United States Patent

Engel et al.

[15] **3,663,323**

[45] **May 16, 1972**

[54] ENERGETIC PROPELLANT COMPOSITION CONTAINING A DIFLUORAMINO BINDER

[72] Inventors: **Lawrence J. Engel,** Green Brook; **John Brooke Gardiner,** Mountainside, both of N.J.

[73] Assignee: **The United States of America as represented by the Secretary of the Army**

[22] Filed: **Apr. 30, 1970**

[21] Appl. No.: **43,618**

Related U.S. Application Data

[63] Continuation-in-part of Ser. No. 671,164, Sept. 27, 1967, abandoned.

[52] **U.S. Cl.** **149/19,** 149/20, 149/42, 149/44, 149/76

[51] **Int. Cl.** **C06b 11/00**

[58] **Field of Search** 149/42, 19, 44, 20, 76

[56] **References Cited**

UNITED STATES PATENTS

3,441,550	4/1969	Zimmerman	260/89.5
3,447,323	6/1969	Allan	149/109
3,441,549	4/1969	Gardiner et al.	160/88.3
3,332,812	7/1967	Guthrie	149/76
3,332,811	7/1967	Guthrie et al.	149/76

Primary Examiner—Carl D. Quarforth
Assistant Examiner—Stephen J. Lechert, Jr.
Attorney—Harry M. Saragovitz, Edward J. Kelly, Herbert Berl and James T. Deaton

[57] **ABSTRACT**

An improved energetic propellant charge, of the solid type useful in rockets and rocket motors, may be formulated with a system of the polymer of tetrakis(difluoramino)amyl acrylate and a large amount of hexakis(difluoramino)propyl ether if trimethylolpropane trimethacrylate and a peroxidic polymerization catalyst such as dichlorobenzoyl peroxide are used to crosslink the tetrakis(difluoramino)amyl acrylate from which the polymer is derived.

4 Claims, No Drawings

ENERGETIC PROPELLANT COMPOSITION CONTAINING A DIFLUORAMINO BINDER

CROSS REFERENCE TO RELATED APPLICATION

This application is a continuation-in-part of copending application, Ser. No. 671,164 filed Sept. 27, 1967 and now abandoned.

BACKGROUND OF THE INVENTION

This invention relates to jet propulsion and particularly to an improved propellant charge of the solid type useful in rockets and rocket motors.

Modern military operations are becoming more dependent upon rocket motors. The very nature of the operations require a motor that will give high and reliable performance so that lives will not be lost. A rocket motor is heavily dependent upon a reliable propellant. A solid propellant motor is most reliable since, once ignited, it will continue to burn until it is consumed. As the usefulness of a propellant is dependent upon the intensity of thrust created by its combustion and the consistent reproducibility of said thrust, it has long been the objective of those engaged in the art to find a means of increasing the thrust by means of energetic propellant compositions. This invention provides an energetic solid rocket propellant composition that is capable of high and reliable performance. The polymer used as the propellant binder must result in a propellant composition with certain mechanical and chemical properties. Among some of the more important mechanical properties are: elongation at maximum stress and at break, stress at the maximum of stress-strain curve; tensile strength; tear strength; and elastic recovery when the stress is removed. Since the polymeric binder and oxidizer are in intimate contact with each other and may be exposed to elevated temperatures for prolonged storage periods, it is important that the polymeric binder and oxidizer possess good mutual chemical stabilities toward each other.

The use of a peroxidic curing catalyst is well known in the art of propellant compositions. It is also known, but not as generally, that the curing catalyst assumes roles beyond that of crosslinking molecules containing the alkenic linkage. An undesirable side reaction of the curing catalyst is the promotion of the degradation of the cured propellant, probably by oxidative degradation of the polymerized fuel-binder. As a consequence of these harmful degradations, the quantity of curing catalyst used to aid in the cure of the propellant should be held to the minimum, the final amount being a compromise between the quantity necessary to achieve an adequate cure of the polymeric binder and that which can be tolerated without undue degradation process.

The polymer of tetrakis(difluoramino)amyl acrylate has been investigated as a material for such binder development. It is a strong polymer readily plasticized by several NF_2 compounds. However, it suffers the disadvantage of losing strength upon adding NF_2 liquid plasticizer and with slight increases in temperature. Therefore, attempts have been made to improve the strength of this acrylate-type binder.

It is therefore an object of this invention to provide a tetrakis(difluoramino)amyl type polymer with improved mechanical properties for use in propellant compositions.

Another object of this invention is to provide a new and improved composition of matter.

A further object of this invention is to provide a polymer which requires low levels of both curing catalyst and crosslinking agent when it is employed in propellant compositions.

One still further object of this invention is to provide a polymer which has good chemical stability with the oxidizer of the propellant composition in which it is employed.

SUMMARY OF THE INVENTION

It has been discovered that an energetic propellant may be formulated with a system of the polymer of tetrakis(difluoramino)amyl acrylate and a large amount of hexakis(difluoramino)propyl ether if trimethylolpropane trimethacrylate and a peroxidic polymerization catalyst such as dichlorobenzoyl peroxide are used to crosslink the tetrakis(difluoramino)amyl acrylate from which the polymer is derived.

DESCRIPTION OF THE PREFERRED EMBODIMENT

The synthesis of tetrakis(difluoramino)amyl acrylate is effected as follows:

Divinylcarbinol + N_2F_4 ⟶

1,2,4,5-tetrakis(difluoromino)-3-amyl alcohol acrylate, $\xrightarrow{\text{acrylyl chloride}}$ tetrakis(difluoramino)amy

The polymer of tetrakis(difluoramino)amyl acrylate is then formed from the monomer of tetrakis(difluoramino)amyl acrylate by copolymerization with trimethylolpropane trimethylacrylate (or other crosslinker containing three functional groups) and a peroxidic polymerization catalyst. For a more detailed explanation of this synthesis, see U. S. Pat. No. 3,441,550.

The polymerization is deferred until the mixing of all of the propellant ingredients has been carried out. Dilatometer data on the propellant system showed the cure to be substantially complete after 72 hours at 50° C.

The synthesis of hexakis(difluoramino)propyl ether is effected by reacting 1,2-divinyl ethylene oxide dissolved in carbon tetrachloride with N_2F_4 at reaction temperatures in the range of 100° to 150° C. The NF_2 group becomes linked to the carbons to give the product having the composition $CH_2(NF_2)CH(NF_2)CH(NF_2)OCH(NF_2)CH(NF_2)CH_2(NF_2)$.

Of course, a suitable oxygen oxidizer, such as ammonium perchlorate, is included as an ingredient in the propellant composition.

Similarly, a suitable fuel, such as aluminum metal, is included as an ingredient in the propellant composition.

A specific example of a propellant composition that has been successfully formulated and fired in both hollow grain and end burning motors containing 10 grams of propellant is included herein for illustrative purposes only. The composition is:

COMPONENTS	WT.%
Hexakis(difluoramino)propyl ether	30.7
Tetrakis(difluoramino)amyl acrylate	11.1
Dichlorobenzoyl peroxide	0.15
Trimethylolpropane trimethyacrylate	0.65
Ammonium perchlorate, (65 microns)	46.5
Aluminum	10.2

Thus, the addition of trimethylolpropane trimethacrylate to crosslink the tetrakis(difluoramino)amyl acrylate has permitted the use of a large amount of hexakis(difluoramino)propyl ether as a plasticizer while still retaining adequate physical properties.

Obviously many modifications and variations of this invention are possible in the light of the above teaching. It is therefore to be understood that, within the spirit and scope of this invention, it may be practiced otherwise than as specifically described.

We claim:

1. A solid propellant composition consisting essentially of an ammonium perchlorate oxidizer, a metal fuel source, at least 25 weight percent plasticizer consisting essentially of hexakis(difluoramino)propyl ether, tetrakis(difluoramino)amyl acrylate, a crosslinker of trimethylolpropane trimethyl acrylate, and a peroxide polymerization catalyst.

2. The composition of claim 1 wherein said fuel source is aluminum.

3. The composition of claim 1 wherein said peroxidic polymerization catalyst is dichlorobenzoyl peroxide.

4. The composition of claim 1, wherein said oxidizer is present in an amount of about 46.5 weight percent, wherein

said metal is aluminum and is present in an amount of about 10.2 weight percent, wherein said plasticizer is present in an amount of about 30.7 weight percent, wherein said tetrakis(difluoramino)amyl acrylate is present in an amount of about 11.1 weight percent, wherein said crosslinker is present in an amount of about 0.65 weight percent, and wherein said catalyst is dichlorobenzoyl peroxide and is present in an amount of about 0.15 weight percent.

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