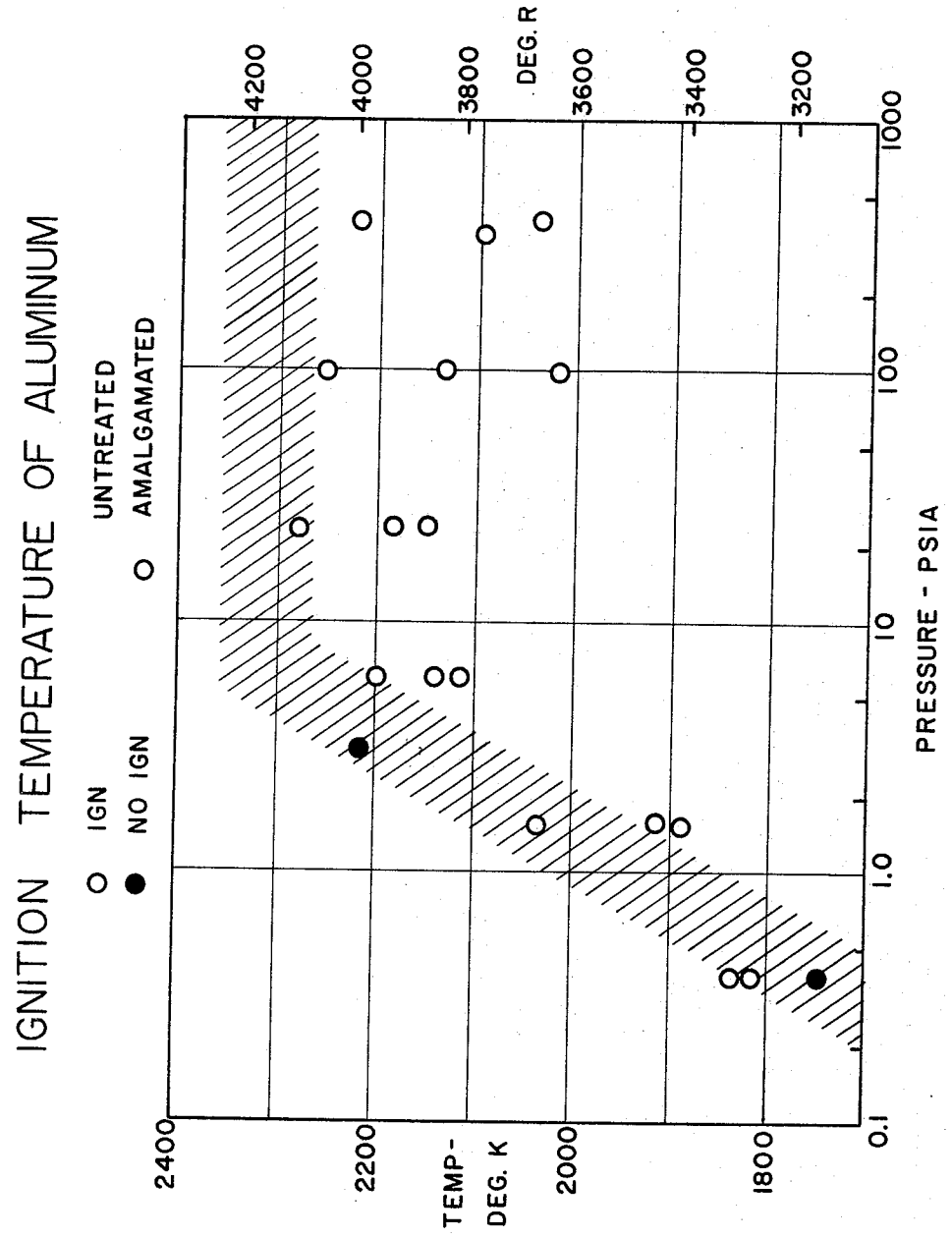
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METHOD OF OPERATING A THRUST PROPELLANT MOTOR USING AMALGAM COATED METAL PARTICLES

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METHOD OF OPERATING A THRUST PROPELLANT MOTOR USING AMALGAM COATED METAL PARTICLES

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This invention relates to metal ignition and more particularly to improving the rate and lowering the temperature of ignition of metals on which protective oxide coatings tend to form when the surface thereof is contacted with oxygen and/or oxygen-containing environments.

This invention is particularly directed to fuels or fuel systems comprising or utilizing metals of the type described as a component.

Metal containing fuels made in accordance with the teachings contained herein are capable of use in a wide variety of transportation and military equipment. They are especially useful as high energy propellants for reaction thrust propellant motors or engines, such as missile, rocket, jet, and ram-jet engines. In motors or engines of the type described, fuel is burned in a combustion chamber and the products of combustion are allowed to escape through a jet, thereby producing a driving force.

An object of this invention is to improve the rate and temperature of ignition of metals on which protective oxide coatings form when the surface thereof is contacted with oxygen and/or oxygen-containing environments, i.e., oxygen containing gases, liquids, or solids, and includes improving the rate and temperature of ignition of compositions containing such metals.

Another object of this invention is to improve the efficiency of combustion of metals of the type described, including compositions containing such metals.

It is another object of the present invention to provide improved metal additives for solid, liquid and/or hybrid rocket propellants.

Another object of the present invention is to increase the delivered performance of metal containing propellants by reducing ignition delay.

Another object of this invention is to provide more complete combustion of metal containing fuels in combustion chambers of thrust propellant engines.

Still another object of the present invention is to provide surface activated metal particles which may be incorporated into solid, liquid, or hybrid high energy propellant systems to improve the performance characteristics of such systems.

Other objects of the present invention will in part be obvious and will in part appear hereinafter.

It is known that the formation of a protective oxide coating on solid, high energy metals retards ignition of such metals until a much higher temperature is reached than would be required in the absence of such a protective oxide.

It is known that when a protective oxide coating forms on the surface of a solid metal being heated to ignition temperature in the presence of oxygen or an oxygen containing environment, a temperature approaching or substantially equal to the melting point of the protective metal oxide coating must be reached before ignition will occur. Since the melting point of the protective oxide is usually substantially higher than the ignition temperature of the base metal, it will readily be appreciated that the presence of the protective oxide considerably hampers the ignition performance of the base metal.

According to this invention, it has been discovered that

formation of protective oxide coatings on metals or particles thereof undergoing combustion may be avoided or substantially eliminated by amalgamating the surfaces of the metals or metal particles prior to subjecting them to the combustion or ignition environment.

Amalgamation of the metal surface may be readily accomplished by contacting the metal or a surface portion thereof with mercury in the liquid or gaseous phase, or in the form of mercury salts or alloys, as will be more fully described hereinbelow.

Amalgamation not only prevents the formation of a protective oxide layer but also provides a surface amalgam which is generally more active than the pure metal itself, thereby further enhancing the ignition properties of the metal.

Because of the high activity of the surface amalgam, it will generally be necessary to store the metal particles of this invention in inert environments which are free or substantially free of oxygen, e.g., under vacuum, in inert gases such as nitrogen, hydrogen, helium, argon, neon, krypton, and the like, or under inert liquids, such as hydrocarbons, e.g., hexane, or in non-reactive binders as in a solid propellant or hybrid fuel grain.

Although particularly directed to metal containing fuels or fuel systems, this invention is generally useful whenever it is desired to burn or otherwise react metals to produce power, heat or light.

Thus, metals, e.g., magneisum or aluminum, treated in the manner described herein could be used to good advantage in camera flash bulbs and flares, as well as in other pyrotechnic display devices. The metals or metal particles described herein could also be used in high temperature cutting or welding torches, e.g., oxyacetylene torches. In such torches, the metal particles could be injected into the oxygen stream.

As a further and important application, metals treated in accordance with the teachings contained herein may be used as components in the so-called metallized explosives, i.e., explosives in the nature of dynamite comprising conventional oxidants, e.g., ammonium nitrate, and metallic particles.

In all of these applications, it is the rapid, low temperature ignition properties of the solid metals of this invention which are exploited to enhance the operational performance and the efficiency of the devices or compositions containing the metals as components.

Although the invention will be particularly described in connection with thrust propellants, it should be understood that the basic concepts are equally applicable to other, more general applications of the type described.

When metals which form protective oxide are heated in an oxygen-containing environment, a protective oxide coating forms and builds up as the temperature is raised to ignition. The protective metal oxide coating adversely affects the performance of the metal, since it results in a longer ignition time and higher ignition temperature than would be required were the coating not present. In thrust propellant motors utilizing metal containing fuels, the fuel has a relatively short, limited residence time in the combustion chamber of the engine. Protective oxide coatings which form on the metal components of such propellants cause delay in ignition time and high ignition temperatures, thereby leading to incomplete combustion and reduction in the delivered performance of the fuel.

The use of metals having amalgamated surfaces as fuel additives in solid, liquid, or hybrid thrust propellant systems will increase the delivered performance through reduction in ignition delay, which will lead in turn to more complete combustion of the metal in the combustion chamber.

Further, hypergolicity of some bipropellant liquid rockets will be improved or obtained by the addition of the activated (amalgamated) metal particles of this invention to the fuel phase of the propellants.

The invention is generally applicable to improve ignition performance of all metals, including alloys, combinations or mixtures of metals, which form an alloy with mercury, i.e., an amalgam.

Typical of the metals which do form amalgams and to which this invention applies are:

Beryllium	Magnesium
Aluminum	Titanium
Zirconium	Chromium

including alloys or mixtures of the foregoing, and alloys or mixtures containing at least one of the foregoing metals. Particularly advantageous results are achieved when the invention is practiced with: aluminum; beryllium; alloys or mixtures of aluminum and beryllium; alloys or mixtures of any of the listed metals with aluminum; alloys or mixtures of any of the listed metals with beryllium; and alloys or mixtures of any of the listed metal with lithium.

Aluminum and beryllium, including alloys or mixtures comprising beryllium and aluminum, treated as described herein, constitute preferred species of the invention. In this category may also be mentioned alloys or mixtures of aluminum, beryllium, or mixtures of aluminum and beryllium, with lithium.

The surfaces of the metals may be amalgamated in a variety of ways.

Thus, mercury could be added to a molten form of the metal or alloy, and the resulting admixture solidified.

Alternatively, the metal or alloy may be contacted or otherwise treated with mercury in liquid (pure or alloy) or vapor form, or may be treated with appropriate aqueous or organic solutions of mercury salts, e.g., mercury iodide, chloride, bromide, nitrate, bromate, chlorate, fulminate, and the like, to thereby form an alloy of the metal with mercury, i.e., an amalgam, at the surface of the metal or alloy.

It will be appreciated that the teachings contained herein are applicable regardless of the form, shape, or size of the metal or alloy being treated.

Typically, however, the metal or alloy will be in a form which will provide a relatively high surface to volume ratio, such as a sphere, cylinder, torroid, and the like.

Ordinarily the metal or alloy will be in the form of spheres, irregularly shaped granules, wires, strips, and the like, which have, as an inherent characteristic, a minimum size in one direction, e.g., a diameter. As used herein, the term metal particle or particles is intended to encompass a wide variety of shapes, including spheres, irregularly shaped granules, wires, strips, slivers, chips, and the like.

The burning efficiency of metals is a function of many variables. For any given metal, however, the burning efficiency can be generally said to vary inversely with the size of the particle sought to be burned—the larger the particle—the lower the burning efficiency.

For rapid burning rate and high burning efficiency, the metal particles should possess, as an inherent characteristic, a small minimum dimension in one direction, e.g., a diameter, so as to provide a large surface to volume ratio.

One of the advantages of the present invention resides in the fact that it permits utilization in thrust propellants of metal particles having a minimum dimension which is larger than has heretofore been generally possible.

Heretofore in thrust propellant technology, metal particles with minimum dimensions of 0.010" to 0.025" were about the largest that could be practically used, even with the biggest motors available, because of the problems encountered with ignition of larger particles. With the amalgamated particles of this invention, use of

metal particles having a minimum dimension of up to 1" or even larger is entirely possible, although the particles will ordinarily have a minimum dimension of up to ½".

Amalgamated metal particles of the type described having a minimum dimension ranging from about 0.001 micron to about ¼" constitute preferred embodiments of this invention.

Dimensions of the metal particles other than the minimum dimension are not particularly significant. Thus, wires or strips having a minimum dimension as described supra and lengths of up to several miles or even longer are entirely within the purview of the invention and entirely feasible for use with the larger rocket engines.

Metallic particles having amalgamated surfaces will ignite more readily in the combustion chamber of a rocket motor and therefore will achieve more complete combustion within the chamber, thereby leading to greater delivered performance. Since the amount of mercury required to produce the amalgamated surface is small, the presence of mercury will not reduce the potential impulse of the metal particles appreciably.

The nature of this invention will be made more clear by the following example.

Example

A large number of runs were made to ascertain the ignition temperature of untreated vs. amalgamated aluminum wire.

The aluminum wires utilized were 99.99% pure and had a length of 4 inches and a diameter of 500 microns.

Amalgamation was accomplished by placing a drop of dilute (5%) alcoholic mercuric chloride on the center of the wire. The wire was then subjected to high vacuum to evaporate the alcohol. The wires treated in this manner were maintained under vacuum until testing to prevent premature oxidation.

The drawing is a graph of the results obtained from these tests.

In the drawing, the wire ignition temperature in degrees Kelvin ("Deg. K") is plotted as ordinate vs. pressure in pounds per square inch absolute (p.s.i.a.) as abscissa.

The shaded portion of the graph represents a band of over 60 data points taken with untreated, pure (99.99%) aluminum wire in a gas containing 30 to 100 percent oxygen. The width of the band gives an indication of the spread of the experimental data. In the horizontal portion of the shaded area the coefficient of variation is less than 1, corresponding to a standard deviation of $\pm 21°$ K.

The circles represent individual data points for the amalgamated wire. Amalgamation, as described above, was accomplished by treatment with dilute (5%) alcoholic mercuric chloride.

In the region below 10 p.s.i.a. there is no statistical significance to any differences between the treated and untreated wires. Above 10 p.s.i.a., and therefore in the region of practical significance, there is a statistically significant difference in the ignition temperature of treated and untreated aluminum wire. From the graph it can be shown that the amalgamated aluminum wires have an ignition temperature on the order of 170° K. lower than the untreated wire.

As will be clear from the example and accompanying graph, the amalgamation treatment of this invention leads to a surprising reduction in ignition temperature of metal particles to be used as components in rocket propellants. This reduction in ignition temperature, as described, supra, is immediately translatable into greater delivered performance when the amalgamated metal particles are utilized as a component in fuels, e.g., rocket propellants.

Metal treated in accordance with this invention can be used in the solid grain of a hybrid or tripropellant rocket to provide improved performance. The binder or matrix material which holds the metal particles in the grain will

also serve to prevent reaction of the activated surfaces in the atmosphere during storage.

Normal storage of the activated metal will however have to be under an inert atmosphere in order to prevent pre-ignition reaction.

Metal treated in the manner described may also be used in a slurry or gel of a non-oxidizing fluid in a bipropellant liquid rocket. In many cases hypergolicity will result from the use of metals whose surfaces have been activated by amalgamation.

Use of an amalgamated metal in solid rockets will also be advantageous but in such formulations, some form of protection for the treated metal will ordinarily be required to prevent reaction with the solid state oxidizer during mixing, curing, or storage.

Besides the metal, rocket propellants will comprise an oxidizer or utilize an oxidizer as a component. By oxidizer is means oxygen or a chemical capable of being decomposed to yield oxygen. When solid, the fuel will comprise a binder. Other constituents of rocket propellants are well known in this art. Fuels, i.e., chemical substances capable of chemically combining with oxygen to release heat, other than the metal component may also be used. It should be understood that the teachings contained here are equally applicable to any thrust propellant system which utilizes metal as a high energy additive.

Typical rocket propellant systems in which metals treated as described herein can be used to good advantage are described in United States Patents Nos. 3,133,842 and 3,112,609.

The invention in its broader aspects is not limited to the specific details shown and described but departures may be made therefrom within the scope of the accompanying claims without departing from the principles of the invention and without sacrificing its chief advantages.

What is claimed is:

1. In a method of operating a thrust propellant motor, wherein fuel comprising metal as a component is burned in a combustion chamber and the products of combustion are allowed to escape through a jet, thereby producing a driving force, the improvement which comprises utilizing, a metal selected from the group consisting of beryllium, aluminum, zirconium, magnesium, titanium, chromium, mixtures and alloys of the foregoing and mixtures and alloys containing at least one of the foregoing, said metal having a surface coating which is an amalgam of said metal, characterized by an ignition temperature which is lower than the ignition temperature of said metal itself.

2. The method of claim **1** wherein the fuel comprises, in addition to said metal, an organic binder.

3. The method of claim **1** wherein the fuel comprises, in addition to said metal, an oxidizer.

4. The method of claim **1** wherein the fuel comprises, in intimate admixture with said metal, an oxidizer and an organic binder.

5. A method for producing power which comprises reacting with an oxidizer a metal selected from the group consisting of beryllium, aluminum, zirconium, magnesium, titanium, chromium, mixtures and alloys of the foregoing, and mixtures and alloys containing at least one of the foregoing, the surface of said metal having a coating which is an amalgam of said metal characterized by an ignition temperature which is lower than the ignition temperature normally associated with the metal itself.

6. The method of producing power which comprises contacting aluminum metal with an oxidizer, the surface of said aluminum metal having a coating which is an amalgam of aluminum having an ignition temperature which is substantially lower than the ignition temperature of aluminum metal.

7. In a method for preparing a fuel composition containing a high energy metal selected from the group consisting of beryllium, aluminum, zirconium, magnesium, titanium, chromium, mixtures and alloys of the foregoing and mixtures and alloys containing at least one of the foregoing the improvement which comprises contacting the metal with a member selected from the group consisting of mercury and salts of mercury under conditions such that an amalgam of said metal, characterized by an ignition temperature which is lower than the ignition temperature of said metal itself, forms as a coating on the surface of said metal.

8. The improvement of claim **7** wherein said metal comprises lithium as a component.

9. A high energy fuel system comprising, as a component, a metal selected from the group consisting of beryllium, aluminum, zirconium, magnesium, titanium, chromium, including mixtures and alloys of the foregoing, and mixtures and alloys containing at least one of the foregoing, said metal having a surface coating which is an amalgam of said metal characterized by an ignition temperature which is lower than the ignition temperature of said metal itself.

10. The system of claim **9** wherein said member is aluminum.

11. The system of claim **9** wherein said member is an alloy of beryllium and aluminum.

12. The system of claim **9** wherein said member is an alloy of aluminum with a member selected from the group consisting of beryllium, magnesium, zirconium, titanium, silicon, chromium, lithium and mixtures of the foregoing.

13. The system of claim **9** wherein said member is an alloy of beryllium with a member selected from the group consisting of aluminum, zirconium, magnesium, silicon, titanium, chromium, lithium, and mixtures of the foregoing.

14. The high energy fuel system of claim **9** wherein said metal comprises lithium as a component.

15. As a new article of manufacture, a metal selected from the group consisting of beryllium, aluminum, zirconium, magnesium, titanium, chromium, including mixtures and alloys of the foregoing, and mixtures and alloys containing one of the foregoing, said metal having a surface coating which is an amalgam of said metal, characterized by an ignition temperature which is lower than the ignition temperature of the metal itself, said metal having a shape which includes a minimum dimension which is less than about 1 inch.

16. The metal particle of claim **15** wherein the metal comprises lithium as a component.

17. An article in accordance with claim **15**, wherein said metal has a shape which includes a minimum dimension which ranges between about 0.001 micron and ¼ inch.

18. The system of claim **9**, wherein said member is beryllium.

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