

Review

Energy-Intensive Materials with Mechanically Activated Components

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Abstract: The production and study of highly dispersed aluminum-based powders represents one of contemporary science's priority fields. This is primarily driven by the practical necessity to develop new materials, a feat that, in some cases, can only be achieved through the utilization of powdered components. This article presents the results of the mechanochemical treatment method employed to obtain highly reactive aluminum particles. It also includes a comparative analysis of aluminum particles generated through various methods and their respective properties. Furthermore, the application of these highly reactive aluminum particles in energy-intensive materials is discussed.

Keywords: aluminum; energy-intensive materials; mechanochemical treatment



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1. Introduction

Currently, the search for energy-intensive materials is carried out in all directions, and these are not only artificially created materials, but also materials of natural origin. Energy-intensive systems are a class of materials with a high content of stored chemical energy, which can be used to solve certain practical problems. Typical classes of energy materials are explosives, pyrotechnic compositions, solid propellants, and other types of fuels (diesel fuel, gasoline, etc.). The advancement of powder technologies and the growing demand for dispersed systems utilizing metals and their alloys have amplified interest in the investigation of these materials. Consequently, one of the present challenges lies in regulating their reactivity to develop materials with superior operational characteristics [1]. Energetic materials have found practical application since the discovery of gunpowder, and contemporary explosives and rocket propellants are essential in various industrial sectors. The destructive potential of explosives for non-military purposes is harnessed in the mining industry, road construction, and within rocket propulsion systems, etc.

2. Physical and Chemical Properties of Aluminum and Methods for Changing the Structure and Composition of the Surface Layer of Aluminum Particles

Aluminum powders are extensively employed in various applications such as explosives, manufacturing high-energy combustible mixtures, and solid propellants for rocket engines. Optimizing the combustion parameters for these applications is a crucial and pressing objective, owing to the high specific heat of combustion exhibited by aluminum powders [2].

Aluminum (CAS number: 7429-90-5, Table 1) is a lightweight, silver-white, paramagnetic metal that is readily malleable, machinable, and castable. It possesses excellent thermal and electrical conductivity and exhibits resistance to corrosion, primarily due to the

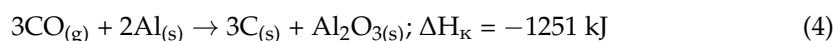
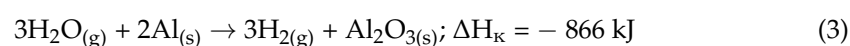
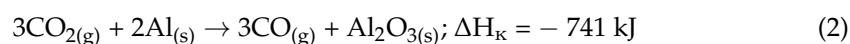
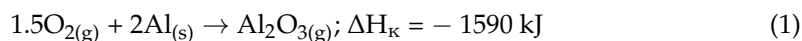
rapid formation of robust oxide films on its surface. These oxide films serve as a protective barrier, preventing further interaction with the surrounding environment.

Table 1. Basic physical and chemical properties of aluminum [3].

Property	Significance
Density	2700 kg/m ³
Melting temperature	660 °C
Specific heat capacity	24.15 J/(mol·K)
Gas thermal conductivity	210.25 J/(m·s·K)
Standard enthalpy of formation	0 kJ/mol
Melting heat	322–394 kJ/kg
Molecular weight	27 g/mol

Aluminum has a high enthalpy of combustion, is non-toxic, and has a low cost compared to other potential reactive materials, making it an excellent candidate for energy material applications (Table 1). The complete oxidation of aluminum to aluminum oxide (Al₂O₃) gives a specific energy of about 31 kJ/g. Comparison with the detonation energy of explosives such as trinitrotoluene (TNT), octogen (HMX), or hexogen (RDX) shows that aluminum has great potential for use as a structural energy material, even in the presence of an external oxide film on the surface of the particles. The comparison shows that the enthalpy of combustion of aluminum is two times higher than the enthalpy of combustion of an explosive, using the example of complete decomposition of TNT. Aluminum is difficult to ignite, which leads to the realization of only a part of its potential energy (about 6–10% of the theoretically realizable energy potential) when used in explosives. Fragmentation of aluminium, combined with ignition delay and relatively long burning time compared to explosives, plays a limiting role in the limited conversion of energy stored in the material [4]. If the fragments produced by the explosive loading of aluminum particles are too large for unhindered ignition, the energy stored in these fragments will be kinetically removed from the explosion zone, i.e., the energy stored in these particles will not be realized. In this case, it practically does not improve the explosion.

As mentioned above, aluminum is a potentially very energy-intensive material when evaluating fuel efficiency relative to its mass. Its ability to oxidize in the atmosphere in a variety of media, including oxygen, water vapor, carbon monoxide, and carbon dioxide, makes it attractive for use in applications such as solid rocket motors and explosives. The addition of aluminum to these systems increases the energy density with negligible losses. The oxidation of aluminum with various oxidizing agents can be characterized by the following main reactions [5]:



The combustion of aluminum with pure oxygen yields the most substantial heat release, with the subsequent reaction of aluminum with carbon monoxide also generating significant energy. Equations (2) and (3) illustrate that the reactions between aluminum and carbon dioxide, as well as aluminum and water vapor, produce comparatively lower energy outputs. Aluminum can undergo oxidation in diverse environments, the rate at which it oxidizes and fully consumes depends significantly on various factors, including temperature, particle size, the presence of oxidizing agents, and pressure [6].

Because of its inherent reactivity, “pure” aluminum will undergo oxidation even at room temperature, resulting in the formation of a protective oxide layer on its outer surface. This oxide layer is typically just a few nanometers thick. The outer alumina layer plays a crucial role in regulating the initial heating and ignition of the aluminum particles and can significantly impact the delay in ignition. In order for the oxidizing agent to reach the aluminum core, the oxide layer must be destroyed, since it has a melting point of about 2350 K. Immediately after explosive detonation, the outer passive shell begins to rupture under load at a high strain rate, and the resulting fragments initially contain pure aluminum, since the oxide layer has not yet formed. Pure aluminum particles will begin to react most intensely at temperatures above the melting point of aluminum, approximately 930 K.

The main factors influencing the combustion process of aluminum particles are temperature, pressure, and fragment diameter. The fragment diameter of the aluminum particle is an important factor to consider as it determines the basic characteristics and burning time of the particles. It was shown [5] that larger aluminum particles (usually more than 20 μm) burn in the classical mode with a limited time, wherein aluminum evaporates and forms a para-phase separated by a flame at a certain distance from the surface of the particle, as shown in Figure 1.

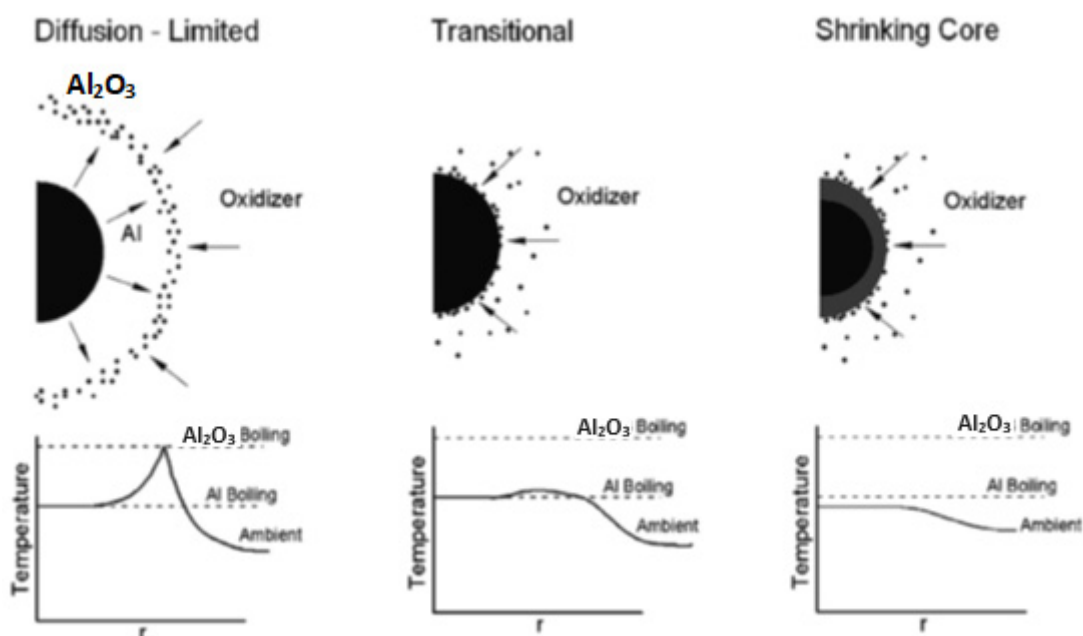


Figure 1. Scheme describing the structure of the flame and temperature profiles for various modes of combustion of aluminum particles [5].

Experimental data from various researchers have shown that the correlation of aluminum burning time provides a weak dependence on pressure and temperature, and also predicts that the burning rate of aluminum in H_2O should be half that in oxygen, while the burning rate of aluminum in CO_2 is approximately one fifth of the burning rate in oxygen. With this slower burning rate in these oxidizing atmospheres, the aluminum fragments are unlikely to rapidly release energy with a high explosive detonation. The large diameter of the fragments also affects ignition, as ignition requires the particle to be heated to the ignition temperature. As the surface shell is destroyed, fragments of larger particles are quickly removed from the explosion zone and move faster than particles of smaller sizes. Therefore, larger fragments may not have sufficient residence time in the elevated temperature blast zone to ignite and provide improved blast efficiency. As the particle size is reduced to below 10 μm , the characteristics of the combustion mode and burning time of the particles change. For particles about 3 to 11 μm in size, experimental data showed a transition to lower diameters with combustion time correlations, as well as

a stronger dependence of combustion characteristics on both pressure and size of oxidized particles [7].

Interest in aluminum powders is due to their possible use in various energy systems. In addition, beryllium and its combustion products are toxic, while boron is refractory and its viscous low-melting oxide blocks the access of the oxidizer to the combustion zone. Therefore, aluminum is the most optimal fuel for metallized mixed compositions. However, a dense oxide film on the surface of aluminum particles makes it difficult to ignite the compositions and causes a low burning rate. Various methods are used to remove or reduce the thickness of the oxide film.

There are two ways to activate aluminum and its alloys: chemical and physical.

One of the chemical methods is the use of mercury salts, which have a promoting effect on the process of chemical dissolution of aluminum powder in nitric acid. In this case, the reaction rate depends not only on the concentration of mercury and HNO_3 salts and temperature, but also on the shape and size of the aluminum particles subjected to activation (powder, wire, or bulk sample) [8]. When studying aluminum samples of various geometric shapes, it was found that with an increase in the surface/volume ratio of aluminum powder, the ratio of its dissolution rate to area decreases, while the ratio of dissolution rate to unit mass remains unchanged. In this case, not only a surface effect occurs that is probably associated with the cyclic reduction and oxidation of mercury on the surface of aluminum particles, but also a “volumetric” effect, including the formation of an aluminum-mercury amalgam.

Another traditional method of metal activation is the treatment of aluminum with iodine vapor [9]. Aluminum is activated by heating in an argon medium in the presence of crystalline iodine, which by penetrating through the defects of the oxide surface film and forming aluminum iodide leads to the destruction of the oxidized surface.

A method has been developed for activating aluminum powder by interacting with water under supercritical conditions (temperature 220,900 °C, pressure 20–40 MPa) [10]. In aqueous media under supercritical conditions, the rate of diffusion of water molecules is very high; accordingly, intense mass transfer develops, which leads to a rapid reaction between aluminum and water. By varying the reaction conditions, various end products can be obtained: AlOOH , $\text{Al}(\text{OH})_3$, $\gamma\text{-Al}_2\text{O}_3$, $\alpha\text{-Al}_2\text{O}_3$.

Of the other methods of chemical activation of aluminum, it should be noted the methods of treating the metal surface with a solution of NaOH or KOH to remove the oxide film [11]. The reactivity of aluminum with respect to water is also achieved by its fusion with dehydrated alkali metal hydroxide with copper additives (up to 5 wt. %) and by activating aluminum with other metals (Ga, Bi, Pb, Sn, In). At present, there are developments using various aluminum alloys in hydrogen generating units [12].

Physical methods of aluminum activation are based on the impact of external mechanical and electrophysical loads. For example, during the electric explosion of an aluminum wire in an argon medium (203 kPa) with the addition of hydrogen (10% vol.), an activated aluminum powder is formed [13]. When this powder interacts with water on the surface of the particles, aluminum oxide is obtained with a specific surface area of 200–300 m^2/g .

When grinding aluminum powders by electric explosion in a solution of a water-soluble polymer (a solution of polyethylene oxide in dioxane), the oxide film is replaced by a polymer [14] which protects the aluminum surface from oxidation. In this way, encapsulation of the activated state is achieved. The use of nanometric aluminum turned out to be effective in reducing the problems associated with the low reactivity of micrometric Al, but its use at the industrial level is hampered by the high cost of production (~500 \$/kg for Russian nAl), wherein the requirement of specialized production is the need to control the accurate dispersion procedure and the need to ensure a high level of security.

Among the activation processes, processing methods based on mechanical action are the most promising. They are based on the grinding of one or several powders simultaneously in mills of various types (friction, centrifugal, or planetary), the so-called mechanical reactors [15]. Mechanochemical treatment leads to an increase in the content of active

metal depending on grinding parameters (time, mass ratio of balls and powder, diameter of grinding bodies, type and mass fraction of the modifying agent, typology, and mass fraction of the activating agent).

For example, it was shown in [16] that aluminum powder processed in a mixture with finely dispersed graphite in vibration mills exhibits high reactivity when interacting with water. The maximum amount of hydrogen corresponding to the complete oxidation of aluminum is released at a mass ratio of aluminum/carbon (90/10). In this case, the dose of absorbed energy during mechanical processing is 10.8 kJ/g. It is assumed that at the first stages a homogeneous Al/C composite is formed, in which particles of finely dispersed aluminum are stabilized in a medium of finely dispersed graphite. During long-term mechanical treatment, the chemical interaction of aluminum with graphite is carried out with the formation of the Al_4C_3 crystalline phase.

Simultaneously with the physical impact during processing in a mill, an activating change in the surface of aluminum particles can be carried out as a result of the presence of special activating additives in the processed powder, as well as aluminum fusion with activating reagents. The chemical processes of fusion during the interaction of grinding working bodies with the material to be ground are possible as a result of local short-term heating to high temperatures [17].

Mechanically, it is possible to obtain nanopowders with a particle size from 5–10 to 200 nm. The disadvantage of this method is the contamination of the powder with the material of the lining and grinding media, as well as the limited minimum particle size and the wide size distribution of the powder particles. The advantages of this method include relatively high productivity, the potential for obtaining nanocomposites, and its superior environmental friendliness. The mechanochemical production method holds promise for practical implementation due to its high efficiency, enabling the production of the desired product in large quantities.

The use of nano-sized aluminum powders offers extensive opportunities for scientists, engineers, and technologists in the realm of developing cutting-edge materials and technologies, as well as entirely new instruments and devices. Nano-sized aluminum powders find applications in high-energy materials, radio-absorbing coatings, the chemical industry, organic electronics, optical technologies, aerospace, medicine, pharmaceuticals, cosmetology, and manufacturing.

3. Physical and Chemical Foundations of Mechanochemical Synthesis

The mechanochemical treatment of materials both organic and inorganic using grinding equipment is presently considered an innovative approach for producing novel materials tailored to specific properties and various functional applications, including energy-intensive systems, catalysts, sorbents, construction materials, etc. [18,19]. Mechanochemical treatment (MCT) involves various processes and phenomena in addition to particle dispersion. These include the following examples: Deformation of Crystals: The mechanical forces applied during MCT can cause the crystals of the material to deform; Formation of Defects: MCT can lead to the creation of numerous defects within the material's crystal lattice; Change in Microblock Size: The size of microblocks that constitute a crystal may change as a result of MCT; Shear Stresses: Mechanical forces applied during MCT can induce shear stresses within the material; Aggregation of Crystallites: Crystallites can aggregate or come together due to the mechanical action; Heat Release: Energy is released in the form of heat during MCT, often leading to a local increase in temperature; Local Temperature and Pressure Changes: MCT can result in localized temperature and pressure changes within the material; Emission of Light and Electrons: Some materials undergoing MCT may emit light and electrons; Phase Transformations: Phase changes or transformations may occur within the material; Amorphization: The material may become amorphous or lose its crystalline structure; Breakage of Chemical Bonds: The mechanical forces can lead to the breaking of chemical bonds; Acceleration of Diffusion Processes: Diffusion processes within the material can be accelerated; Formation of Active

Centers: MCT can create active centers on newly formed surfaces. These diverse effects and transformations make mechanochemical treatment a powerful method for modifying the properties and structure of materials for various applications [20,21]. All the processes involved in mechanochemical treatment contribute to an increase in the activity of the processed solids. This heightened activity is a consequence of several factors, including the following examples: **Vibrationally and Electronically Excited States of Interatomic Bonds:** The mechanical action leads to the generation of vibrationally and electronically excited states within the interatomic bonds, making the material more reactive; **Mechanically Stressed and Broken Bonds:** The mechanical forces applied during treatment can result in the stressing and breaking of chemical bonds, which can create more reactive sites; **Formation of Free Radicals:** Mechanochemical processes can generate free radicals, which are highly reactive species with unpaired electrons; **Coordinatively Unsaturated Atoms:** Some atoms within the material may become coordinatively unsaturated, making them more prone to chemical reactions; **Various Structural Defects:** The formation of structural defects in the material introduces sites with altered reactivity; **Ionization of Particles:** Particles of matter can undergo ionization during mechanochemical treatment, leading to the formation of ions and charged species; **Stabilization of Electrically Charged Centers:** The treatment can stabilize electrically charged centers, which can influence reactivity. These cumulative effects enhance the reactivity and chemical activity of the processed solids, making mechanochemical treatment a valuable technique for the modification and synthesis of materials with tailored properties [22]. The most commonly used are planetary mills. Mechanochemical treatment of powdered mixtures or pulps in energy-intensive apparatuses is accompanied by the formation of vacancies, dislocations, and defects. If in the processed systems the stored energy will be spent on providing chemical reactions in the reactor-activator, then the process is called mechanochemical, and if it is used outside the reactor, then the process is called preliminary mechanical activation [23].

The process of mechanochemical treatment in general can be represented as triaxial loading and shear deformation at the contacts between solid particles. It should be noted that during mechanical treatment, the impact on particles is of a pulsed nature, i.e., chemical and physical processes do not occur during the entire stay in the mechanical reactor, but only at the moment of impact and for some time during the relaxation of the stress field [24]. The relaxation time can be different depending on the substance. In some cases it is seconds, and in others it is months and years.

Mechanical activation (MA) in energy-intensive devices is a complex process that depends on many factors, such as the type of mill, the duration of activation, the heating temperature during processing in the mill, the ratio of the mass of the powder to the mass of the balls, the chemical composition of the reagents, etc. During mechanical activation, these factors make it difficult to predict nonequilibrium processes (phase formation, phase transition). Nevertheless, in [25] a physical and mathematical model was proposed that describes the process of mechanochemical synthesis in a macroscopic approximation. The model is based on the equations of thermal action in the reactor-activator, chemical kinetics, and changes in the reaction surface. Two options were considered: a one-stage process—synthesis in the reactor itself and a two-stage process—synthesis using pre-activation. Results are obtained to describe the processes of mechanochemical treatment of heterogeneous systems under various conditions.

The mechanoactivation of mixed systems leads to the formation of mechanocomposites in which the initial components or the resulting products of their interaction are bound and evenly distributed among themselves. The structure of mechanocomposites depends mainly on the properties of the initial components of the processed charge. Depending on the nature of the components, three main types can be distinguished: plastic-plastic, brittle-brittle, and brittle-plastic. Descriptions of the mechanism of formation of layered composites, including two different plastic components, are shown in the works of Benjamin and Wolin [26]. In the primary collision of grinding bodies with a metal particle, it is flattened, i.e., surface area increases relative to its volume.

For grinding solids, various apparatuses are used, which differ in the type of impact on solids: crushing, splitting, abrasion, impact, etc. Modern dispersing devices usually combine two or more types of action. Efficient MA of a solid occurs mainly in devices with high energy density. Such conditions can be implemented in mills with medium and high loading speeds, such as centrifugal planetary, impact-reflex, and others.

Figure 2 shows the modes of movement of the load of a centrifugal mill. Particles in the drum of a centrifugal planetary mill (CPM) participate in two movements: relative to movement around a common axis and rotation around its own axis. During MCT, there are three modes of movement of grinding bodies: (a) when sliding, the material moves in the opposite direction of rotation of the drum around its axis; (b) shock, which is described by a curvilinear trajectory of the material, located in the form of a segment; (c) vortex, in which the material moving along a curvilinear trajectory fills the entire volume in the drum.

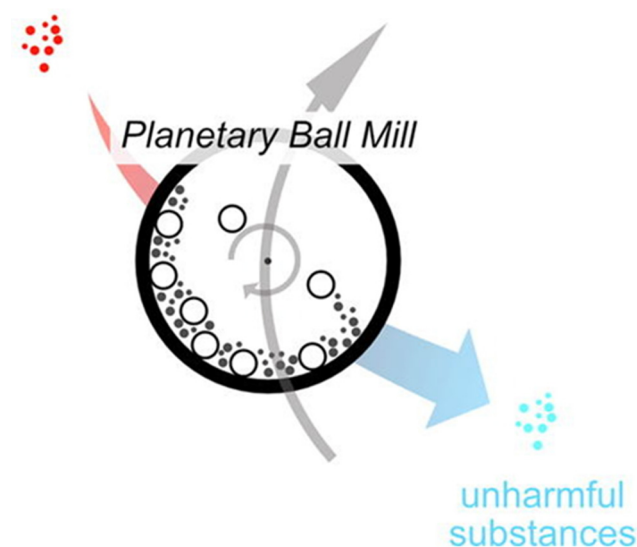


Figure 2. Scheme of movement in the CPM drum [27].

The theoretical speed of the grinding body at the moment of its separation from the drum wall at an angle of 180° or close to it is [28]

$$v = v_1 + v_2 = 2\pi[\omega_1(R_1 + R_2) + \omega_2 R_2], \quad (5)$$

where, ω_1 , ω_2 are the number of revolutions around the common axis and the planetary axis, R_1 , R_2 —radii of general and planetary rotations.

Expressing this equation in terms of the characteristics of the mill, we obtain

$$v = 2\pi\omega_1 R_1 [k + (m + 1)], \quad (6)$$

where, k is the kinematic characteristic of the mill ($k = \omega_1 / \omega_2$), m —geometric characteristic of the mill ($m = R_1 / R_2$).

For metal powders, most often, the average size of the dispersed materials is not in the nanometer, but in the micron and submicron regions. In this case, the powder particles contain nanograins and can be considered as a textureless polycrystalline material with a high density of grain boundaries [29]. Often, crystalline materials ground in ball mills have a particle size of up to 5–30 nm.

In the “brittle-plastic” systems, at the initial stage of grinding, the particles of ductile metal powder are deformed as a result of ball-powder-ball interaction, while brittle oxides or intermetallic phases are fragmented and crushed. The course of the alloying process in the “brittle-viscous” system depends on the solubility in the solid state of the brittle component in the plastic matrix. For example, as a result of the dispersion of

pure zirconium Zr (ductile) and NiZr_2 (brittle), a metal is formed—an oxide system or an intermetallic compound.

Thus, mechanical activation is a complex and multi-stage physical and chemical process at each stage of which processes and phenomena occur, to one degree or another, determining changes in the state and structure of the material and its energy capabilities. Starting from the moment of elastic deformation to brittle fracture, from the moment of accumulation of defects to complete structural rearrangement in an ultrasmall particle, the energy state of the substance changes.

Enhancing the reactivity of aluminum particles through mechanochemical treatment is of particular significance in the production of metallized solid rocket propellants (SRP). Most currently known solid rocket fuels contain up to 15–20% aluminum powder as a metal fuel, which can significantly elevate the combustion temperature, the rate of product flow, and, consequently, the efficiency of rocket fuel [30]. This results in an extended flight range and the capacity to carry a larger payload. Activating aluminum powders allows for the enhancement of the ballistic properties of propulsion systems across a broad range of values.

Rossi [31] wrote in his works that one of the advantages of using aluminum powder as an ingredient in solid rocket fuel is its high heat release during combustion. This elevated heat release during metal oxidation increases the flame temperature, thereby boosting the specific impulse value in aluminized fuel by approximately 10%. Additionally, there is another crucial characteristic to consider: the unstable combustion of aluminum and aluminum-based systems, which becomes significant when employing aluminum in condensed systems [32,33]. The inclusion of highly dispersed aluminum can enhance the propellant capabilities of compositions when compared to pure explosives. The addition of nano-sized aluminum also has the potential to increase the propellant capabilities of explosives, albeit the observed increase is somewhat lower compared to micron-sized aluminum due to the higher oxide content on the surface of nanoparticles.

4. The Use of Mechanochemical Treatment of Materials as a Way to Create Highly Active, Modified Systems

Mechanochemical treatment (MCT) occupies an extremely important place among the many methods of influencing a solid body in order to change its state, structure, and in general, the properties of a substance, in particular, reactivity, and forms the basis of many technological processes for obtaining a material with desired properties [34].

During the treatment of materials in mills, the dispersion of particles is accompanied by a change in their energy state, i.e., activation or interaction between the components of the mixture, resulting in the synthesis of new materials. During mechanochemical treatment in mills of high energy intensity, the formation and migration of dislocations occurs, as well as the appearance of a microplasticity region with a partially destroyed structure. After the elastic and plastic deformation of the particles, the formation and development of cracks and the destruction of the solid body (i.e., dispersion) occur. At the same time, interatomic bonds are broken, and a fresh surface is formed. The emergence of a highly active short-lived state on the surface, the breaking of chemical bonds, and an increase in the surface during the grinding of powders occurs wherein each of the processes individually and collectively can cause chemical interaction between the components of the mixture to be ground.

All processes of structural rearrangement and chemical interaction begin on the surface. The rate of heterophase reactions is known to be proportional not to the amount of substance, but to the surface area of the solid [35]. This surface is enriched with defects and radical centers, which due to their high reactivity, enter into various interactions with gaseous, liquid, and solid components, while the surface structure changes (modification). Due to the high density of defects in the dispersed particle, there is a high probability of formation of amorphous compounds on its surface.

Modification of powder particles directly in the grinding process is one of the directions of mechanochemical treatment of inorganic materials. This process makes it possible to simultaneously bring the material to the required degree of fineness, modify the surface of the dispersed particles, and obtain plastic, well-shaped masses. Mechanochemical treatment expands the range of modifiers used. In particular, the use of carboxylic acids makes it possible to purposefully form active centers on the surface of various layered silicate systems. The volume of scientific papers and publications in the press, as well as those presented at international conferences on mechanochemical modification, has increased significantly in recent years, which reflects the prospects and effectiveness of work in this direction (Table 2).

Table 2. Summary of Al Powders under Investigation [36].

Nomenclature	Source, Production Technique, and Nominal Size D	s, m ² /g
Al-01a	EEW technique (uncoated Al); D = 0.15 µm; produced in 2003	15.3
Al-02a	EEW technique (uncoated Al); D = 0.15 µm; produced in 2003	17.5
Al-02b	EEW technique (uncoated Al); D = 0.15 µm; produced in 2002	14.4
Al-02c	EEW technique (uncoated Al); D = 0.15 µm; produced in 1999	12.6
Al-03a	Pneumatic mill (uncoated Al); D = 0.2 µm	2.0
Al-03b	Pneumatic mill (uncoated Al); D = 0.4 µm	1.5
Al-03c	Pneumatic mill (uncoated Al); D = 0.8 µm	0.9
Al-04a	Planetary mill Al + C 20%; D = 2 µm	9.554
Al-04b	Planetary mill Al + 20% (C ₂ H ₃ OH) _n ; D = 30.5 µm	14.648
Al-04c	Planetary mill Al + 3% C ₁₇ H ₃₅ COOH; D = 38 µm	5.31

The production of aluminum nanopowder from discarded electrical aluminum wire, containing a minimum of 99.5% aluminum, is achieved through electroerosive dispersion in distilled water. This process operates at a pulse repetition rate ranging from 95 to 105 Hz, with an electrode voltage range of 90 to 110 V and a capacitor capacity of 65 µF. Subsequently, the solution is subjected to centrifugation to separate large-sized particles from the nanopowder. This approach leads to reduced energy consumption and enhanced environmental sustainability [36].

The difference between mechanochemical processing and other methods of aluminum production and modification is the production of energy-intensive aluminum-based powders without the use of an inert medium in the presence of modifying additives: polyvinyl alcohol, stearic acid, and graphite (Table 2). The exclusion of an inert environment when producing energy-intensive powders by mechanochemical processing reduces the economic component of the process and is a more technological solution for mass production. In addition, the exclusion of an inert environment makes it possible to reduce the time of mechanochemical treatment to 20 min, compared to similar studies carried out in an inert environment, for which the time of mechanical chemical treatment was 90 min.

Research in the field of energy-intensive materials includes not only volumetric synthesis, engineering, and materials science, but also includes microscopic studies of molecular dynamics and structure (i.e., understanding the molecular level of these systems). To understand the combustion process of energy-intensive materials, the chemical processes of decomposition must be studied in detail. It is necessary to study in detail the individual stages of a chemical reaction, the dynamics of dissociation, and the energy released during combustion reactions.

High-energy materials are of interest for increasing energy release in various energy systems. Numerous studies have been carried out on the addition of reactive metals to high explosives and solid propellants to provide higher energy densities and energy yields [37]. The metal in the composition of the fuel is an additive that increases the energy of the

composition. An increase in the heat output and specific impulse of the fuel is due to the high thermal effect of metal oxidation, as well as a decrease in the average molecular weight of gaseous combustion products as a result of the decomposition of H_2O and CO_2 during their interaction with the metal. For example, some mixed solid rocket propellants (SRFs) for special purposes, such as hydrosensitive SRFs, can contain up to 80% metal, which in this case is the main fuel [38]. The energy characteristics of metals and metal-containing substances increase the efficiency of SRFs by increasing the density of the fuel. In addition, the combustion products of metals in the form of oxides are the stabilizers of the combustion process. Powerful explosives such as hexogen, octogen, etc. are used as energy additives both in ballistic fuels and in SPRs, acting as additional oxidizers.

One paper [39] describes a comparative analysis of the ballistic efficiency of rocket fuels containing either Al or Zr or ZrH_2 . The research results show that the replacement of aluminum with zirconium or its hydride provides an increase in the rocket speed, practically, for any SRFs compositions. However, the melting point of zirconium is higher than that of aluminum, and therefore stronger igniters are required for zirconium-based compositions.

The authors of [40] studied the combustion rate and flame structure of SRFs compositions with a wide particle size distribution of the starting components. However, these works focus on the size distribution of ammonium perchlorate particles and do not pay due attention to the role of the size factor of aluminum particles on the burning rate of aluminumized mixed fuels. An examination of the impact of incorporating powdered aluminum, including nanosized particles, into explosive compositions' propelling performance is detailed in [41]. The inclusion of finely dispersed aluminum can enhance the propelling efficiency of these compositions compared to pure explosives. Moreover, the addition of nanosized aluminum has the potential to improve the propelling performance of explosives, as well. However, it is worth noting that the observed enhancement is slightly less pronounced when using nanosized aluminum in comparison to micron-sized aluminum. This discrepancy can be attributed to the higher presence of oxide on the surface of the nanoparticles, which affects their reactivity.

One paper [42] shows the efficiency of using aluminum, magnesium, and aluminum-magnesium (AM) alloys in a propulsion hydrojet engine. In this work, aluminum-magnesium alloys were obtained by mechanical processing. Fuel compositions based on polybutadiene with terminal hydroxyl groups (hydroxyl-terminated polybutadiene—HTRB) with the addition of metal combustible components demonstrated a high burning rate.

Powdered aluminum is an effective fuel for energy-intensive materials, and in terms of mass calorie content, it is more than twice superior to magnesium, although it is inferior to boron and beryllium in this indicator, but significantly exceeds the latter in density. Therefore, aluminum is the most optimal fuel for metallized mixed compositions. Currently, there are various industrial types of aluminum powders such as PA4, ASD, PAP, etc. But due to the dense oxide film on the surface of aluminum particles, the ignition of the compositions is difficult and a low burning rate is observed. In combustible condensed systems, an important factor is the rapid ignition of the fuel and the high calorific value of the oxidation reaction, which largely depends on the state and structure of the surface of aluminum particles.

5. Features of the Structure and Properties of SHS Systems with Mechanically Activated Components

In recent years, a large number of works have been devoted to the effect of mechanical treatment on the combustion parameters in the SHS regime. First of all, there is a significant decrease in the temperature and time of the beginning of the process of interaction of reagents, an increase in the rate of chemical reactions, and an increase in the completeness of the transformation for pre-activated systems [43,44]. Ignition temperature reduction has been established for a large number of systems. The degree of reduction depends on both the specific material and the time-force conditions of machining. It has been established that the temperature of the beginning of the interaction of reagents in the system after

mechanical activation decreases by 100–300 °C or more. In this case, the nature of the interaction of the reagents changes significantly. For example, for the Al-NiO system, it was found [45,46] that the aluminothermic reduction begins significantly below the melting temperature of aluminum, i.e., the process proceeds in the solid phase mode. The beginning of the process of chemical interaction decreases and there is a significant increase in heat release in the system. Thus, for a mixture of Mo-10% B, it was found [47] that activation for 10 min leads to an increase in the heat released and the rate of heat release by more than three times. The increase in heat release is associated with the “release of mechanical energy” stored in the process of grinding powders, as well as with a decrease in diffusion paths due to the formation of chemical gradients at the nanolevel.

In some cases, on the contrary, a decrease in the thermal effect was observed after activation of the system, for example, for Fe-45% Al [48]. The authors of the work explain this fact by a change in the leading SHS reaction; in contrast to the non-activated mixture, for which the Fe_2Al_5 formation reaction is thermodynamically preferable, and after the activation of the system, the formation of iron monoaluminide (FeAl) is observed, the thermal effect of which is much lower. The change in the leading reaction is due to a decrease in the energy barrier for the second reaction due to an increase in the concentration of defects in the structure of activated particles. In some cases, the decrease in the thermal effect of the synthesis process is due to the formation of an interaction product between the mixture components in the mechanically activated mixture, which reduces the exothermicity and thermal conductivity ahead of the combustion front, which reduces its propagation rate and the local thermal gradient.

Since much attention is paid to the combustion rate (propagation of the reaction front) when studying the regularities of SH-synthesis, during the mechanical processing of the system attention is also paid to the change in this characteristic of the synthesis process. As a rule, mechanical activation (MA) leads to an increase in the burning rate in both low- and high-calorie systems. An increase in the burning rate after mechanical activation was observed in the Ti-C and Ni-Al systems [49] by four and thirty times, respectively. In both cases, melting of the metal reagents occurs. The authors attribute this to the refinement of the microstructure of the reaction medium. Under other activation conditions, a decrease in the burning rate was observed upon complete conversion of the system. These differences are associated with the conditions of heat transfer between the particles of the reaction mixture. The authors explain the decrease in the burning rate by the fact that large aggregates of particles formed during MA require more time for heating, which slows down the propagation of the SHS process. The increase in the burning rate can be associated with the morphological features of the composite particles.

In this case, the uniformity of the distribution of reagents in the charge mixture, the grinding of particles, the increase in the contact between particles, and the increased defectiveness of particles are the most important structural factors of the effect of MA on SHS. The main factor in increasing the burning rate is an increase in the specific surface area, the number of bonds, and adsorbed oxygen on the particle surface. The combustion temperature is related to the burning rate, according to the classical concepts of SH-synthesis. As the speed increases, the combustion temperature increases. However, with the involvement of MA, this dependence is not always exactly fulfilled. For a number of systems, for example, Ni-Al [50], it was found that after activation the combustion temperature decreased below the formation temperature of a low-melting eutectic. The decrease in the combustion temperature of mechanically activated mixtures is associated with a decrease in the temperature of the beginning of the reaction due to the stored energy of mechanical activation, as well as the loss of part of the chemical energy of the system as a result of the interaction of components during mechanical processing. In some cases, the mechanism of interaction between the reagents changes after mechanical activation. The intensification of endothermic reactions, in particular melting, after MA leads to a decrease in the combustion temperature.

Usually, when studying the mechanism of MA influence on heterogeneous processes, two types of influence are considered: microstructural and energetic [51]. The microstructural effect is reduced to grinding the reagents and increasing the contact area between them, which leads to an acceleration of the reaction. The energy effect is associated with the accumulation of additional energy in the crystal lattice due to the accumulation of residual elastic or plastic deformations during mechanical action.

Mechanical activation makes it possible to change the conditions for SHS processes. A decrease in the temperature and time of the beginning of the interaction of reagents in pre-activated systems, as well as an increase in the rate of chemical reactions and an increase in the completeness of the transformation lead to a change in the mechanisms and kinetics of the processes of interaction of the components. The influence of MA on SHS is a consequence of the complex influence of a number of factors: the grinding of reagents, an increase in the contact surface, an increase in the concentration of defects in the crystal structure, the presence of crystallization centers of new phases, and reaction products of the components of the reaction mixture.

Thus, the use of mechanical action to change the structure, state of the material, and its energy intensity is an effective way to influence the process of SH-synthesis and obtain materials with desired properties and functionality. The use of preliminary mechanochemical treatment will promote the formation of various phase structures. By adjusting the parameters of mechanochemical treatment and the state of activated reagents, it is possible to control the reaction pathways in the subsequent SH-synthesis, purposefully changing the thermal effect of reactions and obtaining products with specified morphology and sizes of structural components. Along with well-known methods [52], this approach to the integrated use of mechanochemical treatment and SH-synthesis will expand the possibilities of obtaining materials with a variable set of properties.

6. Conclusions

1. Examples of mechanochemical processing of aluminum are analyzed. The influence of mechanochemical treatment on the microstructure and reactivity of activated and modified aluminum has been studied.
2. Prospects for the use of highly active aluminum in energy-intensive materials are presented. The influence of powdered aluminum additives, including nano-sized particles, on the propellant ability of explosive compositions, and on the combustion and speed of high-energy compositions, as well as on the rapid ignition of fuel and the high calorific value of the oxidation reaction of metallized solid rocket fuels is analyzed.

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