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# 3,017,301
# PROPELLANT COMPOSITIONS CONTAINING A POLYCYANO FUEL COMPONENT

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The present invention relates to propellants of the type which may be used in rockets or missiles. More particularly, it pertains to propellant compositions containing polycyano compounds in admixture with an oxidizing agent.

This application is a continuation-in-part of my prior copending application Serial No. 649,613, filed April 1, 1957, now abandoned.

Propellants of the type used to power rockets and missiles must operate under conditions in which it is difficult or impossible to obtain oxygen for combustion from the surrounding environment. For example, the rarefied atmosphere at high altitudes through which rockets and missiles must sometimes be driven will not supply sufficient oxygen to support combustion of a fuel. In the case of underwater missiles, the lack of oxygen in an amount and form suitable to support combustion is even more acute. The propellants of this invention thus contain, in addition to a fuel, an oxidizing agent in intimate association with the fuel.

It is an object of the present invention to provide novel propellant compositions of the type described which contain within themselves all, or a predetermined percentage, of the oxidant necessary for combustion of the fuel component. It is a further object of the invention to provide propellant compositions that release copious quantities of gaseous reaction products upon combustion which, when exhausted, will develop a high degree of thrust. Other and additional objects will become apparent from a consideration of the ensuing specification and claims.

The novel propellant compositions of the present invention comprise an organic polycyano fuel component and a liquid or solid oxidizing agent therefor. The fuel may be any organic polycyano compound in which the ratio of cyano groups to non-cyano carbon atoms in the molecule is at least 0.8:1. The polycyano compound may be unsubstituted or it may be in the form of a metallic or other salt, as will be more particularly hereinafter described. The preferred organic polycyano fuels are those (including the salts) in which the parent cyano compound contains no elements other than carbon, hydrogen, and nitrogen, and particularly those composed substantially of carbon and nitrogen (i.e., containing two or less hydrogen atoms per molecule). An especially preferred group of organic polycyano fuels within this category are those (including the salts) in which the polycyano compound contains at least three cyano groups per molecule and in which the ratio of cyano groups to non-cyano carbon atoms in the molecule is 1:1 or greater. Many of the organic polycyano compounds within the above definition which may be used as fuels according to the present invention contain readily ionizable (acidic) hydrogen. Such compounds are often more stable and readily isolated in the forms of their salts. Various of these salts will be particularly preferred polycyano fuels for use in the present invention. The cation of these salts may be any known cation, such as ammonium, substituted ammonium, and the ionic forms of the metals; the inorganic cations such as the ionic forms of the metals being preferred.

The salts of the polycyano compounds which may be employed in the present invention are readily prepared in aqueous systems, in which event some of these salts will



contain water of crystallization. Such hydrated salts are fully operable in the compositions of the present invention.

According to the present invention, a mixture of the polycyano fuel with an oxidant, such as a solid oxidant like a metallic nitrate or permanganate is prepared, the mixture being form-stable at ambient temperatures at least up to about 50° C. Such compositions may be prepared and handled with safety under ordinary conditions and may be ignited at the proper time by the application of suitable heat and/or flame. Dispersions of the polycyano fuels in a liquid oxidant such as nitric acid, liquid oxygen or the like are also within the scope of the invention. The invention will be better understood from a consideration of the following examples.

Examples 1 through 3 which follow illustrate the preparation of polycyano compounds which may be used in the present invention.

## EXAMPLE 1

An aqueous solution (50 ml.) of barium chloride containing 12.2 g. of $BaCl_2.2H_2O$ was added slowly to 100 ml. of an aqueous solution containing 16.4 g. of the monohydrate of the sodium salt of 1,1,3,3-tetracyanopropylene (a suitable method for preparing the latter compound is disclosed in Y. Urushibara, Bull. Chem. Soc. Japan, 2, 278 (1927)). White crystals slowly formed. After one hour the mixture was filtered to obtain 11.5 g. of the solid trihydrate of the barium salt of 1,1,3,3-tetracyanopropylene, $Ba[(NC)_2C{=}CH{-}C(CN)_2]_2{\cdot}3H_2O$. A sample for analysis was obtained by recrystallization from water.

*Analysis.*—Calcd. for $BaC_{14}H_2N_8{\cdot}3H_2O$: Ba, 29.2; C, 35.3; N, 23.6; $H_2O$, 11.4. Found: Ba, 30.86; C, 35.05, 36.27; N, 23.51, 23.80; $H_2O$, 10.9.

## EXAMPLE 2

A hot aqueous solution (60 ml.) containing 2.49 g. of $CuSO_4{\cdot}5H_2O$ was added to 60 ml. of a hot aqueous solution containing 4.73 g. of

$$Ba[(NC)_2C{=}CH{-}C(CN)_2]_2{\cdot}3H_2O$$

Barium sulfate precipated. The mixture was kept hot for ½ hour and filtered hot. Evaporation of the filtrate yielded 1.8 g. of $Cu[(NC)_2C{=}CH{-}C(CN)_2]_2{\cdot}4H_2O$ in the form of brown crystals.

*Analysis.*—Calcd. for $CuC_{14}H_2N_8{\cdot}4H_2O$: Cu, 15.1; C, 40.0; H, 2.39; N, 26.75. Found: Cu, 15.07; C, 40.50; H, 2.58; N, 26.82.

## EXAMPLE 3

The procedure of Example 2 was repeated except that 2.63 g. of $NiSO_4{\cdot}6H_2O$ was substituted for the cupric sulfate. Green crystals of

$$Ni[(NC)_2C{=}CH{-}C(CN)_2]_2{\cdot}3H_2O$$

were obtained.

*Analysis.*—Calcd. for $NiC_{14}H_2N_8{\cdot}3H_2O$: Ni, 14.90; C, 42.7; H, 2.03; N, 28.4. Found: Ni, 14.57, 14.42; C, 42.90, 43.09; H, 2.23, 2.32; N, 28.99, 28.60.

The various salts of the polycyano compounds, and a suitable technique for their preparation, referred to in the examples below are described by Middleton et al., J. Am. Chem. Soc. 80, 2795–2806 (1958). The utility of the compositions of the present invention for propellant purposes is illustrated by the following examples.

## EXAMPLE 4

Aluminum 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide pentacosahydrate was partially dehydrated by azeotropic distillation of the finely ground salt with toluene for five hours. The resulting powder was collected

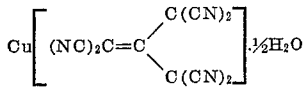
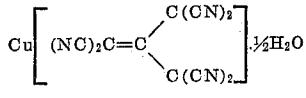
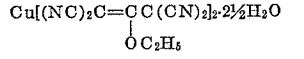
by filtration and dried under vacuum over phosphorus pentoxide to obtain the corresponding heptahydrate.

*Analysis.*—Calcd. for $Al_2C_{30}N_{18}\cdot 7H_2O$: C, 45.5; H, 1.8; N, 31.9; Al, 6.8. Found: C, 45.2; H, 1.9; N, 33.1; Al, 7.5.

A finely ground, intimate mixture of 0.1 g. each of sodium chlorate and aluminum 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide heptahydrate was tightly packed into an aluminum cylinder 2 cm. long, 2.5 mm. inside diameter and of 3 mil. wall thickness, sealed at one end. This projectile was placed at one end of a glass guide tube 1.3 meters long, open at both ends. An illuminating gas flame was brought into contact with the open end of the projectile which immediately fired and traversed the guide tube at a speed too rapid to observe with the eye.

tube. One end of the launch tube was covered with a piece of 0.001" aluminum foil. Brass cylinders 15 mm. long, 4 mm. in diameter and 3 mils. in wall thickness were prepared and sealed at one end. These empty projectiles weighed about 0.2 g. each. Equal weight mixtures of representative salts of cyanocarbon acids and oxidants (see Table I) were finely ground and intimately mixed with a mortar and pestle. These propellants were then packed into the brass projectiles, each projectile containing about 0.15–0.2 g. of propellant. Each of these rockets in turn was placed at the open end of the launch and ignited as described in Example 4. Each flight was observed with regard to qualitative speed and ability of the rocket to pierce or penetrate the aluminum foil buffer at the distal end of the launch. Results are summarized in Table I.

*Table I*

| Cyanocarbon Salt (Fuel) | Oxidant | Flight Performance |
|---|---|---|
|  | $NaClO_3$ | Easy ignition; rapid flight; penetrated aluminum foil; some power remaining after penetration. |
|  | $KClO_4$ | Do. |
| $Cu[(NC)_2C{=}CHC(CN)_2]_2\cdot 4H_2O$ | $NaClO_3$ | Easy ignition; rapid, full length flight; no penetration of aluminum foil. |
| $Ni[(NC)_2C{=}CHC(CN)_2]_2\cdot 3H_2O$ | $NaClO_3$ | Do. |
| $Ba[(NC)_2C{=}CHC(CN)_2]_2\cdot 3H_2O$ | $NaClO_3$ | Difficult ignition; mild explosion. |
| $Na[(NC)_2C{=}CHC(CN)_2]\cdot H_2O$ | $NaClO_3$ | Do. |
|  | $NaClO_3$ | Easy ignition; rapid flight; pierced aluminum foil but did not pass through. |

## EXAMPLE 5

In 300 ml. of water at 80° C. was dissolved 18 g. of barium 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide hexahydrate. To this solution was added a hot solution of 5.1 g. of lithium sulfate in 250 ml. of water. Barium sulfate precipitated, and the mixture was kept at 80° C. for ½ hour and filtered hot. Evaporation of the filtrate yielded a white crystalline product. This was dried under vacuum over phosphorus pentoxide and further dehydrated by azeotropic distillation with toluene for five hours. The resulting white powder was collected by filtration, washed with petroleum ether, and dried under vacuum over phosphorus pentoxide to yield anhydrous lithium 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide.

*Analysis.*—Calcd. for $Li_2C_{10}N_6$: C, 55.0; N, 38.5; Li, 6.4. Found: C, 54.7; N, 40.0; Li, 6.2.

A projectile of shape and dimensions similar to those described in Example 4 was packed with a 0.2-g. propellant mixture of equal parts of lithium 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide and sodium nitrite. When ignited this projectile rapidly traversed a 15-cm. guide tube and continued on a free flight path.

## EXAMPLE 6

A projectile as described in Example 4 was loaded with a 0.2-g. propellant grain of equal parts of lithium 2-dicyanomethylene - 1,1,3,3 - tetracyanopropanediide and sodium chlorate. The projectile was placed in the glass guide tube of Example 4. When ignited with an illuminating gas flame, the projectile detonated, shattering all but a 10-cm. center portion of the guide tube. The shock wave was felt over ten feet away from the source of detonation.

## EXAMPLE 7

A rocket launch of 10 mm. inside diameter was prepared from a copper tube 1.2 meters long. Vent holes about 10 cm. apart were made on the upper aspect of the

## EXAMPLE 8

An aluminum tube closed at one end and having an interior diameter of 4 mm. and a length of 15 mm. is packed with an intimate mixture of 128 parts of powdered tetracyanoethylene and 276 parts of powdered sodium nitrite. The resulting rocket is placed on an inclined metal launching trough and fired by igniting the mixture at the open end of the rocket with a match flame. A rapid oxidation of the tetracyanoethylene occurs, propelling the rocket from the launching trough into the air at high speed.

Tetracyanoethylene may be prepared by the reaction of malononitrile with sulfur monochloride in the presence of an inert liquid diluent, as described more particularly by Cairns et al., J. Am. Chem. Soc. 80, 2775–8 (1958).

## EXAMPLE 9

A rocket is launched in the manner of Example 8, employing as a propellant charge a mixture of 128 parts of powdered tetracyanoethylene and 374 parts of powdered silver nitrate.

## EXAMPLE 10

A rocket is launched in the manner of Example 8, employing as a propellant charge a mixture of 128 parts of powdered tetracyanoethylene and 379 parts of powdered potassium permanganate.

## EXAMPLE 11

A rocket is launched in the manner of Example 8, employing as a propellant charge a mixture of 128 parts of powdered tetracyanoethylene and 1430 parts of powdered lead dioxide.

## EXAMPLE 12

Fifty parts of the dihydrate of the monosodium derivative of 4-cyano-5-dicyanomethylene-3-hydroxy-2-oxo-3-pyrroline (Carboni et al., J. Am. Chem. Soc. 80, 2840 (1958)), was suspended in 157 parts of acetonitrile, and 60 parts of oxalyl chloride was added. The mixture was

stirred and heated at reflux for one hour. The mixture was then cooled and the precipitate which formed was collected by filtration, washed with acetonitrile and dried under reduced pressure to obtain 36 parts (80% yield) of crude 3-chloro-4-cyano-5-dicyanomethylene-2-oxo-3-pyrroline in the form of buff-colored crystals. It was purified by sublimation at 180° C. and 0.1 mm. pressure to give colorless crystals.

*Analysis.*—Calcd. for $C_8NH_4ClO$: C, 47.06; H, 0.49; N, 27.45; Cl, 17.40. Found: C, 47.53; H, 0.77; N, 27.28, 27.46; Cl, 16.99, 17.04.

To a solution of 204 parts of 3-chloro-4-cyano-5-dicyanomethylene-2-oxo-3-pyrroline in 1800 parts of ethyl acetate was added 88 parts of sodiomalononitrile. The sodium salt of 4-cyano-3-dicyanomethyl-5-dicyanomethylene-2-oxo-3-pyrroline precipitated as a brick red solid. This product (168 parts) was recovered by filtration and recrystallized from water.

Equal portions of the sodium salt of 4-cyano-3-dicyanomethyl-5-dicyanomethylene-2-oxo-3-pyrroline and sodium chlorate were mixed and charged into a brass projectile and fired as in Example 7. The projectile was readily ignited by an illuminating gas flame and rapidly traversed the rocket launch but did not pierce the aluminum foil at the end of the tube.

Cyanocarbon acids and their salts which are useful as fuels in the present invention and which may be substituted for the cupric salt of 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide hemihydrate of Table I (Example 7) include the following:

1,1,2,3,3-pentacyanopropene and its trimethylsulfonium, silver, and beryllium salts.

1,1,2,4,5,5-hexacyano-3-azapentadiene and its trimethylsulfonium, acetamidinium, and silver salts.

1,1,2,5,6,6-hexacyano-3,4-diazahexadiene and its silver salt.

2-dicyanomethylene-1,1,3,3-tetracyanopropane and its sodium, potassium, magnesium, silver, cadmium, uranyl, ferrous, vanadyl, nickel, cobaltous, strontium, zinc, beryllium, manganous, mercuric, and ammonium salts.

2-ethoxy-1,1,3,3-tetracyanopropene and its sodium, cobaltous, cupric, and nickelous salts.

2-($\beta$-hydroxyethoxy)-1,1,3,3-tetracyanopropene and its sodium and cobaltous salts.

2-methylthio-1,1,3,3-tetracyanopropene.

2-amino-1,1,3,3-tetracyanopropene and its silver salt.

2-methylamino-1,1,3,3-tetracyanopropene and its sodium salt.

2-dimethylamino-1,1,3,3-tetracyanopropene and its sodium salt.

2-N-nitrosomethylamino-1,1,3,3-tetracyanopropene.

2-bromo-1,1,3,3-tetracyanopropene and its silver and barium salts.

2-chloro-1,1,3,3-tetracyanopropene.

Tricyanomethane and its sodium, barium, and copper salts.

Tetracyanoethylene.

Other polycyano compounds useful as fuels in the present invention include malononitrile, vinylidene cyanide, 1,2-dicyanoethylene, 1,1-diamino-2,2-dicyanoethylene, 1-amino-1-chloro-2,2-dicyanoethylene, 1-amino-2-bromo-2,2-dicyanoethylene, 1-chloro-1,2,2-tricyanoethylene, malononitrile dimer (2-amino-1,1,3-tricyanopropene), 4,4,5,5-tetracyano-2-pentanone, 1,1,2,2-tetracyanoethanesulfonic acid and its sodium, silver, and trimethylsulfonium salts; tricyanovinyl alcohol and its sodium, silver, cobaltous, and zinc salts; tetracyanoethane; 2,3,3,4,4,5,5,6,6,7-decacyano-2,7-dimethyloctane; and the like.

Oxidizing agents suitable for use in this invention are those which, when intimately mixed with the organic polycyano compound, will promote the self-sustained oxidation of the polycyano compound at the temperature of the oxidation reaction. To obtain practical amounts of thrust, it is generally necessary to employ an oxidizing agent having an oxidation potential of at least 0.7 volt. Oxidation potentials of oxidizing agents are discussed by W. M. Latimer in his book, "The Oxidation States of the Elements and Their Potentials in Aqueous Solution," Prentice-Hall, Inc., New York, second edition, 1952. Reactions of oxidizing agents having oxidation potentials in excess of 0.7 volt are particularly summarized in the part of Table 84 shown on pages 343–345 of this reference. It will be noted that the table starts with reducing agents and that the convention has been adopted of expressing the voltages of reducing agents as positive numbers and the voltages of oxidizing agents, with which we are here concerned, as negative numbers. The above-indicated range thus corresponds to those reactions shown in the table as having potentials starting from —0.7 volt and including those shown with negative potentials numerically larger than —0.7, i.e., to —3.0 volts and beyond.

The self-sustained oxidation of the polycyano compounds may reach temperatures above 5000° K. The propellant compositions of this invention may be ignited by heating to temperatures above about 200° C. (473° K.). Accordingly, the preferred oxidizing agents are those which cause oxidation of the fuel at temperatures in the range of 473° K. to 5000° K. Included among the many oxidizing agents which are useful in the propellant compositions of the present invention are sodium nitrate, sodium nitrite, sodium chlorate, potassium perchlorate, potassium nitrate, silver nitrate, potassium permanganate, ammonium chlorate, ammonium perchlorate, ammonium nitrate, potassium bromate, lithium chlorate, lithium perchlorate, lithium nitrate, sodium hypochlorite, sodium peroxide, sodium persulfate, potassium peroxymonosulfate ($KHSO_5$), sodium perborate, sodium perchromate, potassium dichromate, potassium perchromate, sodium periodate, potassium hydrogen iodate, chromium trioxide, cobaltic oxide, nitrogen tetroxide, perchloryl fluoride, liquid fluorine, liquid chlorine, liquid bromine, liquid oxygen, liquid ozone, and the like. Potassium and sodium equivalents of the sodium and potassium compounds above are equally operable as oxidants. All the corresponding lithium salts are also operable.

The rate of oxidation of the polycyano compound in the combustion of the compositions of this invention varies with the oxidizing potency of the oxidizing agent. Since this is characteristic of the individual oxidizing agent used, variation of the thrust developed by the propellant or the rate at which its energy is released may be made by variation of the oxidizing agent. To provide for fine control of the rate of oxidation, mixtures of two or more oxidizing agents as well as two or more polycyano compounds may be employed.

Numerous alternative means for using the compositions of this invention as propellants will be apparent. For example, a rocket tube may be charged nearly full with a mixture of a polycyano compound and an oxidizing agent which yields its oxidizing force at relatively high temperatures. The ignition zone of the rocket may then be packed with a mixture of a polycyano compound and a different oxidant, which latter mixture has a lower ignition temperature. The ignition zone mixture is readily ignited by heat and the heat of its combustion serves to initiate combustion in the main body of the propellant composition.

The propellant compositions may also be prepared in the form of a "hybrid" fuel mixture. In this embodiment, a solid organic polycyano compound is cast as a grain; fitting the sides of a propellant tube and having a central longitudinal hole. Through this hole, a liquid oxidant such as fluorine, oxygen, or ozone is pumped. The burning mixture of fuel and oxidant employs the hole as a combustion chamber, the hole becoming progressively larger as the fuel is consumed.

It is to be understood that the characteristic burning speed of the compositions of this invention may also be varied by incorporating reaction modifiers, inert adjuvants, or the like. Thus, the proportions of the polycyano compound and the oxidant may be varied widely, for example, from molar ratios of 10:1 to 1:10 of the stoichiometric amounts required for complete oxidation of the polycyano compound, the excess of either the polycyano compound or oxidizing agent serving as inert material. In actuality, the proportion of polycyano compound to oxidizing agent will normally lie in the range of 10:1 to 1:10 parts by weight since higher or lower ratios would, though operable, be functionally or economically unattractive, and this proportion range represents the preferred embodiment of the invention. Suitable binder compositions, such as silicones, polyurethanes, polysulfide rubbers, etc., may also be used.

Other combustible materials, such as finely divided aluminum, carbon, or cellulosic materials may also be incorporated into the propellant compositions in minor amounts. However, for practical utility as a propellant, the polycyano compound/oxidizing agent mixture must represent at least about 60% by weight of the composition. For maximum thrust, the molar ratios of polycyano compound to oxidizing agent are within the range of about 8:10 to 10:8 of stoichiometric values, and the mixture of polycyano compound and oxidizing agent comprises at least about 90% by weight of the total composition.

The propellant compositions of this invention may be employed in the form of wall-fitting charges, as bored or tubular or "grain" reactant shapes or may be otherwise physically modified as by variation in the degree of physical intimacy of mixture, i.e., particle size of separate reactants as is well known in the art.

The propellant compositions of this invention are useful not only for powering rockets and missiles but may also be used in boosters for such mechanisms, as well as takeoff assistants for aircraft.

While the invention is described in detail and illustrated with specific examples in the foregoing specification, it will be readily apparent to those skilled in the art that many variations and modifications may be made in the embodiments described without departing from the spirit or scope of the invention. It is intended, therefore, to be limited only by the following claims.

I claim:

1. A propellant composition consisting essentially of an organic polycyano compound in which the ratio of cyano groups to non-cyano carbon atoms in the molecule is at least 0.8:1 and an inorganic oxidizing agent therefor, said polycyano compound and oxidizing agent being present in a proportion of from 10:1 to 1:10 parts by weight and in the aggregate amounting to at least about 60% by weight of the propellant composition.

2. A propellant composition consisting essentially of an organic polycyano compound in which the ratio of cyano groups to non-cyano carbon atoms in the molecule is at least 0.8:1 intimately mixed with an inorganic oxidizing agent therefor, said polycyano compound and oxidizing agent being present in a proportion of from 10:1 to 1:10 parts by weight and in the aggregate amounting to at least about 60% by weight of the propellant composition.

3. A propellant composition consisting essentially of an organic polycyano compound in which the ratio of cyano groups to non-cyano carbon atoms in the molecule is at least 0.8:1 intimately mixed with an inorganic solid oxidant, said polycyano compound and oxidizing agent being present in a proportion of from 10:1 to 1:10 parts by weight and in the aggregate amounting to at least about 60% by weight of the propellant composition.

4. A propellant composition consisting essentially of tetracyanoethylene and an inorganic oxidizing agent therefor in a proportion of from 1:10 parts to 10:1 parts by weight based on the weight of the tetracyanoethylene, said tetracyanoethylene and oxidizing agent in the aggregate amounting to at least about 60% by weight of the propellant composition.

5. A propellant composition consisting essentially of tetracyanoethylene intimately mixed with an inorganic oxidizing agent therefor in a proportion of from 1:10 parts to 10:1 parts by weight based on the weight of the tetracyanoethylene, said tetracyanoethylene and oxidizing agent in the aggregate amounting to at least about 60% by weight of the propellant composition.

6. A propellant composition consisting essentially of tetracyanoethylene intimately mixed with a inorganic solid oxidant in a proportion of from 1:10 parts to 10:1 parts by weight based on the weight of the tetracyanoethylene, said tetracyanoethylene and oxidizing agent in the aggregate amounting to at least about 60% by weight of the propellant composition.

7. A propellant composition consisting essentially of tetracyanoethylene intimately mixed with a inorganic solid oxidant, the solid oxidant being present in an amount sufficient to supply at least 6 atoms of oxygen per molecule of the tetracyanoethylene, said tetracyanoethylene and solid oxidant in the aggregate amounting to at least about 60% by weight of the propellant composition.

8. A propellant composition consisting essentially of lithium 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide and an inorganic oxidizing agent therefor, said lithium 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide and said oxidizing agent in the aggregate amounting to at least about 60% by weight of the propellant composition.

9. A propellant composition consisting essentially of cuprous 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide and an inorganic oxidizing agent therefor, said cuprous 2-dicyanomethylene-1,1,3,3-tetracyanopropanediide and said oxidizing agent in the aggregate amounting to at least about 60% by weight of the propellant composition.

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