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SOLID PROPELLANT COMPOSITION CONTAINING THE REACTION PRODUCT OF POLYVINYLENE GLYCOL NITRATE AND AN AROMATIC DIISOCYANATE

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ABSTRACT OF THE DISCLOSURE

An improved high energy additive for use in a propellant subjected to a desert type environment consisting of the reaction product of polyvinylene glycol nitrate and an aromatic diisocyanate.

This invention relates to an improved additive for use in a propellant. More particularly, this invention relates to additive for use in a propellant thereby producing increased stability coupled with enhanced physical and mechanical properties.

Sometime in the past, polyvinylene glycol nitrate due to its inherent characteristics was thought to possess great potential as an additive for use in a propellant. It was also though to possess special advantages when utilized in a propellant designed for use in a gun or the motor of a rocket. These advantages are derived from the fact that polyvinylene glycol nitrate is polymeric in nature and possesses a relatively high amount of energy. Thus, it could be used to replace ammonium perchlorate, ammonium nitrate, or some of the explosive itself, such as HMX or RDX, with the advantage that it would not in any way degrade the energy level of the propellant but would, in fact, improve the mechanical properties of the final product. Further, it was found that polyvinylene glycol nitrate possesses the ability to colloid with nitro-type plasticizers such as nitroglycerine, butane trioltrinitrate and diethylene glycol dinitrate. Thus, it may be used to partially replace these expensive plasticizers without loss of energy to the system. Also, the volatility of double base propellants ordinarily possessing these types of plasticizers may be reduced by replacement of a portion of such nitro-type plasticizers.

However, conventional warfare under a desert type environment dictates that a propellant possess stability at temperatures in excess of 60° C. But to dismay, it was found that propellants, having polyvinylene glycol nitrate incorporated therein, did not possess the stability desired due to the low softening point and other inherent characteristics of such nitrate compound. It was also found that such polymer decomposed under the conditions desired affecting the ballistic characteristics of the round and rendering firing unreproducible from round to round.

The subject invention answers the needs of the art as described above with special emphasis on improving the stability of polyvinylene glycol nitrate at temperatures usually encountered in the desert.

It is, therefore, an object of this invention to provide a stable form of polyvinylene glycol nitrate for use in propellants.

Another object is to provide a modified form of polyvinylene glycol nitrate which may be utilized in propellant under a desert type environment.

Other objects and many of the attendant advantages of this invention will be readily appreciated as the same becomes better understood by reference to the following detailed description.

We have discovered that polyvinylene glycol nitrate may be effectively stabilized by reaction with an aromatic diisocyanate such as toluene diisocyanate. The polyvinylene glycol nitrate thus stabilized may be incorporated in a propellant for use at temperatures in excess of 60° C., such as that encountered in desert warfare, without any adverse effects to the propellant. In fact, this incorporation has been found to be accompanied by the inclusion of all the inherent advantages of the polyvinylene glycol nitrate itself. This improvement in stability is evidenced by an increase in thermal stability as shown by differential thermal analysis. This improvement is also accompanied by a reduction in sensitivity to impact. It has been found to advantage that the modified form of polyvinylene glycol nitrate is less sensitive than nitroglycerine and comparable to nitrocellulose. Further, a visual observation of the burning rate of the modified form of polyvinylene glycol nitrate was demonstrated to advantage to be many times faster than a nitrocellulose fiber having a nitrogen content of 12.6 percent.

The following are examples of the preparation of the material of this invention.

Example I

A specified amount of polyvinylene glycol nitrate was dissolved in acetone and between 5 and 15 percent of toluene diisocyanate was added. The specific amount of diisocyanate is dependent on the hydroxyl content of the nitrated polymer. For instance, about 10 percent of diisocyanate was added, when the hydroxyl content of the nitrate polymer was about 1 percent. The resulting solution was stirred from 1 to 4 hours and then poured in a large volume of water producing a precipitate. The latter solid was filtered from the solution, washed with water and dried in a vacuum oven. The resulting solid material was then tested as hereafter described.

Example II

As an alternate procedure, the polyvinylene glycol nitrate is slurried in an inert solvent such as heptane and the same proportion of diisocyanate utilized in Example I is added to the solution. The slurry is stirred from 1 to 4 hours and then filtered to remove the precipitate. After drying, the precipitate was utilized in the tests which follow.

The following are the results obtained with a vacuum stability test which measures thermal stability of a particular compound. The test is carried out by heating a 5 gm. sample, under vacuum at specified temperatures, and measuring the evolution of gas. The latter is a measurement of the degradation of a compound when exposed to a particular temperature for a particular length of time.

TABLE I

	Vaccum Stability Test	
Thermal Stability Temp., ° C.	A[1]	B[2]
60	11 mls. (16 hrs.)	1.22 mls. (40 hrs.).
80		4.37 mls. (40 hrs.).
100		11 mls. (40 hrs.).

[1] Polyvinylene glycol nitrate.
[2] Reaction product of the above and toluene diisocyanate.

The above table illustrates the degree of stability achieved at the various specified temperatures. It shows that polyvinylene glycol nitrate exhibits a high degree of instability at 60° C., when heated for only 16 hours, whereas the modified form of the same material exhibits a slight degree of instability of the modified material is only minor, when heated for 40 additional hours at a temperature of 80° C., which is 20 degrees higher than the temperature at which polyvinyl glycol nitrate itself

xhibits complete instability. This is quite an improvement nd is evidence of the fact that the modified material of iis invention may be incorporated into a propellant which iay be utilized in a desert environment without exhibiting ny instability due to temperature. Table II, which fol- ows, illustrates the loss of weight achieved by an 0.6 ample when heated for a specified time at a specified emperature. This evidence indicates the degree of in- tability of the material under study at each of the tem- eratures specified.

TABLE II.—HEAT TEST

	Loss in Weight		
	1st 48 hrs.	2nd 48 hrs.	100 hrs.
[1]:			
60° C	60.8%	4.0%	No Explosion.
80° C	60.7%	4.9%	Do.
100° C			
[2]:			
60° C	3.8%	0.5%	No Explosion.
80° C	22.2%	8.9%	Do.
100° C	51.4%	9.0%	Do.

[1] Polyvinylene glycol dinitrate (material under study).
[2] Reaction product of polyvinylene glycol nitrate and toluene diiso- ranate.

As indicated above, polyvinylene glycol nitrate possesses relatively greater instability than that of the reaction roduct of this material and toluene diisocyanate as videnced by the greater loss in weight at a relatively lower emperature for the initial heating period.

Another indication of the stability of the material of his invention is the impact sensitivity of the material. Vhen a 2 kilogram weight was dropped from a 3-inch eight on top of a sample of the material of this inven- ion, it did not explode. However, when polyvinylene lycol nitrate was subjected to the same test, it detonated. t was found that a 4-inch height was required before he reaction product, a polyvinylene glycol nitrate and an romatic diisocyanate, which is the material of this in- ention, was exploded under otherwise identical testing onditions. This is substantial evidence of the improve- ient achieved in the stability of the polymer by the con- ept of this invention.

The physical strength of both polyvinylene glycol nitrate nd the material of this invention was tested as follows.)ne inch strips of each of the above materials were pre- ared and tested in an Instrom Tensile Tester utilizing onventional procedures. It was found that the tensile trength of the nitrate polymer was 8,700 p.s.i. while the ensile strength of the material of this invention was found o be 11,500 p.s.i. This is a good indication of improve- ient in physical strength of the polymer under study.

The material of this invention may be used to replace solid crystalline oxidizer in both double and triple base ropellants as indicated previously. If this were done, the esulting propellant would be more homogeneous due to he colloiding action of such material with the nitroplas- icizers. The following are examples of the substitutions pecified, to the advantage of the propellant.

	A, percent [1]	B, percent	C, percent
Nitrocellulose	28.0	28.0	28.0
Nitroglycerin	22.5	22.5	10.0
Nitroguanidine	47.7		
Ethyl Centralite	1.5	1.5	1.5
Cryolite	0.3	0.3	0.3
[2]		47.7	60.2

[1] Conventional propellant.
[2] Reaction product of polyvinylene glycol nitrate and toluene diiso- yanate.

	D [1]	E	F	G
HMX	25.0		10.0	10.0
Nitrocellulose (12.6%)	38.0	38.0	38.0	38.0
Nitroglycerin	16.5	16.5	16.5	16.5
Sucrose octaacetate	7.8	7.8	7.8	7.8
Triacetin (glycerol triacetate)	7.7	7.7		7.7
Lead Stearate	3.0	3.0	3.0	3.0
N-methyl-p-nitroaniline	1.0	1.0	1.0	1.0
Ethyl centralite	1.0	1.0	1.0	1.0
[2]		25.0	22.7	25.0

[1] Conventional propellant.
[2] Reaction product of polyvinylene glycol nitrate and toluene diiso- cyanate.

As indicated above, the material of this invention may be partially or wholly substituted for other ingredients of a conventional propellant. It was found that this substitution was not accompanied with any loss of energy of the propellant itself. In fact, the propellant compositions themselves after substitution were assured of increased stability and in some cases a greater amount of energy.

The aromatic diisocyanate which may be used to advantage in this invention includes phenylene-1-4-diisocyanate; p,p[1]-biphenylene diisocyanate; p,p[1]-diphenyl methane diisocyanate.

Obviously, many modifications and variations of the present invention are possible in light of the above teachings. It is, therefore, to be understood that within the scope of the appended claims, the invention may be practiced otherwise than as specifically described.

I claim:

1. In a solid propellant for use at temperatures in excess of 60° C., said propellant containing nitrocellulose and nitrate esters, the improvement of a high energy additive consisting of the reaction product of polyvinylene glycol nitrate and an aromatic diisocyanate.

2. The propellant of claim **1** wherein said aromatic diisocyanate is toluene diisocyanate.

3. The propellant of claim **1** wherein said diisocyanate is present in an amount between 5 and 15 percent by weight of said additive.

4. In a solid propellant for use at temperatures in excess of 60° C., said propellant containing nitrocellulose, nitrate esters and nitroquanidine, the improvement of a high energy additive consisting of polyvinylene glycol nitrate and an aromatic diisocyanate.

5. The propellant of claim **4** wherein said aromatic diisocyanate is toluene diisocyanate.

6. The propellant of claim **4** wherein said diisocyanate is present in an amount between 5 and 15 percent by weight of said additive.

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