(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number: **0 661 252 A2**

(12) # EUROPEAN PATENT APPLICATION

(21) Application number: **94308329.5**

(22) Date of filing: **11.11.94**

(51) Int. Cl.[6]: **C06D 5/06**, C06B 41/00, C06B 47/00

(30) Priority: **10.12.93 US 165273**

(43) Date of publication of application:
05.07.95 Bulletin 95/27

(84) Designated Contracting States:
BE DE ES FR GB IT NL SE

(71) Applicant: **MORTON INTERNATIONAL, INC.**
100 North Riverside Plaza,
Randolph Street at the River
Chicago,
Illinois 60606 (US)

(72) Inventor: **Taylor, Robert D.**
356 South Rosewood Drive
Hyrum,
Utah 84319 (US)
Inventor: **Deppert, Thomas M.**
660 E. 390 South
Brigham City,
Utah 84302 (US)

(74) Representative: **Bankes, Stephen Charles Digby et al**
BARON & WARREN
18 South End
Kensington
London W8 5BU (GB)

(54) **Mixed fuel gas generant compositions.**

(57) In a gas generant composition comprising fuel and oxidizer, the fuel is a mixture of non-salt forms of azoles and alkali metal and/or alkaline earth metal salts of 5-nitrobarbituric acid and/or 5-nitroorotic acid.

EP 0 661 252 A2

The present invention is directed to non-azide gas generant compositions useful, for example, to provide inflation for automobile air bag restraint systems.

Gas generant compositions for inflating automobile air bag restraint systems presently predominantly use sodium azide as the fuel. Because of toxicity problems with respect to sodium azide and instability problems due to reaction of sodium azide with heavy metal contaminants, gas generants based upon alternative fuels have been proposed, although none, so far, has replaced sodium azide compositions to any appreciable extent.

One type of alternative fuel comprises tetrazoles, triazoles and their salts, as described, for example in U.S. Patent No. 4,909,549, the teachings of which are incorporated herein by reference. U.S. Patent No. 5,139,588, the teachings of which are incorporated herein by reference, describes mixtures of tetrazoles and alkali metal salts of tetrazoles in amounts sufficient to reduce amounts of toxic oxides, such as NO_x and CO, in the combustion gases. The mixture of non-salts and salts of tetrazoles combines advantageous features of each to provide an improved gas generant composition. Non-salts have the advantage in that the only combustion products attributed to the non-salt tetrazoles are gases, whereas the tetrazole salts also produce undesirable slag in the form of metal oxides. On the other hand, production of toxic oxides is temperature dependent, being produced in greater amounts at the higher combustion temperatures of the non-salt tetrazole compounds. Accordingly, salts, such as tetrazole salts, are used in conjunction with non-salt tetrazole compounds to control combustion temperature and thereby provide high gas output, without generation of high levels of toxic oxides.

An additional problem with gas generant compositions containing salts of tetrazole compounds, not discussed in U.S. Patent No. 5,139,588, is that such salts are hygroscopic. While care is taken in producing gas generant compositions to maintain them in a moisture-free environment and while gas generant compositions are generally maintained in a hermetically sealed container, any failure of such precautions may seriously affect the performance of a gas generant composition containing a hygroscopic fuel.

Another class of fuels suitable as azide alternatives are described in U.S. Patent No. 5,015,309 to Wardle et al. The fuels described therein are salts of 5-nitrobarbituric acid, 5-nitroorotic acid and 5-nitrouracil (which doesn't form a salt). Of these, 5-nitrouracil is disfavored, having a high burning temperature. The non-salt forms of 5-nitrobarbituric acid and nitroorotic acid might similarly function as fuels, but, like 5-nitrouracil, would have an unfavorably high combustion temperature. In fact, the free acid (non-salt) forms of this class of fuel is considered unfavorable relative to tetrazoles and triazoles in that they produce less gas on a mole per gram basis and burn at higher temperatures.

In accordance with the present invention, a gas generant composition comprises between about 30 and about 65 wt% of fuel, between about 5 and about 75 wt% of the fuel comprising a non- salt component selected from the group consisting of aminotetrazole, tetrazole, bitetrazole, triazole and mixtures thereof and between about 25 and about 95 wt% of the fuel comprising an alkali or alkaline earth metal salt of 5-nitrobarbituric acid, 5-nitroorotic acid or mixtures thereof; and between about 45 and about 65 wt% of an oxidizer.

The azole compound, i.e., the non-salt form of amino tetrazole, tetrazole, bitetrazole, triazole or mixture thereof provided, upon combustion with the oxidizer, a high volume of generant gas which is high in nitrogen (N_2) content. This fuel component produces only gases as combustion products.

The alkali metal or alkaline earth metal salt of 5-nitrobarbituric acid and/or 5-nitroorotic acid is a fuel component which lowers the combustion temperature of the composition (relative to use of the azole component alone). These salts are advantageous relative to azole salts in being non-hygroscopic. Thus, even if precautions against moisture fail, the composition of the present invention should continue to function efficiently as a gas generant.

It is generally desirable to produce as high a volume of gas as possible while achieving a combustion temperature which is preferably not above about 2000 °K so as to minimize formation of toxic oxides. To this end, the mixture is desirably as rich in azole compound relative to the salt of 5-nitrobarbituric acid and/or 5-nitroorotic acid as is consistent with achieving a desirably low combustion temperature.

The oxidizer includes at least one ammonium, alkali metal or alkaline earth metal nitrate, perchlorate or chlorate or a transition metal oxide.

The cations of the 5-nitrobarbituric acid salts and 5- nitroorotic acid salts and the cations of the oxidizer component preferably include both alkali metal cations and alkaline earth metal cations. The former, upon combustion, produce liquid slag components; the latter solid components. The combination of liquid and solid slag components promotes formation of clinkers which are easily filterable.

It is preferred to add SiO_2 up to about 10 wt%, preferably at least 0.5 wt%, to scavenge K_2CO_3, KOH, K_2O and related products from the combustion products, thereby forming K_2SiO_3, a liquid slag component. The silica may be provided as particulates or as a sole or joint component of fibers.

To promote combustion, up to about 10 wt% of a catalyst, preferably at least about 0.1 wt%, may be used. Catalysts may include, for example, transition metal oxides, transition metal chromates, transition metal vanadates, transition metal manganates, boron hydrides and mixtures thereof.

It is frequently desirable to provide a gas generant in pellet form. If the composition does not hold together by compaction alone, a binder in an amount of up to about 10 wt% may be included, typically at least about 1 wt% being used. Binders include, but are not limited to molybdenum disulfide, graphite, polytetrafluoroethylene, Viton ® (copolymer of vinylidene fluoride and hexfluoro propylene), nitrocellulose, polysaccharides, polyvinylpyrrolidones, polycarbonates, sodium silicate, calcium stearate, and magnesium stearate.

Additional components, which may be used at levels totalling about 10 wt% include, but are not limited to graphite fibers, alumina fibers, alumina/silica fibers, boron oxide and titanium dioxide.

The invention will now be described in greater detail by way of specific examples.

Examples

Compositions formulated as per the table below, were prepared by slurry blending the solids with about 25% to 30% water, drying the mixture thoroughly at 80 °C under vacuum (25 mmHg) and screening the composition through a 16 mesh screen. Pellets were prepared by compression molding at 40,000 psi. The table also list combustion temperature and gas output.

Formulations A, B, D, and E in wt% are given as examples. Formulations C, F, G, H and I are given for comparison.

Examples

	A %	B %	C %
KDL*	42.62	39.42	48.89
5-AT***	4.69	7.08	-----
$Sr(NO_3)_2$	46.64	47.89	44.12
SiO_2	6.05	5.61	6.99
	100.00	100.00	100.00
Combustion Temp. (°K)	1675	1765	1524
Moles of gas per 100g of generant	1.806	1.856	1.652

* KDL is potassium salt of nitrobarbituric acid (potassium dilituratе).
*** 5-AT is 5-aminotetrazole.

Examples

	D %	E %	F %
KNOA**	35.17	22.54	47.54
5-AT***	9.57	19.72	-----
$Sr(NO_3)_2$	50.92	56.32	43.04
SiO_2	4.34	1.42	9.42
	100.00	100.00	100.00
Combustion Temp. (°K)	1584	2000	1329
Moles of gas per 100g of generant	1.870	2.150	1.571

** KNOA is potassium salt of 5-nitroorotic acid.
*** 5-AT is 5-aminotetrazole.

Examples

	G %	H %	I %
5-AT	35.73	34.27	33.10
$Sr(NO_3)_2$	62.27	59.73	58.90
SiO_2	2.00	6.00	8.00
	100.00	100.00	100.00
Combustion Temp. (°K)	2661	2609	2571
Moles of gas per 100g of generant	2.417	2.315	2.346

Formulations A, B, D and E make a filterable slag, whereas by comparison formulations C and F in the above Table do not. Also, formulations A, B, D and E burn cooler than 5-AT (examples G, H and I) alone and produce more gas than either KDL or KNOA alone.

Claims

1. A gas generant composition comprising between 30 and 65 wt% of fuel and 35 to 65 wt% of an oxidizer,

   wherein 5 to 75 wt% of said fuel is selected from aminotetrazole, tetrazole, bitetrazole and triazole, and

   25 to 95 wt% of said fuel is selected from alkali or alkaline earth metal salts of 5nitrobarbituric acid, 5-nitroorotic acid and mixtures thereof.

2. A gas generant according to claim 1 which contains at least 45 wt% of said oxidizer and not more than 55 wt% of fuel.

3. A composition according to claim 1 or claim 2 wherein said oxidizer comprises at least one ammonium, alkali metal or alkaline earth metal nitrate, perchlorate or chlorate or transition metal oxide.

4. A composition according to any preceding claim further comprising between 0.5 and 10 wt% SiO_2.

5. A composition according to any preceding claim further comprising between 0.1 and 10 wt% of a catalyst selected from transition metal oxides, transition metal chromates, transition metal vanadates, transition metal manganates and mixtures thereof.

6. A composition according to any preceding claim further comprising between 1 and 10 wt% of a binder.