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PROCESS FOR PRODUCTION OF NITROCELLULOSE PROPELLANTS

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12 Claims. (Cl. 264—3)

This application is a continuation of our copending application Ser. No. 588,065, filed Apr. 12, 1945, now abandoned.

The present invention relates to propellants and more particularly to a new and improved method of fabricating double base propellants, especially those of relatively large dimensions such as the thick webbed grains employed as the fuel in the jet assisted take-off of aircraft, large rockets and other jet propulsion applications.

Broadly speaking, the object of the invention is the production of propellant powders by an improved method that requires only readily available raw materials, simple and inexpensive equipment and a minimum of time and labor.

A more particular object is the production of thick web double base propellant grains from commercially available thin web propellant powders, by a process that obviates the need for either pressing, extruding or rolling equipment of any type.

A still further object is to provide a process of the character described, which lends itself to the production of large grain rocket propellants of new and unusual designs, as well as to those of conventional types.

Other objects and advantages of the invention will become apparent as the description progresses.

The process of the present invention by which the foregoing objects may be attained, represents a method of fabrication that is believed to be radically new in the manufacture of propellants. The invention is based upon the discovery that propellants of excellent ballistic properties may be produced by a process involving a simple casting operation, using two primary components, one being a liquid (hereinafter referred to as the liquid component) and the other a solid (hereinafter referred to as the solid component).

The liquid component comprises a liquid that is capable of plasticizing nitrocellulose. The solid component comprises thin webbed, gelatinized nitrocellulose that is capable of absorbing a substantial proportion of the liquid component mentioned above. The gelatinized nitrocellulose is preferably in a finely divided or granular form and it may or may not contain plasticizers, stabilizer or other conventional propellant components, but in any event it should be (a) thin webbed and (b) capable of absorbing a substantial proportion of the liquid component mentioned above.

In carrying out the process of the present invention, the liquid and solid components mentioned above are mixed together in any suitable manner to form a substantially bubble-free mixture; that is, a mixture in which the interstices between the solid components are substantially completely filled by the liquid component. The ratio or proportion of liquid-to-solid in the mixture should be such that the cast product formed therefrom, as explained hereinafter, possesses the burning properties characteristic of a propellant.

The process of the present invention is based on the discovery that, if a bubble-free mixture of the foregoing character is heated while in a suitable mold to a temperature sufficient to accelerate the rate of absorption of the liquid by the solid, within a fairly reasonable time the individual units of gelatinized nitrocellulose will swell, become tightly packed together and ultimately coalesce to form a unitary, non-porous, cast solid mass. The cast product thus obtained, trued to proper dimensions or otherwise machined and/or finished as desired, possesses the substantially uniform burning properties characteristic of a satisfactory propellant. The method may of course be used to fabricate propellants having any web thickness greater than that of the solid component used in their manufacture. It is, however, particularly useful for fabricating propellants for applications requiring grains of relatively large dimensions or thick webs.

I. THE SOLID COMPONENT

As indicated above, the solid component constituting one of the starting materials in the process of the present invention, comprises thin webbed (preferably granular) gelatinized nitrocellulose that is capable of absorbing a substantial proportion of the nitrocellulose-plasticizer employed as the liquid component in the present process. Thus the solid component may consist of either finely divided single base or double base propellant powder having a substantial capacity for absorbing the liquid plasticizer.

The foregoing types of powders are readily available and may be exemplified, respectively, by "IMR" single base powder and double base "Ball Powder." Such powders are available in various granulations within the range from approximately 0.005 to 0.060 inch in diameter. Of these, the granular double base propellant powders of relatively low (e.g., 12%) liquid nitric ester content constitute attractive casting powders inasmuch as they form hard, homogeneous and tough cast grains after a relatively short cure at moderate temperatures when used in the process of the present invention. However, in addition to the above mentioned commercially available granulated types of powders prepared by special granulating processes, one may use, if desired, a thin-webbed extruded single or double base propellant which has, for example, been machine-cut into finely divided particles, e.g., machine-cut solid cylinders of about 0.030 inch diameter and about 0.30 inch in length.

II. THE LIQUID COMPONENT

In those instances where the finished cast propellant is to be sufficiently hard and strong to be burned, for example, in a rocket motor as a self-supporting grain, it should of course have a nitrocellulose content roughly equal to that of conventional solvent-less and solvent-types of rocket propellants. Since the diffusion rate of a solvent in a plastic composition decreases rapidly with decreasing plasticizer content, the preparation of a satisfactory cast propellant in accordance with the present process requires the use of an active solvent, swelling agent, or plasticizer for nitrocellulose; that is to say, a solvent, swelling agent or plasticizer that is sufficiently active to insure substantial completion of the curing process within a reasonable time in spite of the high nitrocellulose content (and therefore low plasticizer content) of the finished product. The choice of the liquid component for use in the process of the present invention is therefore important.

Generally speaking, the liquid component for use in accordance with the present invention should possess the following characteristics:

(1) It should be an active solvent, plasticizer and/or swelling agent for the nitrocellulose;

(2) It should form with the solid component a finished propellant having the burning properties characteristic of a satisfactory propellant;

(3) It should be of sufficiently low volatility to be retained in the cured and finished propellant; and

(4) Preferably but not necessarily it should be reasonably safe to handle in manufacturing operations, from the point of view of explosive sensitivity.

From the foregoing it will be evident that a large number of compounds and mixtures of compounds are available for use as the liquid component in accordance with the present invention. Generally speaking, the liquid component may consist of one or more explosive, or one or more non-explosive, solvents, swelling agents or plasticizers for nitrocellulose; or a mixture of one or more explosive and one or more non-explosive solvents, swelling agents or plasticizers. Thus the liquid component may comprise one or more liquid nitric esters or other explosive plasticizer for nitrocellulose; for example, nitroglycerine, ethylene glycol dinitrate, diethylene glycol dinitrate, N-methyl-2-nitroxyethylnitramine and the like. In the alternative it may consist of one or more of the well known non-explosive plasticizers, for example, triacetin, dimethyl phthalate, butyl tartrate, and the like. However, the preferred liquid component, possessing all the characteristics listed above, comprises a liquid mixture of (1) a high potential or explosive nitrocellulose-plasticizer as the major constituent and (2) a non-explosive nitrocellulose-plasticizer as the minor constituent, the latter serving (a) to phlegmatize the explosive plasticizer (thereby rendering the mixture safer to hande) and (b) to increase somewhat the rate of gelatinization or swelling of the nitrocellulose by the explosive plasticizer.

Among the available non-explosive nitrocellulose-plasticizers which may be used in the liquid component of the present invention, the following illustrative types of compounds may be mentioned; namely, diethyltartrate, dibutyltartrate, tributylcitrate, butyl levulinate, camphor, isophorone, triethylphosphate, dimethylformamide, butylacetimide, triacetin, dimethylphthalate and the like. The preferred non-explosive solvents comprise triacetin (used for example in an 80–20 mixture of nitroglycerine and triacetin) or dimethylphthalate (used for example in a 70–30 mixture of nitroglycerine and dimethylphthalate).

III. THE SOLID-LIQUID RATIO

The solid-to-liquid ratio of the bubble-free mixture to be cured in the mold may be varied within relatively wide limits, depending in part on the mechanical and ballistic properties desired in the finished product. As previously stated, in order to insure a bubble-free mixture, sufficient liquid should be present substantially completely to fill the interstices between the solid components. In addition, the ratio of liquid to solid should be so adjusted that the finished cast product possesses the burning properties characteristic of a propellant, which means, inter alia, that the cast product should contain a substantial proportion of nitrocellulose. Generally speaking, satisfactory mixtures may be made from 35 or more parts by weight of the solid component and 65 or less parts by weight of the liquid component. The actual proportions selected in any specific case will of course depend in part on the physical structure and composition of the solid component used, the composition of the liquid component employed, the method selected to form the cast mixture, the ballistic properties desired in the finished propellant, and upon other factors.

IV. THE CASTING OPERATION

The casting operation involves the heating of a substantially bubble-free mixture of the solid and liquid component, disposed within a mold, heating being continued at a temperature and for a time sufficient to bring about the formation of a unitary, cast solid mass having satisfactory mechanical and burning characteristics. In effecting this step of the process, various expedients may be used to render the mixture in the mold substantially bubble-free prior to the time the thermal treatment or curing step is initiated. In some cases it may be convenient to pour the preformed mixture into the mold, or to prepare the mixture in the mold, and thereafter to remove entrapped air bubbles by gently stirring or vibrating the mixture from time to time, permitting the entrapped gas to rise to the surface and escape. In general, however, it is preferable to subject the mixture and/or its components to reduced gas pressure (evacuation) in order to hasten the separation of entrapped air. Additional expedients may also be used; for example, a combination of vibration and evacuation, or of evacuation both before and after the mixture or the components of the mixture are placed in the mold.

In this connection, it may be mentioned that, if the mixture is rendered bubble-free by subjecting the mixture to reduced gas pressure, it may be desirable to place the mixture in a vessel of relatively large volume compared to the volume of the mixture inasmuch as the mixture sometimes tends to "boil" or degas vigorously under vacuum. A satisfactory technique is therefore to prepare a sufficient quantity of mixture for several casting operations, using a vessel of ample size to allow for possible "boiling." The mixture is then subjected to reduced pressure and after the air bubbles have been substantially completely removed, the degassed mixture is carefully placed in the individual molds. Thereafter the molds may likewise be subjected to reduced pressure in order to remove any air bubbles clinging to the walls of the mold. In small scale trials it has been found that the time required completely to remove entrapped air bubbles in this manner has ranged from 15 to 30 minutes, depending on the viscosity and depth of the slurry in the container.

After the mold has been filled with a substantially gas-free mixture, the curing operation is carried out simply by heating the contents of the mold to a moderate temperature for time sufficient to produce a unitary, non-porous cast solid mass having the burning properties characteristic of a satisfactory propellant. The time required for the completion of this process will of course depend on the nature and proportions of the components, the temperature of cure and other factors. In general, however, cure periods of one or two days at a temperature of the order of say 60–75° C. have usually been found to be adequate. As previously indicated, cast propellants obtained from single base powders as the solid component in some cases may require a longer cure period while those produced from double base powders of moderately high nitroglycerine content (e.g., 20%) may require a cure of only about one day at about 60° C. in order to produce a satisfactory casting.

Where the mold employed consists of a metal container such as a steel or preferably a non-sparking metal tube, the cured grains may readily be removed by chilling the assembly and then lightly tapping the mold with a wooden mallet. The cast grain readily separates from the metal mold under these conditions, and after removal from the mold the casting may then be finished in any desired fashion to produce the final propellant grain.

The shape of the mold will of course depend on the type of finished grain desired. Simple tubular molds are very satisfactory and if desired they may be provided with co-axial rods in order to form centrally perforated grains. Generally speaking, however, the type and shape of the mold will be determined largely by matters of convenience, having regard to the type of grain to be produced.

V. RESTRICTED BURNING GRAINS

Restricted burning grains may be produced from the cast product formed in the manner described above, by coating any desired part of the surface of the grain with a suitable fire retarding coating composition containing, for example, cellulose acetate, ethyl cellulose, polymethyl methacrylate or other film-forming composition of suitable character. Another method of forming restricted burning grains that is particularly adapted to the produc-

tion of end-burning types, is to cast the grain in a mold which itself consists of a fire retarding or fire resisting plastic, such as cellulose acetate, ethyl cellulose, methyl methacrylate or other non-burning plastic material, preferably one having approximately the same coefficient of expansion as that of the finished double base powder. In this case, the plastic mold adheres tightly to the surface of the cast grain which is subsequently used as a propellant without removal from the mold. In this fashion charges which will undergo end-burning (i.e. a "cigarette-type" of restricted burning) may readily be produced. In some instances it may be desirable to cover the restrictive coating or plastic with a supplementary material such as pressure-adhesive tape in order to protect the restricted burning grain from damage during burning.

VI. ADVANTAGES

The simplicity of the present process will be readily apparent to those skilled in the art and it is unnecessary to elaborate on the fact that the invention enables the production of double base propellants of standard as well as unusual granulations and types, from readily available raw materials, in simple inexpensive equipment with a minimum of time and labor. It will also be apparent that the casting process enables the incorporation of various supplementary solid bodies or masses in the grain with the formation of interesting new types of heterogeneous composite propellants.

Another advantage of the present invention lies in the minimal manufacturing hazards that are involved. The solvent mixtures, constituting the preferred liquid component in the process of the present invention, are significantly less sensitive to impact than an undiluted high potential plasticizer, and this is particularly so in the case of those liquid components which contain 30% or more non-explosive plasticizer. This will be evident from Table I, which provides a comparison of the impact sensitivity of nitroglycerine with that of a number of typical solvent mixtures.

TABLE I

Liquid component:	Impact sensitivity, cm.
Nitroglycerine (NG)	6.3
80–20 NG-diethyltartrate	39
70–30 NG-diethyltartrate	170
80–20 NG-dibutyltartrate	69
70–30 NG-dibutyltartrate	206
80–20 NG-tributylcitrate	125
70–30 NG-triacetin	151

In order more clearly to disclose the nature of the present invention, several specific embodiments will hereinafter be described in considerable detail. It should, however, be understood that this is done solely for the purpose of illustrating, by means of concrete examples, the basic principles involved and that such examples are not intended either to delineate the breadth of the invention or to restrict the scope of the appended claims.

EXAMPLE I

The solid component or "casting powder" employed was a double base Ball Powder containing approximately 12% nitroglycerine. The powder as received from the manufacturer was in the form of approximately spherical balls of about 0.020 to 0.030 inch in diameter.

The liquid component or "casting solvent" employed consisted of a 69–30–1 mixture of nitroglycerine, dibutyltartrate and ethyl centralite.

About 55 parts by weight of the casting powder was added to about 45 parts by weight of the casting solvent and the resulting mixture in a large vessel was placed under vacuum. The mixture "boiled" vigorously for about 15 minutes. The mixture, at atmospheric pressure, was then carefully ladled into steel, tubular-shaped molds of 2⅝ inches in diameter. The filled molds were subject to reduced pressure for 15–30 minutes. The molds were then placed in an oven at atmospheric pressure and maintained at about 60° C. for about two days.

At the end of this time the mixture had set to a tough, hard substantially homogeneous solid. The molds were chilled to −40° C. and the grains removed therefrom by gently tapping the mold with a wooden mallet. The grains were machined to remove the soft material at the top (in this case, the grains were prepared with an excess of liquid) and a ⅜ inch hole was drilled axially through the grain to produce a centrally perforated propellant. The ends of the grain were restricted with sheets of cellulose acetate, thereby producing a neutral burning perforated grain.

Representative pressure-time and ballistic data obtained with a series of grains prepared in the foregoing manner are given in Table II.

TABLE II

Sample	A	B	C	D	E
Length (in.)	2.92	2.80	2.88	2.72	2.96
Charge weight (gms.)	344	331	340	325	345
Burning time (sec.)	1.81	1.62	1.56	1.66	1.56
Av. pressure (p.s.i.)	690	510	-------	1,020	1,090
Burning rate (in./sec.)	0.28	0.30	-------	0.35	0.32
K	362	377	453	474	503

K=Area of powder divided by area of throat of the rocket motor.

EXAMPLE II

The casting powder consisted of machine cut cylinders, about 0.030" x 0.030" having approximately the following composition:

	Percent
Nitrocellulose	84
Nitroglycerine	15
Centralite	1
Carbon black (added)	0.02

The casting solvent had the following composition:

	Parts
Nitroglycerine	75
Triacetin	25
Centralite	1.5
Carbon	0.2

The liquid and solid components were evacuated separately, then mixed to form a slurry, the slurry evacuated for 10 minutes and then poured into suitable molds. The filled molds were evacuated for 15 minutes to remove trapped air and then cured at about 60° C. at atmospheric pressure.

After one day's cure at 60° C. there was produced a homogeneous product having the following approximate composition:

	Percent
Nitrocellulose	47.5
Nitroglycerine	41.2
Triacetin	10
Centralite	1.3

The pressure-time ballistic data for this powder are given in Table III.

TABLE III

	Sample				
	F	G	H	I	J
Type Grain	Perforated end coated	End burning	End burning	End burning	End burning
Length (in.)	2.34	2.05	2.00	1.95	2.05
½ Web (in.)	.465	-------	-------	-------	-------
Charge weight (gms.)	275	218	200	206	216
Burning time (sec.)	0.24	4.3	4.8	3.7	4.2
Av. pressure (p.s.i.)	3,300	1,450	1,300	1,750	1,250
Burning rate (in./sec.)	1.92	0.48	0.42	0.53	0.49
K	520	489	445	490	420
Mold	(1)	(2)	(2)	(2)	(2)

[1] Steel.
[2] Cellulose Acetate.

EXAMPLE III

The casting powder consisted of machine-cut cylinders, about 0.030" x 0.030", having approximately the following composition:

	Percent
Nitrocellulose	84
Nitroglycerine	15
Centralite	1
Carbon black (added)	0.02

Three casting solvents were used, as follows:

	K	L	M
Nitroglycerine	80	70	60
Dimethyl phthalate (DMP)	20	30	40
Centralite (added)	1.5	1.5	1.5

The casting technique was as follows. The casting powder and liquids were separately evacuated. The powder was then poured into the liquid with stirring. The resulting slurry was evacuated a short time (10 minutes), and then poured into the containers (cellulose acetate cups). The cast mixture was re-evacuated a short time (10 minutes) then cured for 48 hours at 60° C.

The ballistic data on this series of powders are given in Table IV.

TABLE IV

Solvent Used	Weight, gms.	Height, ins.	K	Pav.	Burning Time, sec.	Burning rate, in./sec.
K	295	2.4	518	1,600	4.8	0.50
L	261	2.2	518	1,400	5.0	0.44
M	265	2.2	518	600	10.0	0.22
L	266	2.2	580	1,700	4.8	0.46
M	263	2.2	580	1,000	8.2	0.27

Example IV

The casting powder consisted of machine-cut cylinders, about 0.030" x 0.030", having the following composition:

	Percent
Nitrocellulose	79
Nitroglycerine	20
Centralite	1
Carbon black (added)	0.02

The casting solvent had the following composition:

	Percent
Nitroglycerine	15
Isophorone	25
Centralite (added)	1.5

The casting technique was as follows. Equal parts of degassed solvent and powder were used in making up 200 gram grains in cellulose acetate tumblers. A variety of casting techniques were used. These are tabulated below (Table V), together with ballistic data for each. The grains were cured 16 hours at 60° C.

TABLE V
[200 gram grains about 2" long]

Sample	Casting Technique	K	Pav.	Burning Time, sec.	Burning Rate, in./sec.
N	Powder in cup. Pour 100 g. liquid over it and evacuate 10 min.	705	1,550	5.4	0.37
O	Powder in cup. Pour 100 g. liquid over it, stir well, evacuate 10 min.	705	800	14.3	0.14
P	Solvent in cup. Pour powder in slowly with stirring, evacuate 10 min.	516	1,020	7.1	0.28
Q	As P, but after evacuation, re-stir well and re-evacuate 30 min.	546	1,900	4.0	0.50
R	As Q, but with additional stirring and ½ hour evacuation.	546	2,000	3.9	0.51
S	As R, but two additional ½ hr. evacuating, preceded by stirring.	480	950	7.0	0.29

Example V

A mixture of 50 parts of Ball Powder of 12% nitroglycerine content and 50 parts of molten "Methyl-NENA" (N-methyl-2-nitroxyethyl nitramine; see Blomquist and Fiedorek Ser. No. 570,807, filed Dec. 12, 1944, now Patent No. 2,669,576); was made into a slurry at 40° C., and cast in the manner described in the preceding examples. A very hard cast product was formed.

It will be apparent to those skilled in the art that the process of the present invention represents a radical innovation in the art of preparing double base propellants. It will also be apparent that many variations, modifications and extensions of the principles applied in the specific examples may readily be made. Accordingly all such variations, modifications and extensions are to be understood as included within the scope of the appended claims.

We claim:

1. The process for the production of propellant grains which comprises:

(a) forming a substantially bubble-free mixture of (1) a solid component comprising nitrocellulose and (2) a liquid component comprising at least one plasticizer for said nitrocellulose, said plasticizer being selected from the group consisting of at least one explosive plasticizer, at least one nonexplosive plasticizer and mixtures thereof, said explosive plasticizer being selected from the group consisting of nitric esters of glycerin, nitric esters of glycol, their polymers and copolymers and said nonexplosive plasticizer being selected from the group consisting of the lower alkyl tartrates, the lower alkyl citrates, the lower alkyl phthalates, the lower alkyl levulinates, cyclic ketones, n-alkyl amides of fatty acids and glyceryl esters of fatty acids where, in said mixtures, the ratio of explosive plasticizer to nonexplosive plasticizer is from about 60:40 to about 80:20, said solid component being capable of absorbing a substantial proportion of said liquid component, the solid-liquid ratio of the mixture being such that the mixture contains not less than about 35 weight percent solid component and not more than about 65 weight percent liquid component, said mixture being formed at a temperature of about 40° C., and

(b) forming said mixture into a propellant grain by curing the mixture in a mold at atmospheric pressure and at a temperature and for a time sufficient to accomplish substantially complete sorption of said liquid component by said solid component.

2. The process for the production of double base propellants which comprises:

(a) forming a mixture of (1) a solid component comprising a double base propellant powder, and (2) a liquid component comprising about 60 to about 80 weight percent explosive plasticizer for said propellant powder and about 40 to about 20 weight percent nonexplosive plasticizer for said propellant powder, the solid-liquid ratio of said mixture being such that the mixture contains not less than about 35 weight percent solid component and not more than about 65 weight percent liquid component, said mixture being formed at about 40° C.,

(b) subjecting said mixture to reduced gas pressure for a period of time sufficient to render same substantially bubble-free, and

(c) heating the bubble-free mixture, while in a mold and at atmospheric pressure, to a temperature and for a time sufficient to bring about the formation of a unitary, substantially non-porous, cast, solid mass.

3. The process of claim 2 wherein the explosive plasticizer is selected from the group consisting of nitric esters of glycerin, nitric esters of glycol, their polymers and copolymers, and the nonexplosive plasticizer is selected from the group consisting of the lower alkyl tartrates, the lower alkyl citrates, the lower alkyl phthalates, the lower alkyl

levulinates, cyclic ketones, n-alkyl amides of fatty acids and glyceryl esters of fatty acids.

4. The process for the production of powder gains which comprises:

(a) preparing a substantially uniform pourable paste by mixing together (1) particles of gelatinized nitrocellulose and (2) a plasticizer for said nitrocellulose, said plasticizer being present in an amount of at least about 25% of the volume of the paste, said mixing being at a temperature at which substantially no dissolution of the nitrocellulose particles in the plasticizer takes place, said temperature being about 40° C., and

(b) maintaining the mixture, at atmospheric pressure and at a temperature at which the nitrocellulose will dissolve in the plasticizer, until the nitrocellulose is completely dissolved and the resulting substantially homogeneous mass has solidified.

5. The process for the production for powder grains which comprises:

(a) preparing a substatnially uniform pourable paste by mixing together (1) pellets of gelatinized introcellulose and (2) a plasticizer, said plasticizer being present in an amount of at least about 25% of the volume of the paste, said mixing being at a temperature at which substantially no dissolution of the nitrocellulose particles in the plasticizer takes place, said temperature being about 40° C.,

(b) pouring said substantially uniform pourable paste into a mold,

(c) subjecting said paste to reduce gas pressure for a period of time sufficient to render same substantially bubble-free, and

(d) maintaining said paste in said mold, at atmospheric pressure and at a temperature at which the nitrocellulose will dissolve in the plasticizer, until the nitrocellulose is completely dissolved and the resulting homogeneous mass has solidified.

6. The process of claim 5 wherein:

(a) the ratio of nitrocellulose to plasticizer is such that the paste contains not less than about 35 weight percent nitrocellulose and not more than about 65 weight percent plasticizer,

(b) the plasticizer comprises about 20–40 parts by weight of desensitizer and about 80–60 parts by weight of energizing modifier, and

(c) the paste, after being poured into the mold and subjected to reduced gas pressure, is heated at a temperature of from about 40° C. to about 80° C. for a period of from about one to two days.

7. The process for the production of propellants which comprises:

(a) placing thin-webbed gelatinized nitrocellulose powder in a mold to a specific level,

(b) adding a plasticizer for the nitrocellulose, the ratio of nitrocellulose to plasticizer being such that the mixture contains not less than about 35 weight percent nitrocellulose and not more than about 65 weight percent plasticizer, said addition taking place at a temperature of about 40° C.

(c) substantially freeing the mixture of entrapped air, and

(d) subjecting the mixture in the mold to a temperature of from about 60° C. to about 75° C. at atmospheric pressure to form a unitary, solid mass.

8. In the art of making powder grains by a molding process, the method which comprises preparing a substantially uniform pourable paste by mixing together particles of gelatinized nitrocellulose propellant and a liquid plasticizer for the nitrocellulose in the amount of at least about 25% by volume of the resulting mixture and at a temperature at which substantially no dissolution of the nitrocellulose particles in the plasticizers takes place and thereafter maintaining said composition at atmospheric pressure, and at a temperature at which the nitrocellulose will dissolve, until the nitrocellulose is completely dissolved and the resulting substantially homogeneous mass has solidified into a powder grain.

9. In the art of making powder grains by a molding process, the method for making substantially homogeneous and substantially non-porous propellant grains which comprises preparing a pourable paste having a substantially uniform composition by mixing together pellets of gelatinized nitrocellulose propellant and a liquid plasticizer for nitrocellulose in the amount of at least about 25 percent by volume of the resulting mixture and at a temperature at which substantially no dissolution of the pellets in the plasticizer takes place during said mixing, shaping said paste in a suitable mold, and thereafter maintaining said paste in said mold at atmospheric pressure and at a temperature at which the nitrocellulose will dissolve, until the nitrocellulose has completely dissolved in said plasticizer and the substantially homogeneous mass has solidified into a propellent powder grain having dimensions and shape conforming substantially to those of said mold.

10. In the art of making powder grains by a molding process, the improved method for making propellant powder grains having a substantially homogeneous nonporous composition which comprises preparing a pourable paste of substantially uniform composition by agitating together pellets of gelatinized nitrocellulose propellant and a liquid plasticizer for nitrocellulose in the amount of at least about 25 percent by volume of the resulting mixture and at a temperature at which substantially no dissolution of the pellets in the plasticizer takes place during said agitation, said plasticizer including in its composition an energizing modifier of the resulting propellent grain, shaping said paste in a suitable mold, and thereafter, maintaining said paste in said mold at atmospheric pressure and at a temperature at which the nitrocellulose will dissolve until the nitrocellulose has completely dissolved in said plasticizer and the substantially homogeneous mass has solidified into a propellent powder grain having dimensions and shape conforming substantially to those of said mold.

11. In the art of making powder grains by a molding process, the improved method for making propellent powder grains having a substantially homogeneous nonporous composition which comprises preparing a pourable paste of substantially uniform composition by agitating together pellets of gelatinized nitrocellulose propellant and a liquid plasticizer for nitrocellulose in the amount of at least about 25% by volume of the resulting mixture and at a temperature at which substantially no dissolution of the pellets in the plasticizer takes place during said agitation, shaping said paste in a suitable mold, and thereafter, heating said paste in said mold at atmospheric pressure and to a temperature at which the nitrocellulose will dissolve until the nitrocellulose has completely dissolved in said plasticizer and the substantially homogeneous mass has solidified into a propellent powder grain having dimensions and shape conforming substantially to those of said mold.

12. The process for the production of large diameter propellant grains which comprises:

(a) forming a substantially bubble-free mixture of (1) a solid component comprising about 35 to about 60 weight percent of nitrocellulose and (2) a liquid component comprising about 30 to about 65 weight percent of a mix comprising about 60 to 80 percent of a material selected from the class consisting of nitroglycerine, ethylene glycol dinitrate, diethylene glycol dinitrate and N-methyl-2-nitroxyethylnitramine and about 20 to 40 weight percent of a plasticizing agent for nitrocellulose selected from the class consisting of triacetin, dimethyl phthalate, butyl tartrate, diethyltartrate, dibutyltartrate, tributylcitrate, butyl velulinate, camphor, isophorone, tri-

ethylphosphate, dimethylformamide and butylacetamide, and

(b) forming said mixture into a propellant grain by curing the mixture in a mold under atmospheric pressure and at a temperature from about 60° C. to about 75° C. for a period from about one-half day to about two and one-half days until the mixture coalesces to form a homogeneous solid grain.

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