United States Patent Office

3,086,896
Patented Apr. 23, 1963

3,086,896

NITROGUANIDINE PROPELLANT COMPOSITION AND PROCESS OF PREPARATION

Russell L. Trask, Morris Plains, N.J., Samuel Sage, Bronx, N.Y., and Isidore G. Nadel, Little Falls, N.J., assignors to the United States of America as represented by the Secretary of the Army

No Drawing. Filed Jan. 19, 1961, Ser. No. 83,849
11 Claims. (Cl. 149—92)
(Granted under Title 35, U.S. Code (1952), sec. 266)

The invention described herein may be manufactured and used by or for the Government for governmental purposes without the payment to us of any royalty thereon.

The present invention relates to a new product and process for producing a triple-base propellant, for use in guns, howitzers and like weapons.

Triple-base propellants are composed of three major ingredients: nitroguanidine, nitrocellulose and nitroglycerine. These propellants have come into increasing use because of their high force values obtained at low flame temperatures. This results in reduced gun barrel erosion and, because of the low percentages of combustibles in muzzle gases relative flashlessness is obtained. However, the manufacturing time and cost of nitroguanidine-base propellants have proven to be considerably greater than those of either single or double-base artillery propellants. The primary reason for this is that a 4½ to 6-hour final mixing period is required to fully incorporate the nitroguanidine crystals into a uniform colloid, as compared to 1 hour for single-base, and a maximum of 2 hours for double-base propellants. It is believed, too, that despite the precaution of the extended mixing time a small percentage of the nitroguanidine crystals are not as uniformily incorporated throughout the propellant as desired. Therefore, erratic ballistics result when the finished product is fired in a weapon. The poorer ballistic uniformity of nitroguanidine-base propellants can also be attributed to (*a*) the fact that they are more difficult to ignite than any of the standard single or double-base formulations of equivalent energy level and (*b*) their poorer physical properties cause propellant-grain breakup in the gun.

It is, therefore, a primary object of this invention to provide a new propellant composition having improved physical and ballistic properties.

Another object of our invention is to provide a process for making triple-base propellants wherein the processing time and power expenditure during processing are greatly decreased.

Still another object of our invention is to reduce or eliminate the high content of crystalline solids that are present due to the formation of double compounds.

A further object of our invention is to provide a triple-base nitroguanidine propellant that is about fifty percent stronger in physical properties than other nitroguanidine propellants.

These and other objects of the invention will become obvious to those skilled in the art as the invention is hereinafter particularly described.

It has been discovered that a new nitroguanidine triple-base propellant is obtained having superior physical and ballistic properties and requiring fifty percent less final mixing time than those heretofore known when the following disclosed process and ingredients are used. The

triple-base propellant composition of our invention contains in addition to the three base propellants, nitroguanidine, nitrocellulose and nitroglycerine, a liquid dialkylphthalate that functions as a plasticizer and a diphenylamine that functions as a stabilizer.

Any liquid dialkylphthalate plasticizer may be used in the triple-base composition. Some representative compounds of this series that are suitable are, dimethylphthalate, diethylphthalate, dipropylphthalate and other members of the dialkylphthalate series.

In addition to the particular type plasticizer utilized, another critical component of the composition is the stabilizer. The stabilizer must not form double compounds with nitroguanidine. Stabilizers that have been found to be suitable for the composition are substituted diphenylamines. Of this series, optimum results are obtained with nitrodiphenylamine and other nitro-substituted diphenylamines.

Another feature of this invention relates to the type of nitrocellulose that is used in the composition. The average degree of nitration of this nitrocellulose is about 12.6% ±.10 (pyrocellulose), rather than the usual military blend of 13.15% nitrogen. It has been discovered that a more uniform mixture of ingredients is obtained when this nitrocellulose is used. Furthermore, this beneficial result, of uniformity of mixture of ingredients, is obtained at a much lower power expenditure, thus resulting in a considerable economic savings.

The triple-base propellant found suitable for our composition contains from 50 to 65 percent nitroguanidine having a specific surface of from about 9,000 to 22,000 sq. cm. per cc., preferably above about 12,000. The nitroguanidine is dispersed in 16 to 23 percent nitrocellulose, which is gelatinized by about 16 to 21 percent nitroglycerin, or other suitable liquid explosive nitric esters such as triethyleneglycol dinitrate, diethyleneglycol dinitrate and butanetriol trinitrate. The dialkylphthalate plasticizer is present in the composition at a concentration of about 4.5 percent and the diphenylamine stabilizer is present in a concentration of about 1.5%.

A process for carrying out our invention is as follows (percentages being by weight):

A pre-mix was prepared in a Schrader bowl, or other like mixer, by adding about one-half amount, 27.3 percent, of nitroguanidine having a specific surface of 15,000 $cm.^2/cm.^3$ and 20 percent nitrocellulose having a nitrogen content of 12.6%. These ingredients were mixed for about five minutes. Then 9 percent of nitroglycerin desensitized with acetone (NG/acetone 82/18) was added and the mixing was continued for an additional five minutes.

This pre-mix was transferred to a sigma blade mixer and mixed for about five minutes before adding the remaining ingredients. The balance of the nitroguanidine, 27.4 percent and 0.3 percent cryolite were added to the mixer followed by the addition of 4.5 percent of dibutylphthalate and 1.5 percent of 2-nitrodiphenylamine dissolved in a solvent, such as 65 parts ethyl alcohol and 35 parts acetone. The total volatile solvent, which includes the acetone for desensitizing the nitroglycerin and the residual ethyl alcohol in the dehydrated nitrocellulose was about 14.5 percent based on the weight of all the ingredients of the composition.

The mixing in the sigma blade mixer was continued for three hours from the time the final addition of nitroguanidine was made. The temperature in the sigma blade mixer was maintained between 80° F. and 105° F. Mixing the composition yielded a uniform, extrudable colloid.

The colloid is then treated in the conventional manner to form shaped charges. That is the colloid is cooled, transferred to a press, blocked, screened, reblocked, extruded through dies of desired dimensions, cut to the desired length and dried.

The following table discloses the preferred composition with thermochemical values and physical properties of the triple-base propellant.

Table I

Composition:

Nitrocellulose (12.6% N) ____percent by weight__	20.0
Nitroglycerin _____do__	19.0
Nitroguanidine (15,000 cm.²/cm.³) _____do__	54.7
Dibutylphathalate _____do__	4.5
2-nitrodiphenylamine _____do__	1.5
Potassium aluminum fluoride (cryolite) _____do__	0.3
Total volatiles _____do__	0.3
Hygroscopicity _____	0.44
Specific gravity (15.5° C./155° C.) _____	1.65
Gravimetric density _____lbs./ft.³__	56.0

Thermochemical values:

Flame temperature _____° K__	2599
Force _____ft.-lbs./lb__	334,000
Heat of explosion _____cal./gm__	807
Unoxidized carbon _____percent by weight__	8.7
Combustibles _____do__	49.8

Physical properties:

Compression test _____percent by weight__	24.8
Side-impact test _____inch-grams__	3460

An alternate process for preparing the above propellant composition is as follows:

20 parts nitrocellulose was slurried in water maintained at about 80° F. until all the fibers were well disbursed. This takes about ten minutes. The water to nitrocellulose ration was approximately 11 parts water to 1 part nitrocellulose, by weight. 19 parts nitroglycerin and 4.5 parts dibutylphthalate were added, as a mixture, to the slurry and agitated for about ten minutes. A water solution of 1.5 parts finely divided nitrodiphenylamine is added to the slurry and said slurry is mixed for ten minutes. The slurry is then centrifuged whereby the water content was brought down to approximately 20 percent. This mixture was dried at about 120° F. with forced air to a moisture content of from about 4 to 6 percent.

This above pre-mix was transferred to a sigma blade mixer, the solvent, a mixture of ethyl alcohol and acetone, was added followed by the addition of 54.7 parts of the composition nitroguanidine. Mixing of the composition is continued for about three hours and the temperature range is maintained between 80° F. and 105° F. The composition is now ready for extrusion.

Test have been made with a conventional triple-base propellant to show the differences in the physical strength between the present product and conventional triple-base propellants. The compression test value for the product of the instant invention as indicated in Table I above, is 24.8, whereas the ascertained value of the conventional product was 16.3. The flexibility of the propellant grains as measured by the standard compression test is 90% to 115% greater than that of triple-base propellants now in use. This improved physical property results in better ballistic regularity, and greater flexibility, resulting in less low temperature brittleness and the elimination of grain breakup.

The impact-test for the composition was found to have

a value of 3460 whereas for a conventional triple-base propellant the value was 1500.

The following table discloses a comparison of the results obtained firing our propellant, identified as composition I, and a conventional triple-base propellant.

Table II

[Ballistic comparisons in 90 mm. gun, M1 series. Shell: 90 mm. HE, M71]

Propellant	Ammo Temp., ° F.	Muzzle Velocity			Pressure		
		Avg., f./s.	Std. Dev., f./s.	Max. Disp., f./s.	Avg., p.s.i./100	Std. Dev., p.s.i./100	Max. Disp., p.s.i./100
Composition I	70	2,758	14.4	52	374	9.4	30
Conventional	70	2,740	24.5	89	385	15.6	51
Composition I	−65	2,703	15.4	54	352	21.6	76
Conventional	−65	2,630	27.4	92	332	28.7	65
Composition I	160	2,857	9.0	35	412	12.9	46
Conventional	160	2,820	17.1	58	410	15.2	54

The superior physical properties of the propellant composition is clearly evidenced by these results set forth above.

We claim:

1. In the process of preparation of an extrudable propellant composition which comprises forming a mixture consisting essentially of, in proportions by weight, 50–65% nitroguanidine, in a state of comminution such that the particles have a surface of from 9,000–22,000 cm.²/cm.³, 16–23% pyrocellulose, and 16–21% of a nitric acid ester of a polyhydric alcohol from the group consisting of diethylene glycol, triethylene glycol, glycerin and butanetriol, the improvement which comprises stabilizing and increasing the dispersion of the said nitroguanidine and the said nitrocellulose in the said nitric acid ester by adding to the said mixture thereof a minor proportion of a dialkylphthalate, wherein the said alkyl groups contain from 1–6 carbon atoms, and a minor proportion of a nitrodiphenylamine and homogenizing the said mixture, at an elevated temperature, until a homogeneous colloid is formed.

2. The process as fet forth in claim 1 wherein the dialkylphthalate is dibutylphthalate.

3. The process as set forth in claim 1 wherein the nitrodiphenylamine is 2-nitrodiphenylamine.

4. The process as set forth in claim 1 wherein the dialkylphthalate is added in the proportion of about 4.5% by weight of the total composition.

5. The process as set forth in claim 1 wherein the diphenylamine is added in the proportion of about 1.5% by weight of the total composition.

6. The process as set forth in claim 1 wherein the homogenizing is carried out at a temperature of from 80 to 105° F.

7. An extrudable colloidal propellant composition comprising substantially, in proportions by weight, 50–65% nitroguanidine, in a state of comminution such that the particles have a surface of from 9,000–22,000 cm.²/cm.³, 16–23% pyrocellulose, and 16–21% of a nitric acid ester of a polyhydric alcohol from the group consisting of diethylene glycol, triethylene glycol, glycerin, and butanetriol and a minor proportion of a dialkylphthalate, wherein the said alkyl groups contain from 1–6 carbon atoms, and a minor proportion of a nitrodiphenylamine.

8. The composition as set forth in claim 7 wherein the dialkylphthalate is dibutylphthalate.

9. The composition as set forth in claim 7 wherein the nitrodiphenylamine is 2-nitrodiphenylamine.

10. The composition as set forth in claim 7 wherein the

dialkylphthalate is present in the proportion of about 4.5% by weight of the total composition.

11. The composition as set forth in claim 7 wherein the nitrodiphenylamine is present in the proportion of about 1.5% by weight of the total composition.

References Cited in the file of this patent

UNITED STATES PATENTS

2,027,114	Olsen et al.	Jan. 7, 1936
2,557,463	Pring	June 19, 1951