the like and without impairing the finely divided catalyzer preferably employed.

As to the source of the carbon for the reaction,—this is material principally in so far as the maintenance of an adequate supply of carbon to the catalyzer is concerned and further in that sulfur or like impurities,—which might result in the formation of soluble alkali compounds or the like,—are excluded.

The material in which the cyanid or the like is to be formed is preferably briqueted, and here again the formation of the briquets should most preferably involve the introduction of no sulfur or the like.

It is for this reason that the procedure of said Bucher in using the insoluble carbonate or like salt, itself as the binder for said briquets, is of great value when incorporated as a step of the present process.

The step of cooling the cyanized briquets, as described in the Bucher Patent No. 1,120,682, also most advantageously knits into the steps of the herein described improvement; since, as the solvent in question is one that volatilizes at a relatively low

temperature, under normal pressure conditions, it could not be used to advantage upon a hot mass.

The cyanized charge, therefore, is cooled sufficiently to prevent undue evaporation of said solvent and when sufficiently cool, is subjected,—preferably in a receptacle distinct from the retort in which the cyanid or the like was formed,—to the selective solvent action of liquid ammonia; a stream or quantity of the latter preferably being permitted to pass through or permeate the mass, to leach out the cyanid or the like therefrom.

In this connection it may be well to emphasize the importance of having the cooled cyanized material in a form which affords a multiplicity of interstices, extending substantially therethroughout, to the end that an actual stream of the liquid ammonia may be passed through the mass, as aforesaid. The liquid ammonia should permeate all parts of the material under treatment and, as the cyanid or the like goes very readily into solution in said ammonia, a practically saturated solution is speedily formed in any given part of the mass, if the liquid ammonia is stagnant or idle, or nearly so.

Obviously if such saturated ammonia is not removed prior to the steps of draining and evaporating the solvent, then upon evaporation of the liquid ammonia, the cyanid will deposit out very nearly where it was when it went into solution.

Where, however, the charge to be cyanized is properly briqueted in accordance with the disclosure of said Bucher, hereinbefore referred to, and the briquet formation is, after cyanization, then utilized during the extraction step of the process, it is evident that said formation affords the desired interstices; so that the stream of liquid ammonia can pass freely through it and the development of idle saturated masses of liquid ammonia is avoided.

As will be more specifically pointed out hereinafter, there are still other advantages residing in the treatment by liquid ammonia of a cyanized charge in the form of briquets, or the like.

The container for the mass to be leached may be iron; but since the liquid ammonia possesses the ability to penetrate the pores of many cast iron receptacles, care must be exercised to either use a practically non-porous iron or the like, or to allow the pores of the latter to choke up with cyanid, for example, if waste of the solvent is to be avoided.

The leached mass is then allowed to drain and may thereafter be gently heated to volatilize any remaining ammonia present therein; the gaseous ammonia being collected in any suitable manner for ultimate return to the process.

It is surprising how quickly the residues may be completely dried, and this especially when one bears in mind the tedious drying necessary after lixiviation with water as per the Bucher disclosures.

The dried residues are at once available for return to the process; being, especially if in briquet form, repulverized to have added thereto sufficient carbonate or the like and carbon, to replace that converted into cyanid, if that be the product sought. The catalyzer is wholly unaffected by the ammonia and its surface (or multitudinous surfaces) is unimpaired, while no loss there- of occurs as when aqueous lixiviating liquids are used.

Above all, also, no ferro-cyanid or the like is formed and the solvent contains practically nothing else but the cyanogen compound to be produced; while, further, this solvent reclaims practically all of the cyanid or the like, the percentage left in the residual mass being really negligible.

The cyanized solvent is, of course, collected and is then preferably subjected to moderate heat to drive off the ammonia and leave the alkali cyanid, for example, as a white, finely divided mass of almost chemically pure crystals; much like granulated sugar in appearance.

It is believed that this product is distinctively new, as it seems to possess the property of entering into solution in water with great ease, whereas, to the best of my knowledge, the alkali cyanid now on the market which is available for ore treating operations, dissolves in water quite slowly; and certainly very slowly as compared with the product aforesaid.

To return to the process, proper, the evaporated ammonia is of course conserved, being collected and recondensed preparatory to its further use in the process; air being preferably carefully excluded from the ammonia circuit.

In fact it is most desirable to exclude air from the retort in which the cyanogen compound is formed and especially to preserve the cyanized mass from exposure to any oxidizing agent unless the product sought is cyanate; since when the mass of heated cyanized briquets, for example, is exposed to the air, cyanate will form therein and this, as above stated, is soluble in the leaching agent.

After the cyanized mass has been properly cooled, oxidation of the product does not readily occur; but even then, in order to save ammonia and insure purity of the product, air should be excluded as far as practicable; the entire operation preferably being conducted with careful exclusion of air from the time that the reactive pulverulent or briqueted charge is introduced into the retort until the crystalline alkaline

cyanid, or the like, is recovered from the ammonia.

When, as is to be preferred, the charge is in briqueted form, a further advantage is derived from the ammonia treatment subsequent to cyanization and in combination therewith; since the briquets after such treatment, having parted with their cyanid content, are not,—as one might at first think would be the case,—rendered shapeless or more or less plastic or fragile; but, to the contrary, they are found to have become quite hard and brick-like; so that they may be dropped from a moderate height without breaking, and are hence, subsequently, more readily handled.

Their appearance also seems to be modified by subjection to the ammonia bath; as they become somewhat more shiny or lustrous in appearance. This phenomenon seems to convey a hint that some ammoniacal compound, possibly involving cyanogen, is formed; but if this be so, there is left in said briquets at most but a trace thereof, after the solvent has been completely removed.

In conclusion, it should be observed that it is extremely desirable to use the same catalytic material, to wit, iron,—over and over again; since it has been found in practice that after it has been used as a catalyzer once or more, especially after treatment as aforesaid, its condition for the purpose intended seems to be better than that of, for example, finely powdered iron which has never been so used or treated. The importance of using ammonia as a leaching reagent, in the process in question, rather than water, is hence additionally enhanced; there being no deleterious action thereof on the catalyzer, whatsoever, but rather, apparently, a beneficial one; and this in contradistinction to the deleterious and destructive action of such a fluid as water, which forms hydrates of iron, ferro-cyanids, etc. The step, too, of recovering the cyanid from the cyanized charge by means of liquid ammonia has the added advantage that the formation of the cyanid solution is not alone almost instantaneous, but it furthermore, is effected at room temperatures, or lower.

The low temperature of the operation obviates any necessity for the application of heat to the solvent or to the charge at this stage of the process and correspondingly does away with the necessity for providing heating appliances, other than for the subsequent evaporation of the solvent; and even this latter is vastly less expensive than is the evaporation off of a large amount of water.

Thus, for example, in removing the residual ammonia from the leached charge after the latter has been drained as aforesaid, a gentle heat will in the course of half

an hour render the mass practically perfectly dry. The evaporated ammonia is preferably, in every case, passed into a suitable refrigerating apparatus to effect re-

condensation thereof, preparatory to its return to the receptacle in which it may be stored until it is to be used for further leaching out of cyanid or the like, in the manner above described.

Finally, I may state that to the best of my knowledge, my invention, herein described, is the first to afford a practical nitrogen-fixation process in which pure cyanid of an alkali metal may be synthetically produced in a single heating operation, at a moderate temperature (red heat) adapted to conserve the life of the apparatus employed,—coupled with a simple, inexpensive and almost instantaneous selective leaching operation which does not impair the reaction residues and which when followed by the third step of evaporating the selective solvent off at a gentle heat (212° F, for example), yields the product sought in extraordinarily pure and finely divided condition. Moreover, the reaction residue is in the best possible condition to be supplemented by fresh alkali compound and carbon for reuse in the process.

It is, too, believed that the present invention is the first catalytically-operating, cyanid-forming process, in which the catalyst, unimpaired, may be cyclically utilized, to form substantially uncontaminated cyanid, and as pointed out, in effect, in the said patents to John E. Bucher, unless a catalyst is used in the cyanid formation, the temperature requirements in so far as the conversion of such an inexpensive substance as soda ash, or sodium carbonate, into cyanid, are concerned,—become prohibitive for practical purposes on account of the inability of the necessary apparatus to stand up under temperatures materially in excess of a bright red heat.

Electric furnaces intended to effect the direct synthetic formation of cyanid at upward of 1800° C. have been proposed; but so far as I am aware they exist only on paper.

Having thus described my invention, what I claim is:

1. The process of obtaining alkali metal cyanid, which comprises catalytically effecting a synthetic reaction, in which participate carbon, free nitrogen and a compound which constitutes the source of the alkali metal base of the cyanid to be formed, the catalyst for said reaction being a metal in finely divided condition, cooling the reaction residues, and thereafter subjecting the products of said reaction, while cool and intimately associated with said catalyst, to the action of a solvent for said alkali cyanid which solvent is incapable of chemically re-

acting upon said catalyst to form any compound therewith at the temperature of the operation, recovering said alkali cyanid in substantially pure condition from said solvent, and subjecting the remaining and undissolved residues, including said catalyst, to further participation in said reaction.

2. The process of obtaining alkali metal cyanid, which comprises catalytically effecting a synthetic reaction, in which participate carbon, free nitrogen and a compound which constitutes the source of the alkali metal base of the cyanid to be formed, the catalyst for said reaction being iron in finely divided condition, cooling the reaction residues, and thereafter subjecting the products of said reaction, while cool and intimately associated with said catalyst, to the action of a solvent for said alkali cyanid which solvent is incapable of chemically reacting upon said iron catalyst to form any compound therewith at the temperature of the operation, recovering said alkali cyanid in substantially pure condition from said solvent, and subjecting the remaining and undissolved residues, including said catalyst, to further participation in said reaction.

3. The process of obtaining an alkali metal cyanogen compound, which comprises forming a briqueted charge from a mixture of pulverulent materials which includes a compound capable of acting as the source of the alkali metal base of the cyanogen compound to be formed, intimately mixed with a carbonaceous substances, the briquets of said charge being both firm and porous, cyanizing said briquets by heat treatment while subjecting them to the action of free nitrogen, cooling the still briqueted but now cyanized charge, and so separating the alkali metal cyanid therefrom as to leave said charge in briqueted form, by treating said charge with a selective extracting agent for said cyanid, the briqueted form and porous condition of said charge favoring said separation.

4. The process of obtaining an alkali metal cyanogen compound, which comprises forming porous catalytic briquets by using as a binder for the catalytic material to be briqueted, an alkali compound which constitutes the source of the alkali metal of the cyanogen compound to be obtained, subjecting said briquets to heat treatment while supplying carbon and free nitrogen to the surface of said catalytic material exposed in the pores of said briquets, so conducting said treatment as to maintain said surface effective to cause the formation of said cyanogen compound through the chemical reaction of said alkali compound with said free nitrogen and carbon, cooling the briquets and separating therefrom the so formed cyanogen compound through the instrumentality of a substance which is in-

capable of disintegrating said briquets and which leaves unimpaired the catalytic material therein.

5 The process of obtaining an alkali metal cyanogen compound which comprises forming porous catalytic briquets by using as a binder for the catalytic material to be briqueted, an alkali compound which constitutes the source of the alkali metal of the
10 cyanogen compound to be obtained, subjecting said briquets to heat treatment while supplying carbon and free nitrogen to the surface of said catalytic material exposed in the pores of said briquets, so conducting
15 said treatment as to maintain said surface effective to cause the formation of said cyanogen compound through the chemical reaction of said alkali compound with said free nitrogen and carbon, cooling the briquets and separating therefrom the so
20 formed cyanogen compound through the intermediacy of a substance which acts selectively upon the reaction products present in said briquets, to eliminate therefrom substantially only said cyanogen compound,
25 while maintaining the integrity of said briquets and leaving them resistant to fracture.

6. The process of obtaining an alkali metal cyanogen compound, which comprises
30 subjecting porous briquets to heat treatment to effect, within the pores of said briquets, a cyanogen compound forming reaction in which participate carbon, an alkali metal and free nitrogen, cooling said briquets, and
35 separating the so formed cyanogen compound therefrom through the intermediacy of a substance which acts selectively upon the reaction products present in said briquets while maintaining the integrity of said
40 briquets and leaving them resistant to fracture, to facilitate their subsequent handling.

7. The process of obtaining an alkali metal cyanogen compound, which comprises catalytically effecting a reaction in which
45 participate the elements carbon, an alkali metal, oxygen and free nitrogen, the products of said reaction including the alkali metal cyanogen compound to be obtained, intimately commingled with an oxid of said
50 alkali metal, the catalyzer through the instrumentality of which said reaction was effected, a compound which constitutes the source of said alkali metal, and carbon, cooling the said reaction products while excluding
55 air therefrom, and subjecting said cooled products to the action of ammonia.

8. The process of obtaining an alkali metal cyanogen compound, which comprises catalytically effecting a reaction in which
60 participate the elements carbon, an alkali metal, oxygen and free nitrogen, the products of said reaction including the alkali metal cyanogen compound to be obtained intimately commingled with an oxid of said
65 alkali metal, the catalyzer through the in-

strumentality of which said reaction was effected, a compound which constitutes the source of said alkali metal, and carbon, cooling the said reaction products while excluding air therefrom, subjecting said cooled
70 products to the action of ammonia in a circuit of the latter from which air is substantially excluded, and recovering from said circuit the product sought.

9. The process of obtaining an alkali
75 metal cyanogen compound, which comprises effecting a reaction in which participate the elements carbon, an alkali metal, oxygen and free nitrogen, the products of said reaction including the alkali metal cyanogen com-
80 pound to be obtained, intimately commingled with an oxygen compound of said alkali metal and carbon, cooling the said reaction products, and subjecting them to the action of a stream of liquid ammonia
85 while said products are in a form which affords interstices through the mass thereof, to extract said cyanogen compound, said interstices permitting access of said stream of liquid ammonia to substantially all parts
90 of the material under treatment.

10. The process of obtaining an alkali metal cyanogen compound, which comprises effecting a reaction in which participates the elements carbon, an alkali metal, oxygen
95 and free nitrogen, the products of said reaction including the alkali metal cyanogen compound to be obtained, intimately commingled with an oxygen compound of said alkali metal and carbon, cooling the said
100 reaction products, and subjecting them to the action of liquid ammonia while maintaining through the mass thereof a multiplicity of interstices of size and extent sufficient to insure free and rapid penetration
105 of said liquid ammonia into substantially all parts of the material under treatment.

11. The process of obtaining an alkali metal cyanogen compound, which comprises effecting a reaction in which participate the
110 elements carbon, an alkali metal, oxygen and free nitrogen, the products of said reaction including the alkali metal cyanogen compound to be obtained, intimately commingled with an oxygen compound of said
115 alkali metal and carbon, cooling the said reaction products, and efficiently extracting said cyanogen compound therefrom by effecting a sufficiently free flow of liquid ammonia, through substantially all parts of the
120 material under treatment, to insure against the formation within the bulk of said material of idle masses of liquid ammonia holding in solution a high content of the cyanogen compound to be extracted.

12. The process of producing a cyanogen
125 compound of an alkali metal which comprises effecting a reaction in which said compound is formed at a temperature sufficiently high to at least partly fuse a carbonate of
130

said alkali metal, while utilizing the fused carbonate in said reaction as the source of the alkali metal which forms the base of the cyanogen compound sought, said carbonate, just previous to its fusion as aforesaid, being substantially free from water, cooling the non-gaseous reaction residues to render all of them solid, treating said residues with a non-aqueous solvent for said alkali cyanogen compound which is substantially incapable of chemically changing or dissolving any of the solid alkali carbonate which is present at such time in said residues, removing said solvent from the undissolved residues, with said cyanogen compound in solution therein, to leave said undissolved residues substantially dry, and returning to said process the residual alkali carbonate together with an additional quantity of said carbonate, to replace that consumed.

13. The process of producing a cyanogen compound of an alkali metal which comprises effecting a reaction in which said compound is formed at a temperature sufficiently high to at least partly fuse a car-

bonate of said alkali metal, while utilizing the fused carbonate in said reaction as the source of the alkali metal which forms the base of the cyanogen compound sought, said carbonate, just previous to its fusion as aforesaid, being substantially free from water, cooling the non-gaseous reaction residues to render all of them solid, treating said residues with a non-aqueous solvent for said alkali cyanogen compound which is substantially incapable of chemically changing or dissolving any of the solid alkali carbonate which is present at such time in said residues, removing said solvent from the undissolved residues, with said cyanogen compound in solution therein, and returning to said process the residual alkali carbonate still mixed with the remaining reaction residues.

In testimony whereof I have affixed my signature in the presence of two witnesses.

JOHN COLLINS CLANCY.

Witnesses:

J. M. ARCHER,
G. R. MORRIS.