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Improving aluminum particle reactivity by annealing and quenching treatments: Synchrotron X-ray diffraction analysis of strain

Authors

McCollum, Jena
Pantoya, Michelle L
Tamura, Nobumichi

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Corresponding Author: Prof. Michelle Pantoya, PhD

Corresponding Author's Institution: Texas Tech University

First Author: Jena M McCollum

Order of Authors: Jena M McCollum; Michelle Pantoya, PhD; Nobumichi Tamura, PhD

Abstract: In bulk material processing, annealing and quenching metals such as aluminum (Al) can relieve residual stress and improve mechanical properties. On a single particle level, affecting mechanical properties may also affect Al particle reactivity. This study examines the effect of annealing and quenching on the strain of Al particles and the corresponding reactivity of aluminum and copper oxide (CuO) composites. Micron-sized Al particles were annealed and quenched according to treatments designed to affect Al mechanical properties. Synchrotron X-ray diffraction (XRD) analysis of the particles reveals the thermal treatment increased the dilatational strain of the aluminum-core, alumina-shell particles. Flame propagation experiments also show thermal treatments effect reactivity when combined with CuO. An effective annealing/quenching treatment for increasing aluminum reactivity was identified. These results show that altering the mechanical properties of Al particles affects their reactivity.

Suggested Reviewers: Emily Hunt PhD
Professor, Engineering, West Texas A&M University
ehunt@wtamu.edu
Expertise in aluminum combustion

Keerti Kappagantula PhD
Professor, Ohio University
kappagan@ohio.edu
Expertise in energetic materials

Eric Vogel PhD
Professor, Materials Science & Engineering, Georgia Tech
eric.vogel@mse.gtech.edu
Expertise in metals

Zuhir Munir
Professor, Materials Science & Engineering, UC Davis
zamunir@ucdavis.edu

Expertise in metals and combustion

TEXAS TECH UNIVERSITY
Department *of* Mechanical Engineering

June 16, 2015

Re: Manuscript for publication consideration in *Acta Materialia*

Dear Dr. Mahajan,

Please find our manuscript entitled “Improving Aluminum Particle Reactivity by Annealing and Quenching Treatments: Synchrotron X-ray Diffraction Analysis of Strain,” submitted for publication consideration in *Acta Materialia*. We have not submitted this article elsewhere at any time. Support for this project was provided by a grant from the Army Research Office.

This manuscript focuses on synchrotron x-ray diffraction analysis of aluminum particles that have been treated to improve their mechanical properties. The aluminum is then examined for its reactivity with a metal oxide to show the relationship between material processing and the mechanical properties of the metal then extend this understanding towards application for energy generation technologies. We felt this combination of in-depth analysis of the material property of strain based on annealing and quenching processing of a metal powder and its application to reactivity were the perfect combination of mechanical and functional behavior appropriate for *Acta Materialia*.

Thank you for considering our manuscript.

Sincerely,

Prof. Michelle Pantoya, Ph.D.
J. W. Wright Regents Endowed Chair Professor
2703 7th Street
Mechanical Engineering Department
Texas Tech University
Lubbock, TX 79409-1021
Phone: 806-834-3733
Mobile: 806-438-8671
Email: michelle.pantoya@ttu.edu

Improving Aluminum Particle Reactivity by Annealing and Quenching Treatments: Synchrotron X-ray Diffraction Analysis of Strain

Jena McCollum and Michelle Pantoya

Mechanical Engineering, Texas Tech University, Lubbock, TX 79409

Nobumichi Tamura

Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

Abstract

In bulk material processing, annealing and quenching metals such as aluminum (Al) can relieve residual stress and improve mechanical properties. On a single particle level, affecting mechanical properties may also affect Al particle reactivity. This study examines the effect of annealing and quenching on the strain of Al particles and the corresponding reactivity of aluminum and copper oxide (CuO) composites. Micron-sized Al particles were annealed and quenched according to treatments designed to affect Al mechanical properties. Synchrotron X-ray diffraction (XRD) analysis of the particles reveals the thermal treatment increased the dilatational strain of the aluminum-core, alumina-shell particles. Flame propagation experiments also show thermal treatments effect reactivity when combined with CuO. An effective annealing/quenching treatment for increasing aluminum reactivity was identified. These results show that altering the mechanical properties of Al particles affects their reactivity.

Key Words

Aluminum, dilatational strain, mechanical properties, reactivity, annealing, quenching, synchrotron XRD, energetic materials

Introduction

Composite energetic materials consist of a metallic fuel (i.e., aluminum (Al)) and a metal oxide (i.e., copper oxide (CuO)). These composites are widely studied due to their high energy densities and heats of combustion. A main goal of much published research studying aluminum combustion is to understand and improve particle reactivity. Towards this end, many have proposed new Al synthesis strategies: such as altering the native aluminum oxide (Al_2O_3) coating with another passivating agent such as alkenes[1] or applying self-assembled monolayers (SAM) to the particle surface[2]. Various explanations for Al oxidation mechanisms have also been proposed, each strongly tied to the ignition mechanism and heating rate[3]–[11]. All theories share a common theme for mass transport of fuel and oxidizer, but differ in how that diffusion is achieved (i.e., via (a) dispersion[3], [4], (b) phase changes in the polymorphous passivation shell[5]–[7] , (c) reactive sintering[8], (d) pressure gradient driven processes[9], [10], and (e) induced electric field influences[11]). This article will not directly deal with any particular reaction mechanism, but rather investigate the influence of a new parameter, mechanical strain, which has only recently been considered in the study of Al oxidation[3], [4], [12], [13].

A typical aluminum particle consists of an aluminum core passivated by an alumina shell as seen in Fig. 1. Residual stress within an aluminum particle is induced during production. These are thermally induced stresses that are generated during the cooling stage. Stress builds in two stages during particle synthesis: (1) within the aluminum core during cooling and prior to oxide shell formation; and, (2) during alumina shell formation until the core-shell system reaches ambient temperature conditions.

Within the molten aluminum droplet absent of a shell, stress develops as the molten aluminum droplet cools, because external surface layers start to shrink while the core is still hot

and free to contract but its contraction is constrained by the external layers that have solidified and become rigid. In this way, the stress distribution in a naked aluminum particle is tensile in the core and compressive at the surface. Smaller particles cool at faster rates[14]–[16] and should manifest larger internal stresses during production.

The shell is formed when oxygen is introduced at temperatures below the melting temperature of aluminum (i.e., < 660 °C[17]) and no higher than 440 °C (i.e., the phase transition of amorphous to gamma alumina[18]). At this point, another residual stress develops because there exists a mismatch of thermal expansion coefficients (i.e., *a*, linear coefficient; or, *b*, volumetric coefficient) between the newly formed alumina shell (i.e., a_{ox} =5 x 10^{-6} K^{-1} and b_{ox}= 8 x 10^{-6} K^{-1}[18]) and aluminum core (a_m = 23 x 10^{-6} K^{-1} and b_m = 69 x 10^{-6} K^{-1} [17]). Cooling from 440 °C to ambient will induce shrinkage in the core while the shell remains relatively rigid, i.e., there is an order of magnitude difference in expansion coefficients. The internal radius of the oxide sphere is forced to decrease and stresses arise at the interface of the core-shell.

Isothermal stress relief is a common approach to stress relaxation[19]–[21]. This technique involves the uniform heating of a material to a specified temperature below the melting temperature, holding at that temperature for a period of time, followed by cooling to control the re-introduction of desirable thermal stresses. This sequence of steps is referred to as annealing followed by quenching. Timoshenko and others[19]–[23] explain significant microstructural changes take place that promote stress relief at about two-thirds the temperature at which the stresses were formed. In the case of Al particles, the amorphous alumina shell is formed at roughly 440 °C such that annealing temperatures should at least be 293 °C. Interestingly, Firmansyah et al. examined the stress state of nano-aluminum powder upon

continuous heating using a high temperature XRD and found that Al particles experience a zero-stress state at 300 °C[24].

The main mechanism that causes relaxation of locked-in stresses for annealing temperatures < 400°C is classical diffusional creep. This mechanism enables counterbalancing regions of tensile and compressive stresses to contract or expand slightly, and thus to redistribute. This is also a time dependent process and determining the optimum temperature and duration for annealing and quenching has not previously been investigated for aluminum particles. Some experimental work on other metals has revealed a relevant conclusion: while the annealing time is important, creep is a logarithmic process such that most relief is obtained at a given temperature rapidly[20]. Data presented by Adeyemi et al. for carbon steel suggests annealing times on the order of 10 minutes should be sufficient to relieve stress[25]. Even for the annealing temperature of 500^{o}C, 80% of the residual stress is relieved in 5 minutes. Bulk aluminum alloys have been studied for creep behavior [26]. Prasad et al. studied creep at 87 and 200 °C and found the effects of stress increments and decrements to be different[26]. This is an important finding because it implies stress variations are significant in the temperature ranges we are most interested (i.e., < 400 °C).

Quenching is another important variable to consider. Evancho et al. showed that slow cooling may re-introduce thermal stresses that are undesirable (e.g., like the residual stress) and lead to lower strength[27]. Faster cooling rates may effectively 'freeze' the stress state such that purposefully induced desirable thermal stress do not have time to relax, leading to higher strength. The hypothesis is: faster cooling rates will produce aluminum particles with optimal mechanical properties promoting optimal reactivity.

Dikici et al. showed preliminary evidence that annealing and quenching Al particles affects reactivity [13]. They heated Al particles to a prescribed temperature then cooled them to room temperature and found that some thermal treatments lead to improved flame speeds. Their powders were allowed to cool at two different rates. The samples that were cooled at 0.06 KPS (Kelvin per second) showed flame speeds comparable to the untreated samples, but when the cooling rate was raised to 0.13 KPS, the flame speeds for micron scale Al and molybdenum trioxide (MoO_3) improved for one case and worsened for the other. More recently, Levitas et al. extended the work by Dikici et al. by focusing on micron scale Al particles and using a different metal oxide (i.e., copper oxide (CuO))[12]. They examined flame speeds and showed certain thermal treatments were consistent with predictions of the melt dispersion mechanism, effectively expanding the realm of this reaction mechanism towards optimization of micron scale Al particles [12].

The objective of this study is to fundamentally quantify changes in strain associated with Al particle thermal treatments and examine energy propagation behavior of treated Al particles combined with CuO. These extensions enable an understanding of how thermal treatment and mechanical properties couple to influence the reactivity of Al particles with a solid oxidizer. Improving Al reactivity with a relatively simplistic heat treatment approach may greatly improve their functional application.

Experimental

Sample Preparation

Aluminum particles were procured from Alfa Aesar (Ward Hill, MA). Their average diameter was determined using an AccuSizer 780 optical particle size analyzer (Santa Barbara,

CA). The Al particles were suspended in filtered water (specifically, 2 mg Al/80 ml water) and sonicated for 60 minutes in cycles of 10 seconds on, 10 seconds off in order to avoid heating. The sample was then pulled via a syringe pump for analysis. Particle length (diameter) is calculated and plotted as a count-based measurement. The plot for the untreated Al is shown in Fig. 2a and the size data is given in Fig. 2e. The oxide thickness used for purity calculations was estimated at 3 nm (i.e., >1% oxide concentration, see Fig. 1). The untreated Al particles have an average diameter of 5 microns.

Aluminum particles (200 mg) were loaded into ceramic trays and subjected to various annealing temperatures and quenched to room temperature. This process used a Neytech Qex vacuum oven (Torrance, CA). This is a programmable oven, such that the powder was annealed to 100, 200 or 300°C at a controlled rate of 10 Kelvin per minute (KPM), held at the prescribed temperature for 15 minutes then removed from the oven and quenched via refrigeration to room temperature in air at standard pressure. Powder temperature was monitored using an InstruNet Direct to Sensor system (Charlestown, MA) and Type K thermocouples from Omega Engineering (Stamford, CT). Aluminum temperature was monitored for each thermal cycle and Fig. 3 shows the temperature response as a function of time.

For the natural convection conditions that exist in these experiments, materials cool according to a lump capacitance model. The quenching rate is an exponential function of time as shown in Eq. (1). The temperature evolution reduces exponentially and experimental results are approximated by Eq. (1).

$$T = T_a + (T_0 - T_a) \exp(-At) \text{ with } A = 0.0078 \text{ s}^{-1} \quad (1)$$

In Eq. (1) T_a is ambient temperature, T_0 is annealing temperature, t is time and A is determined by examining the exponential plots of the cooling curves and identifying the

coefficient. An average quenching rate ranges from 0.13 to 0.38 KPS depending on annealing temperature.

The Al particles were then mixed with 50 nm average particle diameter spherical CuO particles (Sigma Aldrich (St. Louis, MO)) to an equivalence ratio of 1.2 (i.e., slightly fuel rich). The mixing process is well documented[28]–[30] but will be summarized here. The dry powders were weighed and suspended in hexane and mixed using a Misonix Sonicator 3000 probe for 2 minutes. The solution is poured into a Pyrex dish and hexane evaporated in a fume hood for 24 hours. The dry powders were retrieved and sieved to break up large agglomerations.

The powder is carefully loaded into 3 mm inner diameter, 8 mm outer diameter, 10 cm long quartz tubes containing 850 mg of powder each. The theoretical maximum density (TMD) of the loose powder is determined by a weighted average of the bulk densities of Al (2.7 g/cm^3), Al_2O_3 (3.95 g/cm^3), and CuO (6.31 g/cm^3). This is calculated using Eq. (2) where M is the percent mass of reactant i and ρ is the density for each reactant.

$$TMD = \frac{1}{\sum_i M_i \frac{1}{\rho_i}} \qquad (2)$$

The calculated TMD is 5.46 g/cm^3 and the bulk density is 1.04 g/cm^3 such that tubes were loaded to 19% TMD.

Strain Measurements

X-Ray Diffraction (XRD) experiments were performed at the Advanced Light Source on beamline 12.3.2 using a micron focused synchrotron x-ray beam. This is a unique facility that allows measurement of micron-scale samples. Aluminum powder samples were spread over glass slides and scanned under the x-ray beam (either polychromatic or monochromatic) while a diffraction pattern was collected at each step using a DECTRIS Pilatus 1 M detector. The

measured relative small shifts in the reflection positions in the Laue pattern provides the deviatoric strain tensor of the material while the measurement of the energy of one reflection provides the dilatational component. Data were processed using XMAS software[31]. The beamline experimental setup and capabilities have been described elsewhere[32].

Flame Speed Measurements

Figure 4 illustrates a typical powder filled tube arrangement for measuring flame speed as well as representative still frame images of flame propagation. The apparatus and procedure are described in more detail elsewhere[13] but summarized here. Both ends of the tube were sealed with one side securing a length of nickel-chromium wire for ignition. Five experiments per annealing temperature were performed to establish repeatability. Each tube was placed inside a blast chamber for ignition and flame propagation experiments. The powders were ignited and flame propagation was observed through a viewing window in the chamber (as seen in Fig. 5). The reaction was recorded with a Phantom v7 (Vision Research, Wayne, NJ) high speed camera at a rate of 29,000 frames per second and 512 x 128 resolution. The camera was aligned perpendicular to the direction of flame propagation. Flame speed was determined by tracking the flame front through a referenced time and distance using the Vision Research Software. The resolution of the flame speed for this diagnostic is 0.1 m/s. The largest source of uncertainty in the measurement is due to repeatability and is shown for each data set in the results.

Results

Further particle size analyses were performed for each Al sample after thermal treatment. Figures 2b-d show the plots for Al particle sizing and the average particle diameter and

distribution is given in Fig. 2e. This data shows that the thermal treatments did not result in sintering or agglomerations because the average diameter remains consistent (about 5.9 μm).

Figure 6 shows results of dilatational strain (i.e. change in volume) distribution measurements of aluminum particles that were (a) untreated and annealed to (b) 100^{o}C, (c) 200^{o}C and (d) 300°C. Table 1 presents the count based averages for dilatational strain for each annealing temperature. The average strain for the samples annealed to 100 and 200 ^{o}C fall within the resolution of the machine ($2x10^{-5}$), so their percent increase was negligible. However, the aluminum particles annealed to 300°C and cooled to room temperature showed a significant dilatational strain increase from the baseline Al particles (i.e. 660%). It is noted that these measurements are of the aluminum core because the alumina shell is too thin (i.e., less than 5 nm) to resolve volume based strain measurements using this approach.

Figure 7 shows flame speed as a function of annealing temperature. The samples annealed to 100°C experience a 2% increase in flame speed from the baseline; and, when annealed to 200°C flame speed increases by 3%. Once the samples are annealed to 300°C, they experience a 23% increase in flame speed. This is a significant increase in reactivity for the composite annealed to 300°C but not for the other annealing temperatures. These results are also summarized in Table 1.

Discussion

Figure 6 shows that compared to the baseline, volumetric strain increases as annealing temperature increases. According to Fig. 6 and Table 1, average dilatational strain increases by 660% after thermal treatment to 300 ^{o}C but is unchanged in the samples annealed to 100 and 200 ^{o}C. Diffusional creep resistance occurs in samples when the annealing temperature is below a

critical fraction of the melt temperature (i.e., T_m= 660 ^oC for Al)[33]. Kitagawa et al. investigated creep response in bulk Al samples subjected to a variety of temperatures (i.e., from $0.32T_m$ (~27 ^oC) to $0.55T_m$ (~240 ^oC)) for different number of thermal treatment cycles. They were interested to see the average creep rate for each temperature over a variety of thermal cycles and found that the transition temperature for Al to exhibit creep acceleration is $0.4T_m$ (i.e., 100 ^oC) and the maximum acceleration of creep for Al occurs at about $0.52T_m$ (212 ^oC). Figure 4 shows that the samples annealed to 100 and 200^oC show little response in terms of dilatational strain, consistent with annealing temperatures below the critical temperature for maximum acceleration of creep (i.e., 212 ^oC). However, once this critical temperature is passed (i.e., 300 ^oC), the Al particles responded with a 660% increase in dilatational strain. The effect of this trend is seen in the flame speed results. The Al samples annealed to 100 and 200 ^oC showed little change from the untreated case, but for the 300 ^oC case, flame speeds increased by 24%.

Ultimately, the annealing and quenching treatment effectively increased the overall strain. It is important to note that at higher annealing temperature or longer annealing times, particles could experience shell growth and/or oxide phase change. Amorphous alumina begins to transition to gamma-phase alumina around 440 ^oC, such that annealing beyond this temperature would affect the shell microstructure, which may also impact reactivity. Also, annealing to temperatures past 440 ^oC for an extended time (60+ minutes) would result in oxide layer growth as shown by Gesner et al.[34]. They annealed 95 nm diameter Al particles to 480 ^oC for a variety of hold times and saw an increase in oxide shell thickness from 2.7 to 8.3 nm when held for 150 minutes but negligible shell growth for times applied in this study (and in this study annealing temperatures did not exceed 300 °C).

The thermal treatment is purposefully designed to relax residual stresses by annealing then reintroduce desirable stresses by quenching. While thermal treatments are relatively established for bulk metal processing, manipulation of mechanical properties for particles that consist of a core-shell structure are not well understood. In this study, results for thermal treatment of particles appear to be consistent with theory for bulk metals. The benefit of thermal treatment from Figs. 6 and 7 is that the 300 °C annealed and quenched Al powders now exhibit higher strain that also correlates with higher macroscopic energy propagation.

Conclusion

This study examines the effect of aluminum powder annealing and quenching treatments on strain in aluminum particles and corresponding reactivity. Micron-sized aluminum powder was annealed and quenched according to treatments designed to affect mechanical properties. Synchrotron X-ray diffraction (XRD) analysis of the particles reveals annealing increased the dilatational strain of the Al particles. Diffusional creep is the primary mechanism affecting strain and for powder annealed to 300 ^{o}C, 660 % increase in dilatational strain was observed. However, annealing to 100 or 200 ^{o}C showed no significant increase in dilatational strain. Treated Al powder was then mixed with CuO to assess reactivity. Flame speed measurements similarly showed the 300°C annealed sample produced the highest flame speed. These results reveal that altering the mechanical properties of aluminum particles affects their reactivity, particularly when combined with a solid oxidizer.

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Jena McCollum and Michelle Pantoya

Mechanical Engineering, Texas Tech University, Lubbock, TX 79409

Nobumichi Tamura

Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

Figures

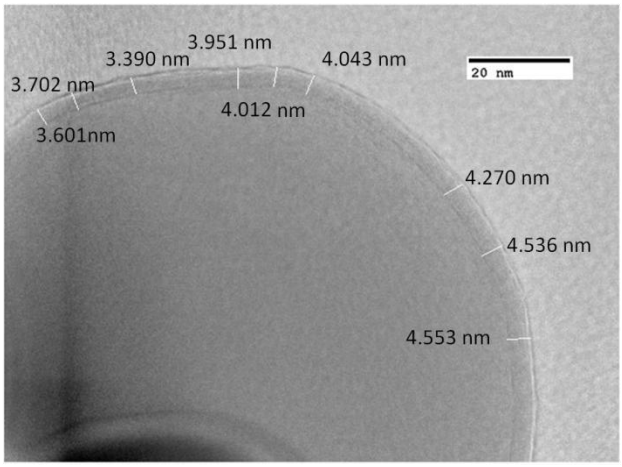
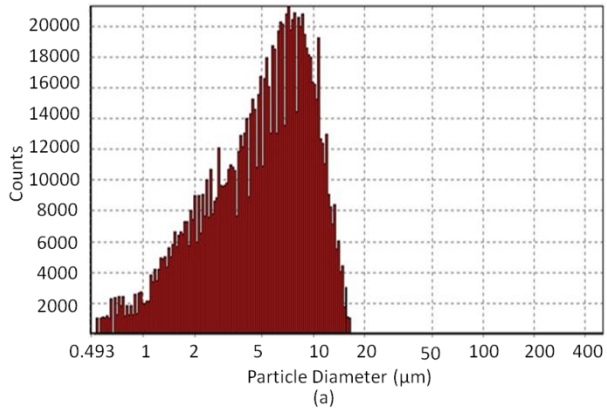


Figure 1. Transmission electron spectroscopy (TEM) image of a nano-scale Al particle to show alumina shell thickness. The average thickness is 4.01 nm. This thickness was used in the purity calculations for micron-scale Al particles (< 1% by mass oxide).


Counts
20000
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8000
6000
4000
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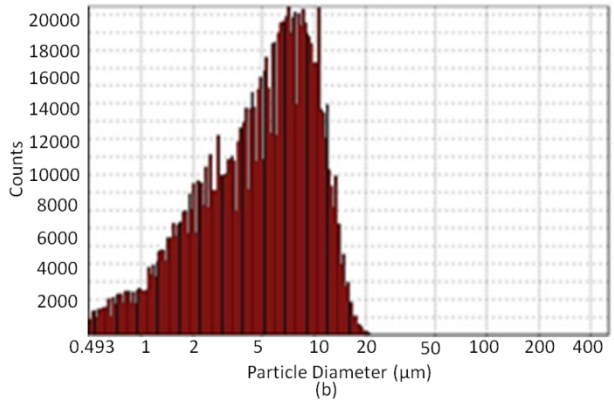
0.493 1 2 5 10 20 50 100 200 400
Particle Diameter (µm)
(a)


Counts
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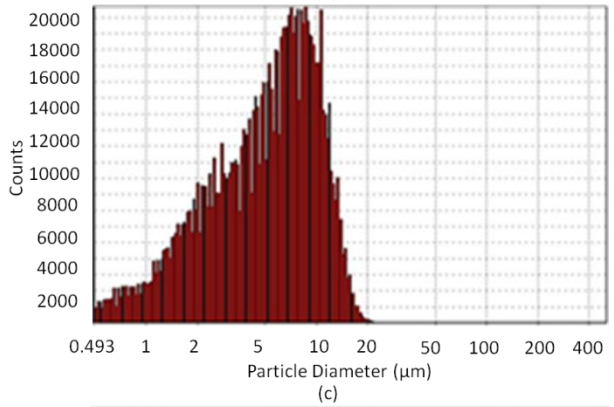
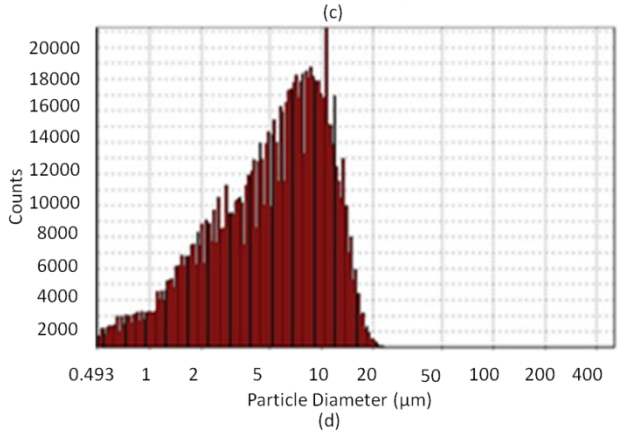
Particle Diameter (µm)
(b)


Counts
20000
18000
16000
14000
12000
10000
8000
6000
4000
2000
0.493 1 2 5 10 20 50 100 200 400
Particle Diameter (µm)
(c)


Counts
20000
18000
16000
14000
12000
10000
8000
6000
4000
2000
0.493 1 2 5 10 20 50 100 200 400
Particle Diameter (µm)
(d)

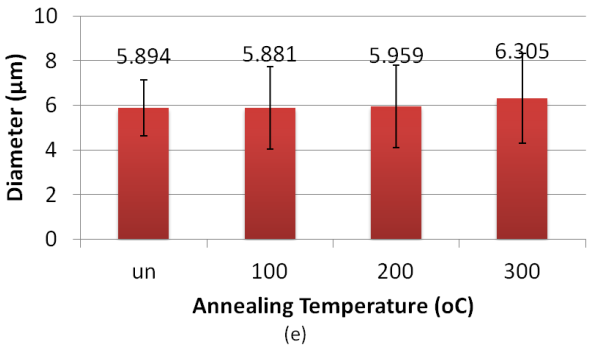


Figure 2. Particle size distribution of (a) untreated Al and Al annealed to (b) 100 ^{o}C, (c) 200 ^{o}C, (d) 300 ^{o}C and (e) average particle diameter with standard deviation for each Al powder sample.

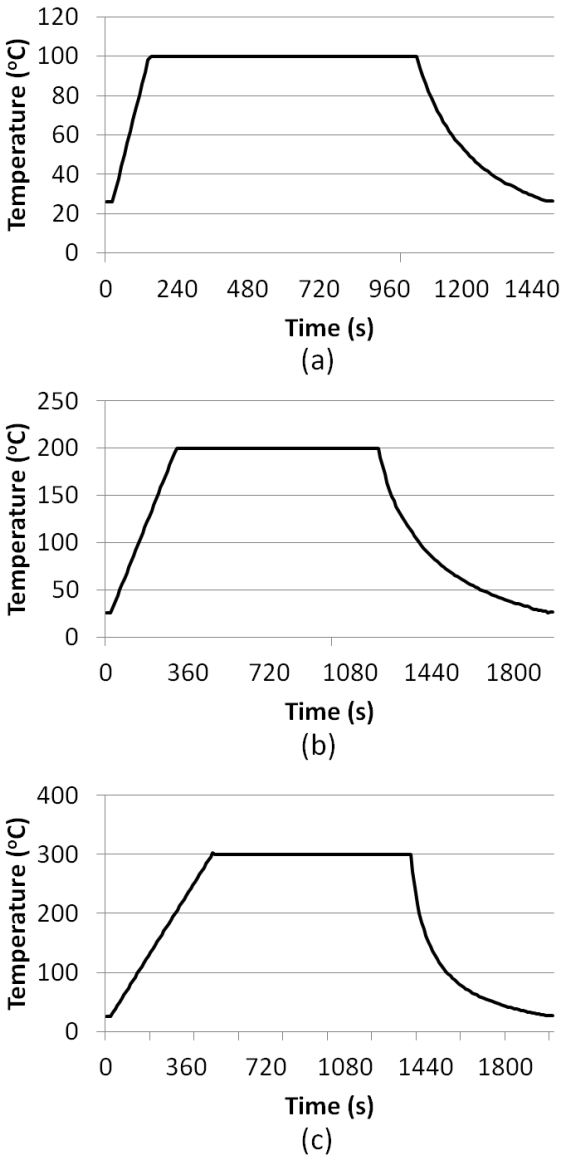


Figure 3. Temperature plots for Al thermal cycles to (a) 100 ^{o}C, (b) 200 ^{o}C and (c) 300 ^{o}C.

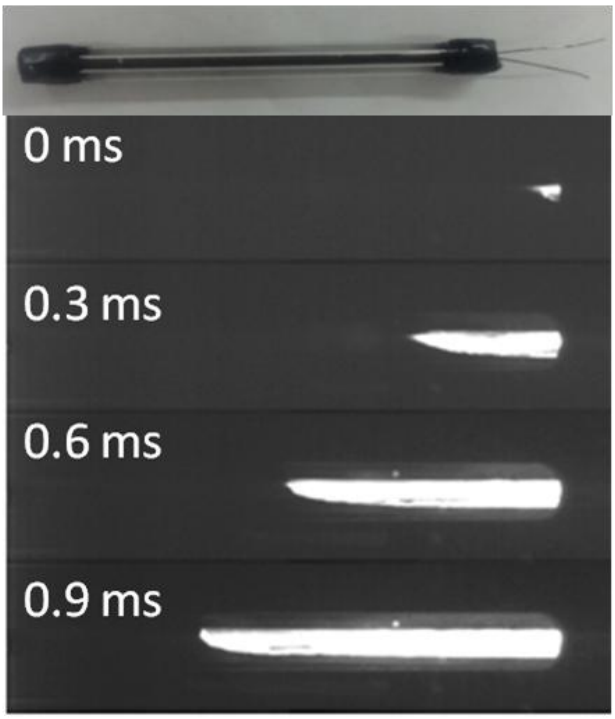


Figure 4: Powder filled quartz tube and representative still frame images time stamped of powder filled quartz tube and flame propagation. Bulk density is 19% theoretical maximum density. Note nichrome wire extruding for left end of the tube.

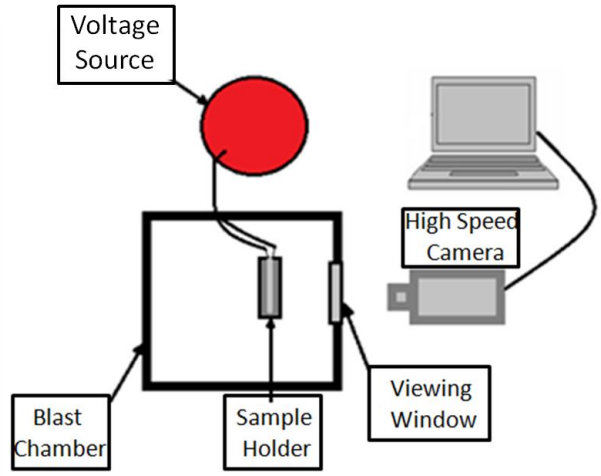


Figure 5. Ignition setup for tracking energy propagation.

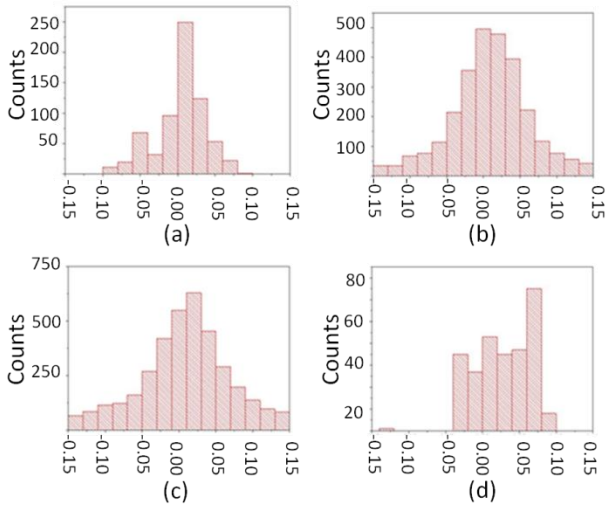


Figure 6. Dilatation strain for (a) untreated particles and particles annealed to (b) 100 ^{o}C, (c) 200 ^{o}C and (d) 300 ^{o}C.

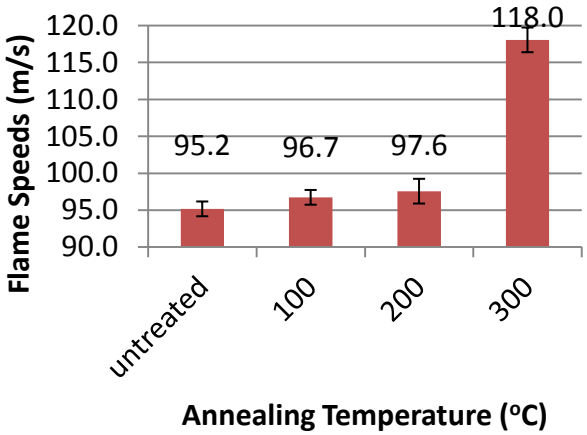


Figure 7. Flame speed as a function of annealing temperature. Average values for flame speed are reported above the bar and standard deviation in the measurements is also shown.

Improving Aluminum Particle Reactivity by Annealing and Quenching Treatments: Synchrotron X-ray Diffraction Analysis of Strain

Jena McCollum and Michelle Pantoya

Mechanical Engineering, Texas Tech University, Lubbock, TX 79409

Nobumichi Tamura

Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

Table 1. Dilatational strain in Al particles, flame speeds when combined with CuO, and percent increase for all Al annealing temperatures. The average strain for the samples annealed to 100 and 200 ^{o}C are within the resolution of the machine, so their percent increase is negligible.

Annealing Temperature (^{o}C)	Average strain	% increase	Flame Speed (m/s)	% increase
untreated	1.00E-05	---	95.2	---
100	1.47E-05	---	96.7	---
200	1.53E-05	---	97.6	---
300	3.25E-05	660	118	24

*Graphical Abstract