Integrated Quality Analysis Method of Aluminum for Composite Propellant Production

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Abstract

Quality analysis of Aluminum powder for solid fuel composite propellant was carried out to determine the critical parameters of the composite propellant characteristics. Two types of aluminum were analyzed for quality, including bulk and true density using the Archimedes principle, particle shape, and size using SEM (Scanning Electron Microscope), and purity using XRD (X-ray Diffraction), particle area with BET (Brunnauer-Emmet-Teller) and BJH (Barret-Joyner-Halenda) adsorption isotherm. Composite propellant made with 87.5% solid content and 18% AL content, then tested the characteristics of the propellant. The results of the analysis of the quality of aluminum for composite propellant raw materials have a very large influence on the quality of the resulting propellant, so an integrated, quick, and efficient quality analysis is needed. The critical parameters of Aluminum quality as propellant fuel are density, purity, particle shape and size, and porosity/surface area. Fast and efficient integrated analysis can be performed using new instruments, such as shape and size analysis with SEM, crystal structure and purity analysis with XRD, density analysis with a densitometer. AL2 which has a smaller particle size, better density and shape parameters value, less pores and surface area than AL1 can be used to produce a higher quality composite propellant.

***Keywords**: Aluminum, Composite Propellant, Quality Analysis*

1. Introduction

Composite propellants are widely used in modern solid rockets because they have high combustion energy, moderate burning rate, low sensitivity, good mechanical properties at operating temperatures, and are economical (Wibowo, 2018). Conventional composite propellants generally use ammonium perchlorate as an oxidizing agent, aluminum powder material as solid fuel material, and polybutadiene as binder material. Many kinds of research on propellant composition have been carried out to obtain propellants that have very high energy (Wibowo, 2019); (NourEldin et al., 2020). A propellant composition is sometimes difficult to replicate because of the different characteristics of the raw materials used. For example, aluminum powder has quite a lot of parameters to form its characteristics. The key parameters that affect the quality of aluminum for solid propellants are different from the key parameters for other applications. Therefore, an integrated analysis method is needed for the quality of the aluminum powder, especially as a propellant raw material. Several key parameters determine the quality of aluminum powder as a raw material for composite propellants. The characteristics of propellants are not only combustion energy but include physical properties (hardness, surface smoothness), ballistics (burning rate), energetic (combustion energy), chemical properties (sensitivity), ease of processing (casting and curing), and mechanical properties (Gligorijević). et al., 2014).

This paper discusses the characteristic parameters of aluminum and their effect on propellant performance. Then the development of an integrated and inexpensive aluminum quality measurement method was carried out. It is hoped that the development of aluminum quality analysis methods can be used to determine the quality of aluminum as a raw material for composite propellants and meet the standards of several manufacturers.

2. Methodology

2.1. Related Works

2.1.1. Parameter quality characteristics of aluminum powder

The key parameters of aluminum powder as raw material for composite propellants are particle shape and size, purity, actual density and surface density, pore volume and surface area, and the number of layers. The aluminum powder will increase the combustion temperature of the propellant. In general, the greater the amount of aluminum powder will affect the energy of the propellant produced (Babuk et al., 2009).

The rate of the combustion reaction of propellant and aluminum is influenced by the concentration of aluminum and the size of the aluminum grains. The smaller the particle size of aluminum powder, the more surface area it will have. Based on the kinetic theory of the propellant combustion reaction, the larger the aluminum surface area, the faster the reaction rate (Kiani et al., 2020). The shape of the particles affects the combustion kinetics because of the contribution to the surface area of the particles (Forte et al., 2018). Particle shape and shape variables (shape factor, roundness, aspect ratio, and solidity) of aluminum powder can also determine the quality of the propellant during the combustion process. Particles with a spherical shape will have a better mass transfer rate and particle velocity in the nozzle than non-spherical shapes such as ellipsoidal, and cube (Wang and Yang, 2019). The surface properties of the material such as surface area, and pore properties will affect the mechanical and hygroscopic properties at the microstructural level of the composite material (Joshi et al., 2017). Powders with larger volume and pore surface area can increase the friction coefficient and wear coefficient of the material where affects the service life and storage period of the material (Li and Olofsson, 2017).

Other variables such as the aspect of processibility brought about from the use of aluminum powder in composite solid propellants are also influenced by the nature of the density (bulk) and the actual density (true). Powder flow characteristics will affect the processibility. Powder materials with a larger bulk density will have a greater resistance to flow than powder materials with a smaller bulk density (Forte et al., 2018), the true density variable shows the opposite phenomenon where powders with the ability to flow the better one have a higher actual density. The actual density of the higher powder will also affect the mechanical properties of the resulting material (Seyda, Herzog, and Emmelmann, 2017).

2.1.2. Existing parameter measurement methods.

Bulk density is the density of the powder material under dry conditions which can be expressed as the mass of the sample in dry conditions in a container with a fixed volume. The presence of water molecules in the powder material will affect the voids between the particles which affect the volume of the powder material. The bulk density measurement method for aluminum powder samples can use ISO 17828, while the ASTM D7481-18 method has a weakness in measuring the bulk density of dry samples (Eisenbies et al., 2019).

The true density can be measured by using the Archimedes principle of fluid which has a theoretical density lower than the density of the material to be measured. The fluid that can be used is a liquid with a simple molecular structure such as water molecules. Fluids with complex molecular structures, such as polymers, cannot be used for density measurements because of the influence of excessive buoyancy forces (Wilson, 2012). The aluminum powder will be immersed in the liquid fluid and will have a contact area with the liquid fluid. Changes in the detected volume can be expressed as a sample volume that can be used to measure sample density (Mohazzab, 2017).

Measurement of particle shape variables from SEM (Scanning Electron Microscopy) images can be analyzed with the ImageJ application. ImageJ application is an application that can be used to measure shape variables from particle images analyzed by SEM. Variables such as size, shape factor, roundness, and aspect ratio are used as variables to characterize the shape of the particle material (Mahmoud et al., 2021). There are many particle characterization methods with ImageJ, where the segmentation between particles, and the skeletonization zone are important parameters in the analysis of SEM images with ImageJ (Hutauruk, Bura, and Wibowo, 2020). BET and BJH isotherms can be used in determining the surface properties of aluminum powder materials which can be analyzed by the Multi-Point method. Wherefrom multi-point analysis can be obtained particle surface area, pore-volume, pore area, constant C as the level of sample affinity in the gas adsorbate adsorption process (Mel'gunov and Ayupov, 2017; Bardestani, Patience and Kaliaguine, 2019).

2.2. Problem Definition

The quality of propellant manufactured during the propellant processing is directly tied to the properties of the main components such as the aluminum powder as metal fuel, the Ammonium perchlorate (AP) as the oxidizer, and the liquid content (binder, curing agent, and additive). The quality of the aluminum powder as one of the main compositions needs to be determined. Currently, the lack of method employed to characterize the properties of aluminum powder proved to be a stumbling block in the development of composite propellant.

The quality of different aluminum powders, evaluated by employing parameters such as density (true density and bulk density), particle (particle size and shape), crystal (structure and purity), and the surface area profiles can be analyzed using appropriate tools and methods. Aluminum powder's quality directly influencing the quality of manufactured composited propellant. Higher quality propellants can be manufactured using superior quality materials. In the case of high solid loading propellant, the quality of aluminum powder resulted in a stark difference in the propellant characteristics and mechanical properties.

In regards to processibility, the propellant slurry rheological properties (the viscosity and working life) are influenced by the shape and size of solid loadings other than the binder's nature. A finer grade of Aluminum powder employed in the propellant mixing and casting process resulted in preferable rheological properties of the propellant slurry. The longer working life of propellant slurry is preferable in the aerospace industry can be achievable by using the higher quality of aluminum powders.

2.3. Method

2.3.1. Materials

The aluminum powder material used is an aluminum powder for composite propellant produced by Dalian-China, produced by Hanwa-Korea, and produced by LAPAN-Indonesia as a comparison. Al materials were directly analyzed without special treatment.

2.3.2. Tools

The tools used are analytical digital balance and aluminum tube for density analysis, SEM analysis with Phenom World® Pro X Desktop (maximum electrical optical magnification range up to 150.000 times), XRD analysis with Shimadzu® MAXima_X XRD-7000 (3kW) with scintillation detector, and OneSight Wide-Range high-speed detector, BET analysis with Quantachrome® ChemBET Pulsar TPR/TPD with N_2 gas, input pressure of 70-140 kPa, with furnace temperature up to 1100^{o}C and dual-filament diffusion type detector.

2.3.3. Aluminum quality analysis

Experiments on measuring bulk density and true density were carried out in the HTPB Laboratory of Rocket Technology Center at a temperature of 25oC, with humidity ranging from 60-70%. Sample test with SEM instrumentation was carried out in the Liner and Thermal insulation instrument laboratory; and sample testing with XRD and BET-BJH instrumentation was carried out at the integrated laboratory of the Diponegoro University, Semarang. Data processing was carried out using the MS Excel application and SEM image sample processing was carried out with the ImageJ 1.53e application, BET isotherm data processing, and BJH method calculations using the Quantachrome® ASiQwin™ application (Hutauruk, Bura and Wibowo, 2020).

Bulk density analysis

Analysis of bulk density using the ISO17828 method by measuring the mass of powder in a container whose volume has been measured and filled with powder without undergoing a tapping process (Amidon, Meyer, and Mudie, 2017). The aluminum powder sample was put into an aluminum tube whose volume had been measured with a glass funnel until it was full. The surface of the powder was then leveled with a spatula and the sample mass was weighed. The difference between the measured mass and the mass of the empty tube will be expressed as the mass of the aluminum powder sample at a predetermined tube volume. Furthermore, the sample in an aluminum tube was heated in an oven at a temperature of 100^{o}C for 1 hour. After 1 hour, the sample in the aluminum tube was measured again. The difference between the mass of the sample after and before being heated in the oven is the mass of water contained in the aluminum powder, which can be expressed as a hygroscopicity index. The specific gravity (bulk density) can be expressed as the mass of the sample in the tube after being heated in the oven divided by the volume of the tube.

True density analysis

Analysis of real density (true density) uses the Archimedes principles method (Fluid Displacement Method) with the selection of aquabides as the fluid used in density measurements (Mohazzab, 2017). The aluminum powder sample was put into a 10 mL measuring cup with a spatula twice (± 1-2 mL) and the mass of the sample was weighed in the measuring cup with an analytical balance. The sample in the tube was then added with 5 ml of distilled water with a measuring pipette and the sample volume and sample mass were measured. The volume of aluminum samples can be calculated as the difference between the sample volume after the addition of aquabides and the volume of aquabides added.

Particle Size and Shape Analysis

Identification of particle size and particle shape of aluminum powder was carried out by SEM. SEM can detect the morphological shape of the particles, with the image scale that can be adjusted to get the best resolution (Kim, Han, and Han, 2020). The aluminum powder sample was analyzed by SEM, then the SEM image obtained was analyzed by Image processing technique using the watershed segmentation method to separate particles that stick to each other or close together (Kornilov and Safonov, 2018), then analyzed the particle size (mean size and particle size range), Perimeter, Shape Factor (Circularity), Aspect Ratio, Roundness, and Solidity with Java-based ImageJ software.

Crystal Structure and Purity Analysis

Analysis of purity, lattice parameters, and crystallite size can be carried out by XRD. The purity of a metal sample can be measured as an index of similarity between the sample diffractogram and the standard diffractogram of the sample (single crystal, 99.99% purity). The lattice parameter (a) can be calculated for Aluminum crystals (fcc) by the reflection selection rule, the crystallite size can be calculated using the Scherrer equation for each diffraction peak. The crystallite size calculated by the Scherrer equation can be expressed as the sample crystallite size at the peak that does not violate the reflection selection rules (Holder and Schaak, 2019).

Particle Surface Area Analysis

The particle surface area analysis was carried out by the gas adsorption method. Two theories of adsorption kinetics models BET (Brunnauer-Emmet-Teller) and BJH (Barret-Joyner-Halenda) are used in interpreting the adsorption isotherm. In the physical adsorption process at very low relative pressure, the surface site that will be covered first with the adsorbate is the more energetic site. On the surface of heteroatomic particles such as organic materials or materials containing impurities, there will be variations in the adsorption potential. The more the number of energetic sites that bind to the adsorbate, the higher the adsorbate pressure, where this will increase the residence time of the adsorbate at the energetic site so that it will increase the probability that the next adsorbate will be bound to the site that still binds to the adsorbate causing the adsorption to become multi-layered. The ability of BET theory which can calculate the amount of adsorbate material needed to form single layer adsorption even though single layer adsorption is never formed can be used to measure particle surface area and adsorbent affinity on adsorbate molecules (Allen, 1990).

The presence of pores will cause the presence of high-energy adsorption sites, where these sites are different from the adsorption sites on the surface. In addition, the BET theory ignores the lateral interactions between adsorbate molecules in multilayer adsorption. The presence of pores will cause the BET model to be less accurate to be used in measuring the surface area, and the size and surface area of the pores of the material. The BJH method uses the desorption isotherm of N2 and calculates the amount of adsorbate that is desorbed against the average pore size involved in the desorption process. After evaporation of the condensed adsorbate liquid (core), the single layer can be calculated by the thickness equation. The thickness of the layer will decrease every time there is a reduction in pressure. The measurement of the amount of gas desorbed is equivalent to the amount of vaporized core liquid. Pores in solid materials can be classified based on the pore width. Pores with a width below 2 nm, between 2 nm to 50 nm, and above 50 nm are classified as Micropores (Mi), Mesopores (Me), and Macropores (Ma) (Allen, 1990).

2.3.4. Testing of Composite Propellant with AL Addition

The AL test in the propellant was carried out by making a composite propellant based on Ammonium perchlorate (AP). The composition used is AP with a size of 200 and 50 microns mixed with a weight ratio of 4:1. The binders used are Hydroxyl Terminated Polybutadiene (HTPB) and Toluene Diisocyanate (TDI) with a weight ratio of 14:1. The number of AL and AP used was 18% and 69.5%, with a binder of 12.5%. The AL used are AL1 and AL2. The mixer used is a 4 kg horizontal mixer equipped with a vacuum pump, stirrer, and heater.

The mixture of propellant ingredients was mixed in a mixer for 1 hour at a stirring speed of 60 rpm, a temperature of 50oC, and in a vacuum. The propellant slurry was put into a tubular mold with a diameter of 5 cm and a length of 20 cm. The mandrel is inserted into the mold to make a hollow grain with an inner diameter of 2.5 cm. The mold was put in the oven for 3 days at 60oC. The mandrel is removed, the hard propellant is removed from the mold. The propellant is then tested for its characteristics including specific gravity, combustion rate, mechanical properties, and propellant performance.

3. Result and Analysis

3.1. Density

Analysis of the bulk density of a particle can be carried out by several methods: direct measurement of the density of the sample in dry conditions in the volume of the measuring container as in the ISO 17828 method, and indirect measurement method by measuring porosity with the gas adsorption method. Bulk density can be calculated as the relationship between porosity and true density which can be expressed as equation (3-1) where ρ_b is the bulk density, f is the porosity, and ρ_s is the true density of the material.

$$\rho_b = (1 - f) \cdot \rho_s \qquad (3\text{-}1)$$

The bulk density analysis method chosen is the direct measurement method for the density of the sample in dry conditions in the volume of the measuring container according to the ISO 17828 method because of the practicality and accuracy of measurement of the method so that it is used as a general standard in determining bulk density.

The sample AL1 represents the Aluminum powder produced by Dalian-China, AL2 is the Aluminum powder produced by Hanwa-Korea, and AL3 is the Aluminum powder produced by LAPAN, Indonesia. The results of the analysis of aluminum powder for each sample are shown in table 3.1. The deviation from the measurement is very small, namely 4.0729E-06 or 0.0001%. The results of the analysis show that AL1 and AL2 have the same bulk density, while AL3 has a lower bulk density. Thus, AL1 and AL2 have higher packing density with interstitial air voids and lower porosity than AL3. If viewed from the aspect of processibility, AL3 with the smallest bulk density will flow more easily than AL1 and AL2. The results of the analysis of AL powder showed that the bulk density of AL1, AL2, and AL3 was 1.2712, 1.2712, and 1.1238 g/cm^3.

Metal particle density analysis can be carried out in several ways, namely by using the Archimedes principle or by calculating the density and bulk density correlation using equation (1). The results of the density analysis of AL1, AL2, and AL3 powders using the Archimedes method and the reduction of bulk density are shown in table 3.1. Pure AL has a density of 2.74 g/cm3 (Holder and Schaak, 2019). AL content purity which is closest to pure AL will increase from AL2>AL3>AL1. Based on the density results, it is estimated that AL2 has a higher purity than AL1 and AL3. The results of calculations using equation (3-1) show a density value that is similar to the calculation using the Archimedes principle.

Table 3.1: Al Density

Sample	ρ (g/cm^3) (Archimedes)	ρ (g/cm^3) (Derived from bulk density)
AL1	2.5163	2.5103
AL2	2.6210	2.6212
AL3	2.4767	2.4735

The validity of the aluminum density measurement method using aquabides as a measurement fluid by considering the value of the hydration reaction rate constant. The rate constant for the hydration reaction between aluminum and water molecules under standard conditions (298 K, 1 bar) is $4.6 \pm 0.8 \times 10^{-13}$ cm^3/s, with a small value for the reaction rate constant, the hydration reaction between aluminum and water under standard conditions it runs very slowly so that the effect of the hydration reaction on density measurements can be neglected. The actual density of the aluminum powder sample can be calculated as the mass of the aluminum sample divided by the volume of the aluminum sample. Statistical analysis of variance and standard deviation of the sample density of aluminum powder and specific gravity of water was carried out as a correction factor (error) from the measurement. Aluminum reactivity in water is expressed as the reaction rate of Al hydrate formation.

3.2. Particle Shape and Size Determination using SEM

The results of the SEM analysis of Al1 and AL2 powders are shown in Figure 3.1. The results of observations on SEM images show that the particle size of AL1 is larger than AL2. Similarly, the shape of the particle AL2 is more spherical than AL1. To prove the results of the analysis, a BET analysis will be carried out. The results of SEM image processing are shown in Tables 4.2 and 4.3. The average particle sizes of AL 1 and AL2 are 34 microns and 11 microns, respectively. The particle size of AL2 is better with AL1 of circularity values of 0.600 and 0.725. The closer to one, the more perfect the roundness will be.

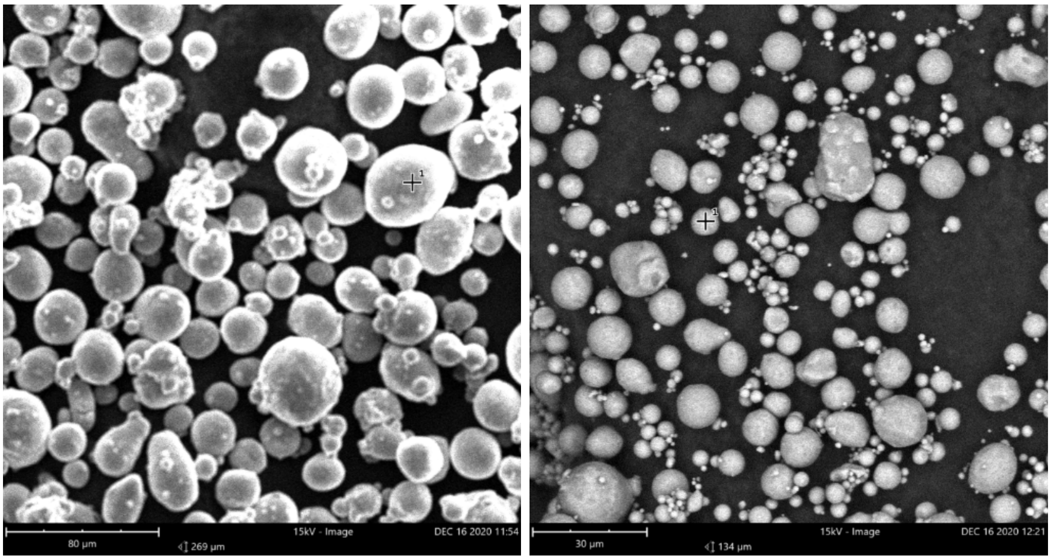


Figure 3.1: SEM Image of Aluminum Powder AL1 (left) and AL2 (right)

Table 3.2: SEM Image Interpretation

Sample	Particle size (micron)	Circularity	Solidity	Roundness	Aspect Ratio
AL 1	33.697	0.600	0.820	0.710	1.792
Al 2	11.135	0.725	0.882	0.728	1.496

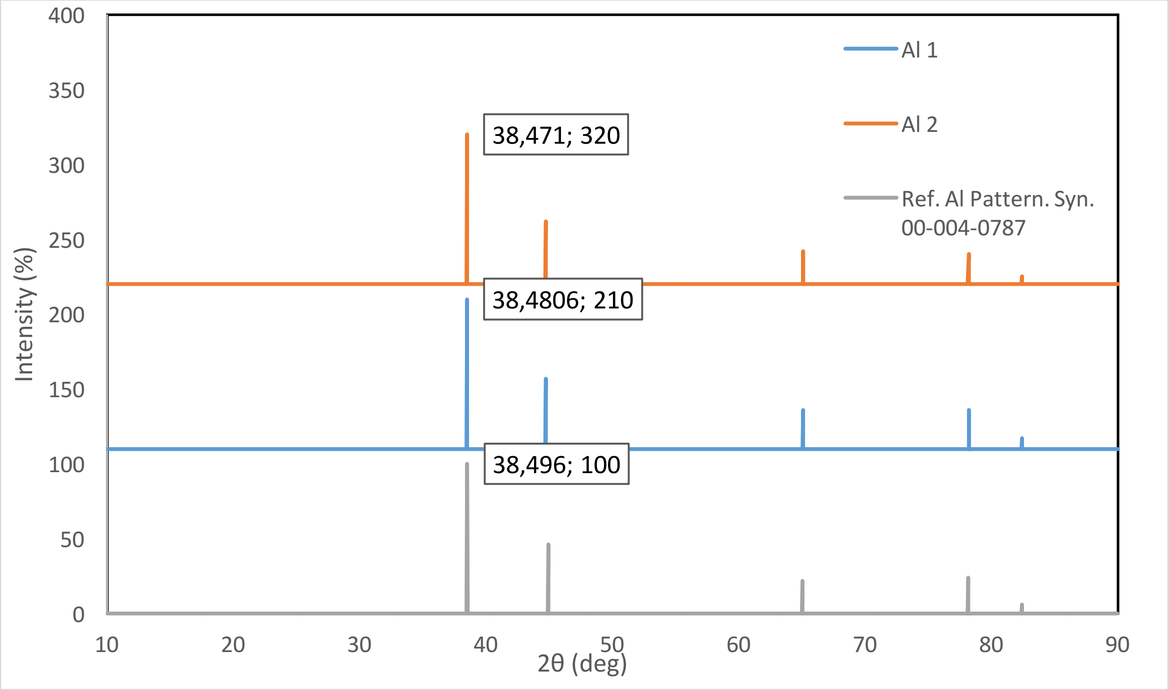
Based on the circularity and roundness values, AL1 and AL2 are classified as well spherical. The roundness of AL2 is better than AL1. AL2 has a wider angle than AL1. AL1 has a more pointed angle. The average particle size is calculated from the results of the particle size distribution. For AL1 used size frequencies from 15-54 microns, while for AL2 used size frequencies from 5-19 microns. The calculation results show that AL1 has an average particle size of 33,697 microns and AL2 has an average particle size of 11,135 microns. Based on the COA of the product, it is stated that AL1 has a size of 37 microns and AL has a size of 10 microns.

Table 3.3: Particle Size Distribution

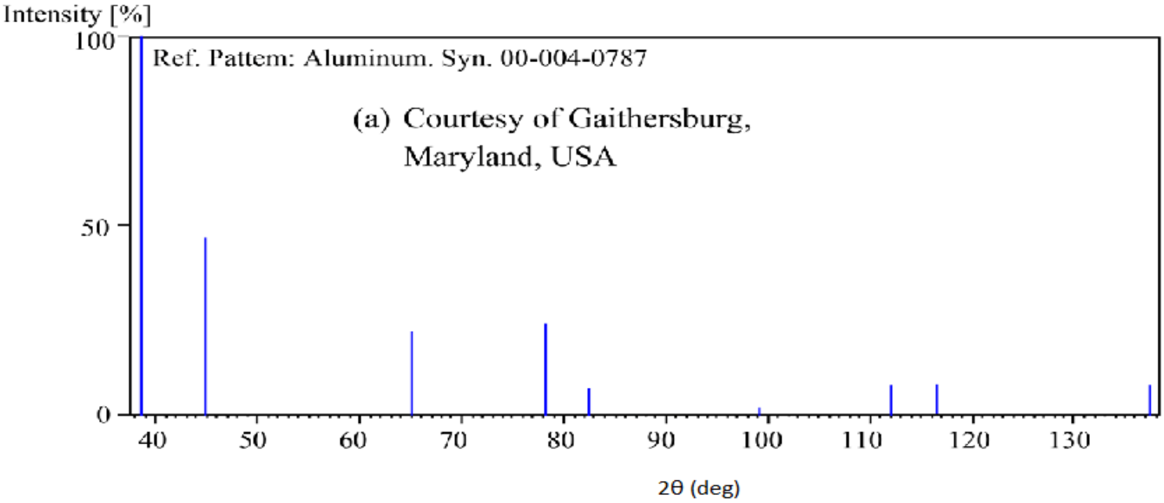
AL1 Size Distribution				AL2 Size Distribution			
Index	Size (μm)	Count	Percentage (%)	Index	Size (μm)	Count	Percentage (%)
0	15.045	2	9.52	0	5.021	18	18.95
1	20.661	5	23.81	1	7.012	19	20.00
2	26.278	5	23.81	2	9.003	16	16.84
3	31.894	3	14.29	3	10.994	8	8.42
4	37.511	0	0.00	4	12.985	11	11.58
5	43.127	3	14.29	5	14.976	12	12.63
6	48.743	0	0.00	6	16.967	5	5.26
7	54.360	3	14.29	7	18.958	6	6.32

3.3. Crystal Structure and Purity of Aluminum

AL purity analysis can be carried out in various ways such as chromatography, spectrometry, gravimetry, volumetric, and X-Ray diffraction (XRD). XRD analysis is interesting because the system works fast, can analyze the crystal structure of AL. The results of XRD analysis for AL1 and AL2 powders in the form of a diffractogram are shown in Figure 3.2. Quantitative analysis was carried out by calculating the intensity at the highest peak from table 3.4. The analysis results are based on the peak of 38.48 degrees for AL2 and 38.47degrees for AL1 in table 3.4 compared with the reference peak AL at a peak of 38.49 degrees.



(a)



(b)

Figure 3.2: AL Powder Stacked Diffractogram of (a). Al1 Al2 and (b). Al reference

Table 3.4: Diffraction Peak of AL1 and AL2

AL1

Peak No	2Theta (deg)	d (A)	I/I1	FWHM (deg)	Intensity (Counts)
1	384.806	233.757	100	0.1216	3687
2	447.252	202.462	47	0.1227	1736
3	650.804	143.207	26	0.1317	944
4	781.974	122.142	26	0.1385	967
5	823.969	116.948	7	0.1415	273

AL2

Peak No	2Theta (deg)	d (A)	I/I1	FWHM (deg)	Intensity (Counts)
1	384.710	233.813	100	0.1280	3941
2	447.138	202.511	42	0.1361	1669
3	650.711	143.226	22	0.1524	856
4	781.897	122.152	20	0.1750	769
5	823.943	116.951	5	0.1670	216

The purity of Al powder from other elements as impurities can be calculated based on the Al powder diffractogram compared to the reference diffractogram. The peaks of each diffractogram can be compared and the overlap coefficient (Szymkiewicz–Simpson coefficient) is calculated for the diffraction angle value data (2θ) of the five Al diffractogram peaks.

Table 3.5: Comparison of Peak Al and interpretation crystal index

peak no.	Ref Al pattern. Syn. 00-004-0787		AL1		AL2		Miller Index
	2Theta (deg)	I/I1	2Theta (deg)	I/I1	2Theta (deg)	I/I1	
1	38,496	100	38,4806	100	38,4710	100	111
2	44,921	46	44,7252	47	44,7138	42	200
3	65,016	22	65,0804	26	65,0711	22	220
4	78,14	24	78,1974	26	78,1897	20	311
5	82,377	6	82,3969	7	82,3943	5	331

From the calculation of the overlap coefficient, the value for AL 1 is 0.998655 and AL 2 is 0.998609. When compared to AL reference Syn. 00-004-0787 with a purity of 99.9999%, it can be stated that the purity of AL 1 is 99.8655% and for AL2 purity is 99.8609%. The purity of both samples (AL1 and AL2) was relatively high with AL1 containing fewer impurities than AL2.

Crystal parameters such as lattice length can be measured using the diffraction selection rule for the first peak. The Miller index values allowed by the diffraction selection rules for fcc crystals are h, k, l must be all even or all odd, where the value 111 is considered eligible to be used. Using the parameter d (interatomic-spacing), the value of the lattice parameter a (lattice length) can be calculated. For fcc type crystals such as Al crystals, the value of the variable a can be calculated as $a = d \cdot \sqrt{3}$. AL1 has a crystal lattice length of 4.04879 Å, while sample AL2 has a slightly larger crystal lattice length of 4.04976 Å. When compared with the size of the Al crystal lattice from the literature reference, which is 4.04950 Å, both crystal samples have a crystal lattice length that is relatively close to the reference crystal lattice length value. This can indicate that the purity of the two Al samples is relatively high, where the presence of impurities will change the crystal structure in the interspace (interstitial impurities) or replace Al atoms in the aluminum crystals (substitutional impurities) where both will change the size of the crystal lattice length and the diffraction peaks of the sample.

3.4. Aluminum Surface Area

The graph of the BET adsorption isotherm model obtained can be analyzed using the Multi-Point analysis method. The characterization of the sample using the BET isotherm includes the surface area and constant C as the sample's affinity for the adsorbate in the gas-solid adsorption process. The obtained adsorption isotherm graph was also analyzed by the BJH method.

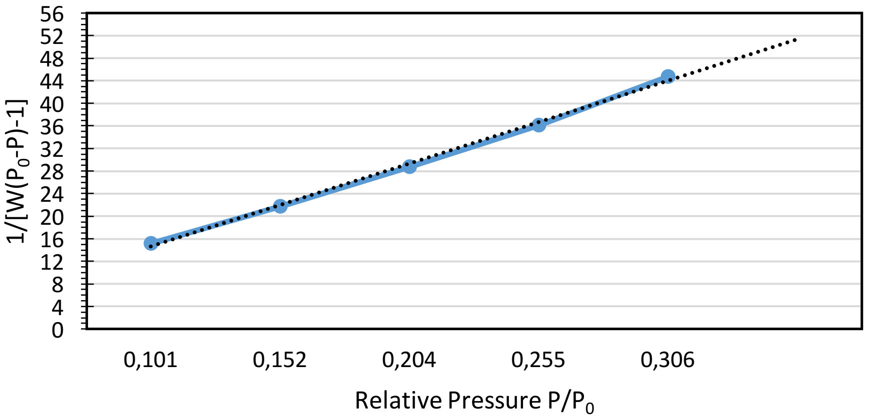


Figure 3.3: BET Isotherm of AL1 Sample

Characterization of samples with the BJH isotherm included pore surface area, pore volume, and pore size in the adsorption and desorption processes. The results of the surface characterization of samples with BET theory can be compared with the BJH method. Surface area and porosity analysis can be done in several ways. An analysis method that is proven to be relatively easy and quick is the BET analysis.

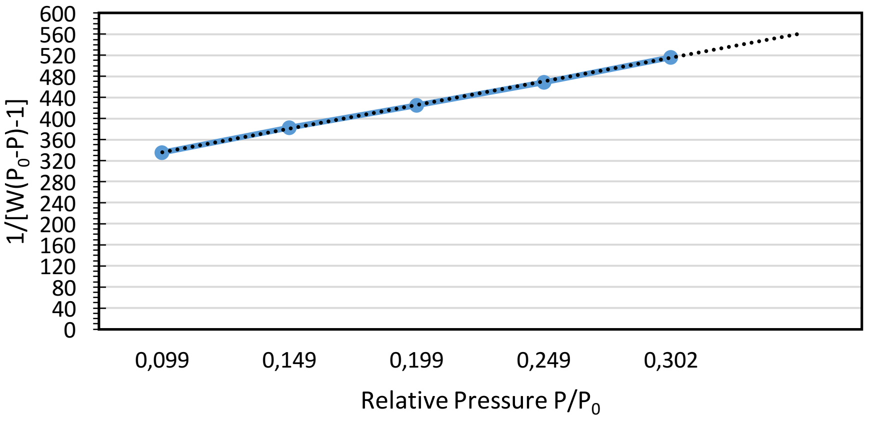


Figure 3.4: BET Isotherm of AL2 Sample

The results of BET image interpretation with the help of image processing, obtained surface area, pore-volume, average pore radius of AL particles as shown in table 3.6. The average surface area of AL1 (23.977 m2/g) is higher than AL2 (3.150 m2/g). Although the pore size of AL2 particles is larger than AL1, the number of pores of AL1 is 600 times more than that of AL2 particles. Thus, the pore volume of AL1 particles is more than that of AL2 particles. The large surface area of the particles will have an impact on the viscoelasticity of the propellant mixture when casting and the rate of combustion.

Table 3.6: Adsorption Data of BET Isotherm

Sample	BET Adsorption			
	Surface Area (m^2/g)	Pore Volume (cm^3/g)	Pore Radius (nm)	C Constant
AL1	23,977	0,009	0,157	1546,754
Al2	3,150	0,004	0,261	4,480

The presence of pores in the Al powder sample which has been proven on SEM images and BET adsorption isotherm indicates the possibility of over-estimate in the measurement of surface properties. The BJH model using desorption isotherm can be used as a comparison.

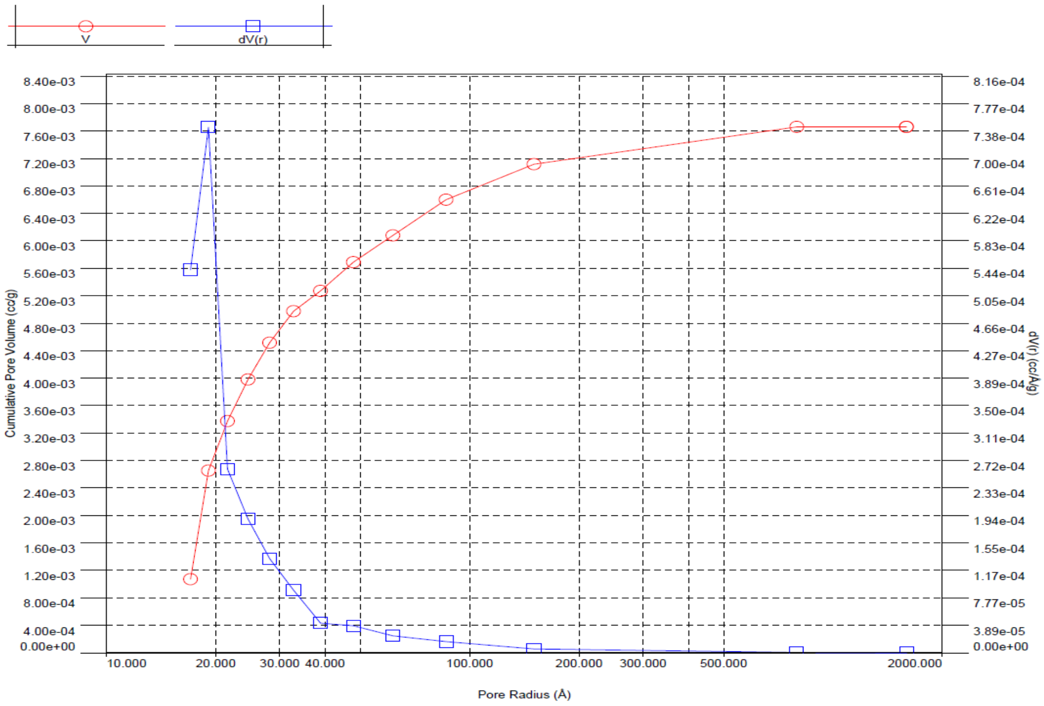


Figure 3.5: Desorption Isotherm BJH of AL1 Sample

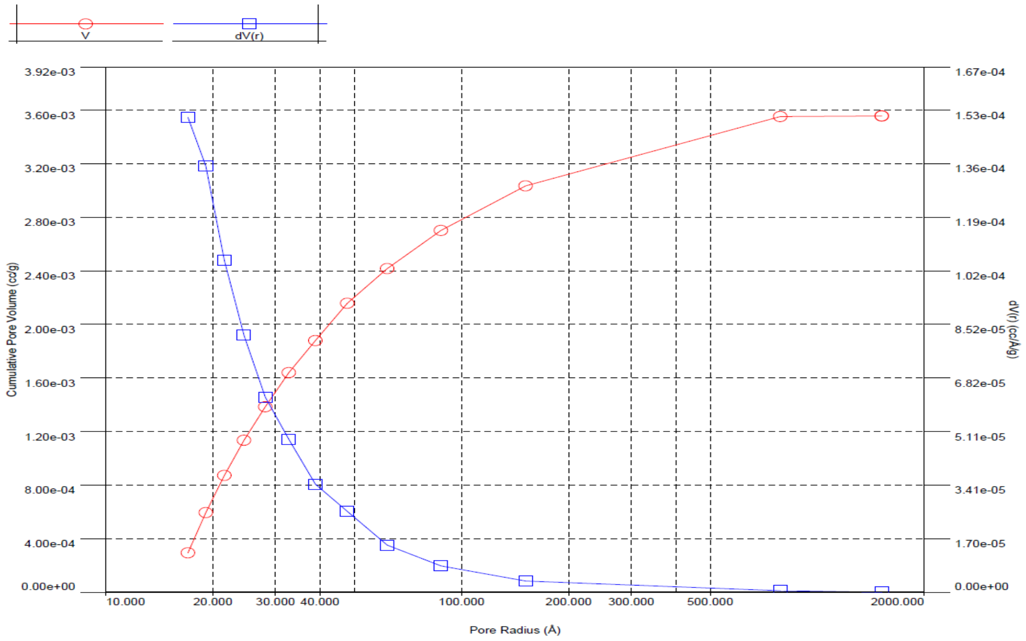


Figure 3.6: Desorption Isotherm BJH of AL2 Sample

Table 3.7: Desorption Data of Isotherm using BJH Methods

	BJH Desorption			
Sample	Surface Area (m^2/g)	Pore Volume (cm^3/g)	Pore Radius (A°)	Average Pore Radius (A°)
Al1	5,398	0,008	19,088	1,57439
Al2	1,912	0,004	17,022	2,60930

From the isotherm desorption data using the BJH method, it is known that the surface area of the sample Al1 (5.398 m2/g) is larger than Al2 (1.912 m2/g) with a maximum pore size of the sample Al1 (1.9088 nm) slightly larger than Al2 (1, 7022 nm), other than that the pore volume size and mean pore size is similar to the results shown by the BET isotherm. The striking difference is shown in the results obtained in the surface area. The surface area of the sample AL1 with the BET isotherm is 23.977 m2/g, where BJH recorded a surface area of 5.398 m2/g, while for the surface area of the Al2 sample, BET recorded a yield of 3.150 while BJH was 1.912 m2/g. The BJH method can measure the surface area of porous materials without neglecting lateral adsorption and does not consider all adsorption sites equally because of potential differences, where the pore surface adsorption site is different from the surface site outside the pore. BJH will produce more accurate calculation results for porous materials and additional parameters of maximum pore size that cannot be measured with the BET isotherm.

3.5. Characteristics of composite propellants with various Aluminum qualities

The propellant characteristic test was carried out to test the difference in the quality of the propellant produced if the aluminum quality was different. The analysis of propellant characteristics includes combustion energy, combustion rate, mechanical strength, and hardness. The results of the analysis are shown in table 3.8.

Table 3.8: Propellant Characteristics

Characteristics	Propellant - AL1	Propellant -AL2
Heat of Combustion, Kcal/g	187	194
Burning rate, cm/sec	0.45	0.53
Tensile strength, kg/cm2	5.6	5.4
Elongation, %	67	71
Hardness, shore A	87	80

Specific gravity, g/cm3	1.72	1.75

Based on the performance of the propellant, it can be seen that the calorific value of the propellant was increased by using AL2 compared to AL1. The calorific value of propellant combustion is influenced by the density of AL, the greater the density of AL, the more AL content in a propellant will be. The more Al will increase the heat of combustion so that the calorific value of combustion increases (Kanagaraj, Chakravarthy and Sarathi, 2017). AL1 has a lower density than AL so that the resulting propellant with the same composition will have a higher density, which is 1.75 g/cm3 compared to 1.72 g/cm3.

The results of the propellant burn rate test showed that the propellant fuel rate with AL1 gave a slower combustion rate than the propellant with AL2. AL1 has a larger particle size than AL2. AL1 has a particle size of 33,697 microns, while AL2 has a particle size of 11,135 microns. According to the Arrhenius combustion reaction theory (Gligorijević et al., 2014), the smaller the particle size, the larger the surface area, so the frequency of collisions between particles will be greater. This result is also supported by the particle shape of AL. AL1 has a more pointed particle shape and a smaller surface area than AL2, with an Al2 surface area of 600 times the surface area of AL1. The larger the surface area, the greater the frequency of collisions between particles, thus strengthening the surface area together with the smaller particle size.

The results of the mechanical properties test of the propellant showed that the propellant with Al1 had greater hardness than the propellant with AL2. AL1 has a more pointed surface shape than AL2 so that the arrangement of the Aluminum particles becomes stronger and intertwined. Thus, the resulting propellant becomes harder.

The results of the working life of the propellant give a change in the viscosity of the propellant slurry when printed. Working life aims to ensure that the rheological properties of the propellant slurry are still possible to print. For the propellant molding process using a pressure casting system, it is expected that the propellant slurry has sufficient viscosity (16,000 -20,000 Poise) so that the propellant printing operation can still be carried out. The viscosity of the slurry propellant with AL1 shows that it thickens faster, as indicated by the viscosity of the slurry at the 90th minute which is 15,445 Poises compared to the propellant with AL2 which has a slurry viscosity of 13,057 Poises. The rheological properties of the propellant slurry are highly dependent on the nature of the binder used (NourEldin et al., 2020), however, the shape and size of the solid particles also have an effect. More amorphous particles, sharper angles will make the particle arrangement irregular and stack each other, so that the composite structure becomes hard, but the hardness is uneven. This has an impact on the mechanical properties which have lower tensile strength.

Based on the results of the evaluation of the characteristics of the propellant, the characteristics of the propellant are important to determine the characteristics of Aluminum. Some parameters of propellant quality that need to be known are density (bulk and true density), particle shape and size, AL purity, surface area, and particle pore. The density and purity of AL affect the performance of the propellant. The shape, size, and surface area affect the mechanical properties, combustion rate, and viscoelastic properties of the propellant.

4. Conclusions

The results of the analysis of the quality of aluminum for composite propellant raw materials have a very large influence on the quality of the resulting propellant, so an integrated, fast, and efficient quality analysis is needed. The critical quality parameters of Aluminum as propellant fuel are density, purity, particle shape and size, and porosity/surface area. AL2 which has the best value for the quality parameters compared to AL1 and AL3 can be used to produce a higher quality composite propellant that has stronger mechanical parameters, higher heat of combustion, and better ballistic performance. Processability was also improved by using better quality Aluminum powder to produce the better rheological characteristics, which AL2 propellant had lower slurry viscosity of 13,057 Poises compared to AL1 propellant with the slurry viscosity of 15,445 poises.

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In writing this paper, the main contributors are H. R. Sitompul and H. B. Wibowo, while the other authors are member contributors. H. R. Sitompul contributed to conducting laboratory analysis, calculations, and data interpretation. H. B. Wibowo contributed to research design, discussion, propellant testing, and coordinating research. M. Baiquni contributed to the SEM analysis. K. Hartaya contributed to the proofreading. L. H. Abdillah and R. Ardianingsih contributed to the experiment of making propellant. A. Restasari contributes to measuring viscoelasticity. R. S. Budi contributed to X-Ray testing. L.H. Abdillah, R. Ardianingsih, and A. Restasari supervised the laboratory works and the writing of this paper.

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