

REPORT DOCUMENTATION PAGE

Form Approved
OMB NO. 0704-0188

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1. AGENCY USE ONLY (Leave Blank)		2. REPORT DATE 4/17/2010	3. REPORT TYPE AND DATES COVERED
4. TITLE AND SUBTITLE Melt dispersion mechanism for fast reaction of aluminum particles: extension for micron scale particles and fluorination		5. FUNDING NUMBERS W911NF-04-1-0217	
6. AUTHOR(S) V. I. Levitas, M. L. Pantoya and K.W Watson		8. PERFORMING ORGANIZATION REPORT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Texas Tech University, Mechanical Engineering Department, Lubbock, TX 79409		10. SPONSORING / MONITORING AGENCY REPORT NUMBER	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) U. S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211		11. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.	
12 a. DISTRIBUTION / AVAILABILITY STATEMENT <i>Approved for public release; federal purpose rights</i>		12 b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) The theoretically predicted relationship for the relative flame rate versus relative particle size based on the melt dispersion mechanism MDM, which was previously confirmed for oxidation of 40-120 nm diameter aluminum particles, is found to be in agreement with experiments for 1-3 μm diameter Al particles and fluorination. The main physical parameters for MDM pressure in molten particle, cavitation threshold, and nanoclusters' velocity have been estimated for micron scale particles. The results suggest parameters that could be controlled during particle synthesis that would enable micron scale Al particles to react and achieve the performance of nanoscale Al particles.			
14. SUBJECT TERMS Reaction Mechanisms, Thermite Combustion, Reactive Materials, Aluminum oxidation, Aluminum nanoparticles		15. NUMBER OF PAGES 3	16. PRICE CODE
17. SECURITY CLASSIFICATION OR REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION ON THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT <i>unclassified</i>

NSN 7540-01-280-3500

Standard Form 298 (Rev. 2-89)
Prescribed by ANSI Std. Z39-18
298-102

Enclosure I

Melt dispersion mechanism for fast reaction of nanothermites

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(Received 15 February 2006; accepted 16 June 2006; published online 16 August 2006)

An unexpected mechanism for fast oxidation of Al nanoparticles covered by a thin oxide shell (OS) is proposed. The volume change due to melting of Al induces pressures of 0.1–4 GPa and causes spallation of the OS. A subsequent unloading wave creates high tensile pressures resulting in dispersion of liquid Al clusters, oxidation of which is not limited by diffusion (in contrast to traditional mechanisms). Physical parameters controlling this process are determined. Methods to promote this melt dispersion mechanism, and consequently, improve efficiency of energetic nanothermites are discussed. © 2006 American Institute of Physics. [DOI: 10.1063/1.2335362]

Thermites are mixtures of metal fuel (e.g., Al, Mg, and B) and oxidizers (e.g., Fe₂O₃, MoO₃, and CuO) which react to produce high temperatures with slow oxidation reactions. Since fuel particles are covered by a thin OS, oxidation is controlled by diffusion of molecules through a growing OS. However, when the initial external radius \tilde{R} of the fuel particle is reduced to 10–60 nm (in contrast to traditional 1–100 μm size), reaction rates drastically increase and flame propagation rates C reach 0.9–1 km/s, ^{1–3} in contrast to cm/s for traditional thermites. Rise time for temperature and pressure at the reaction front is $t_f \approx 10 \mu\text{s}$ which characterizes the reaction time. For self-diffusion coefficients of oxygen and Al in α -alumina at 800–950 °C, $D = 10^{-19}$ and $10^{-18} \text{ cm}^2/\text{s}$,⁴ the diffusion length $l_d = 2\sqrt{Dt_f} = (2-6) \times 10^{-5} \text{ nm}$, which is 10^5 times smaller than the OS thickness. Also, C and ignition time delay t_{ig} are independent of particle size for $R < 40-50 \text{ nm}$, while for diffusion controlled oxidation $C \sim 1/R$ (Ref. 2) and $t_{ig} \sim R^n$.⁵ Thus, the only known oxidation mechanism via diffusion does not apply for nanoparticle combustion; finding such a mechanism for nanosize thermites is one of the most important problems in combustion physics. In this letter, we suggest and justify theoretically and experimentally an unexpected oxidation mechanism during fast heating which explains numerous puzzles observed experimentally in flame propagation of Al+MoO₃ (Refs. 1, 2, and 6) and Al+Fe₂O₃ (Ref. 3) nanothermites. Fast heating creates huge thermal stresses due to a difference in thermal expansion coefficients α of Al and Al₂O₃ and volumetric strain $3\epsilon^m$ during the Al melting. Due to the small size of the amorphous OS, $\delta = 1-4 \text{ nm}$, it is defect-free and its strength approaches the theoretical strength. For small particles (small $M = R/\delta$) the OS fractures after melting of an Al particle (in contrast to slow heating or micrometer size particles). Pressure in the melt is very high (0.1–4 GPa). Dynamic spallation of OS results in complete exposure of the liquid Al droplet which leads to an unloading wave within the liquid Al droplet and creates high tensile pressure. The tensile pressure disperses the Al droplet into small clusters that are ejected at a high velocity. The

oxidation of these clusters is not limited by diffusion. The suggested melt dispersion mechanism (MDM) is much faster than diffusion and is independent of the size of Al particle (below some critical size); fast reaction provides fast flame propagation and fast heating rates for the next particles. The necessary conditions for operation of this mechanism and controlling physical parameters are found. By altering controlling physical parameters we suggest ways to promote the MDM, and thereby improve the efficiency of energetic nanothermites. Increasing the temperature T_0 at which initial OS is formed, creating initial porosity in the Al, and alloying Al to decrease the cavitation pressure are ways to induce the MDM. Theoretical results are consistent with observations. A similar mechanism is expected for nitridation and fluorization of Al. The MDM may also be tailored for Ti and Mg fuels.

Let us consider internal stresses during the heating of spherical Al particle with radius R covered by a thin alumina OS with external radius \tilde{R} . The surface temperature of the particle varies from $T_r = 300 \text{ K}$ to the final $T_f \approx 1000 \text{ K}$ (just above melting temperature of Al, $T_m = 933.67 \text{ K}$) during a time t_f . It is easy to show that for $R < 9 \mu\text{m}$ the temperature in the Al sphere is homogeneous. Stresses for a large Al particle covered by OS were found in Ref. 7; we generalize these results for small particles and apply the proper strength criterion. Internal stresses arise during heating due to a difference in inelastic strains $\epsilon_2^i - \epsilon_1^i$ (subscript 1 is for Al and 2 is for Al₂O₃). Surface tensions at the Al–Al₂O₃ interface, Γ_1 , and between alumina and gas, Γ_2 , cause jumps in pressures of $2\Gamma_1/R$ (at $r=R$) and $2\Gamma_2/\tilde{R}$ (at $r=\tilde{R}$). Pressure in the Al sphere, p , and maximum hoop stresses in OS at $r=R$, σ_h , can be found from the elasticity theory:⁸

$$p = \frac{12(m^3 - 1)(\epsilon_2^i - \epsilon_1^i)G_2K_1K_2}{H} + \frac{2K_1(4G_2 + 3m^3K_2)\Gamma_1}{RH} + \frac{2m^2K_1(4G_2 + 3K_2)\Gamma_2}{RH}, \quad (1)$$

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TABLE I. Material parameters at $T=T_m$.

K_1^s (GPa)	K_1^m (GPa)	K_2 (GPa)	G_2 (GPa)	α_1^s (10^5 K^{-1})	α_1^m (10^5 K^{-1})	α_2 (10^5 K^{-1})	ε^m	Γ (GPa nm)	ρ_1^s (kg/m^3)	ρ_1^m (kg/m^3)
71.1 ^a	41.3 ^b	234.8 ^c	149.5 ^c	3.032 ^a	4.268 ^d	0.778 ^c	0.02 ^e	1.05 ^f	2530 ^g	2380 ^d

^aSee Ref. 13.^bSee Ref. 14.^cSee Ref. 12.^dSee Ref. 16.^eSee Ref. 7.^fSee Ref. 17.^gSee Ref. 15.

$$\sigma_h = -\frac{6(m^3 + 2)(\varepsilon_2^i - \varepsilon_1^i)G_2K_1K_2}{H} + \frac{4(m^3 + 2)G_2K_2\Gamma_1}{RH} + \frac{2m^2(-2G_2K_1 + 3(2G_2 + K_1)K_2)\Gamma_2}{RH}, \quad (2)$$

$$\varepsilon_1^i = -(\alpha_1^s(T_m - T_0) + (1 - f)\alpha_1^s(T - T_m) + f\alpha_1^m(T - T_m) + f\varepsilon^m), \quad (3)$$

$$\varepsilon_2^i = -\alpha_2(T - T_0).$$

Here K and G are the bulk and shear moduli, $K_1 = fK_1^m + (1 - f)K_1^s$, $m = \tilde{R}/R = 1 + \delta/R$, f is the volume fraction of the melt in Al particle, subscripts s and m are for the solid and melt phases, and $H = 3m^3K_1K_2 + 4G_2(K_1 + (m^3 - 1)K_2)$. The criterion for brittle fracture of OS (maximum tensile stress reaches the ultimate tensile strength σ_u) is $-\sigma_h = \sigma_u$.

All material parameters are given in Table I ($\Gamma_1 = \Gamma_2 = \Gamma$). For amorphous OS of $\delta = 1.5 - 8$ nm, defects are neglected, thus the ultimate strength is equal to theoretical strength, σ_{th} . For crystalline materials, $\sigma_{th} = E/20 - E/10$ but we conservatively take $\sigma_u = E_2/30 = 11.33$ GPa and consider much smaller σ_u as well. Also, because of lack of stress concentrators and fast loading, fracture and spallation occur throughout the entire OS.

Using Eq. (2) and applying the condition $-\sigma_h = \sigma_u$, the value of volume fraction of melt, $f_f(\sigma_u, M, \delta, T_0)$, necessary for the fracture of the OS is obtained. The functions $f_f(M)$ for $T_0 = 600$ K and several values of σ_u and $\delta = 3$ are shown in Fig. 1(a). They do not change significantly for $1.5 < \delta < 8$ nm. In small particles the fracture criterion is not fulfilled even at complete melting. Then OS spallation will occur at $T > T_m$, due to growth of thermal stresses and reduction in σ_u with the temperature increase. In such a case, an unloading wave in Al particle after OS spallation produces tensile pressure and disperses the liquid Al droplet (see below).

Relationships $p_0(M)$ for pressure in Al particle at the instant of OS fracture are shown in Fig. 1(b). For $\sigma_u = 9.0$ GPa and $M = 19.0$, pressure at the complete melting is 1.0 GPa; for smaller σ_u , complete melting occurs at smaller M and pressure is even larger. For comparison, for $T_0 = 300$ K we obtain complete melting at $M = 10.8$ and $p = 1.8$ GPa; for $T_0 = 900$ we have $M = 80$ and $p = 0.24$ GPa. Thus, T_0 is an important parameter controlling which MDM can be produced even for large particles.

Thus at fast heating, the major part of the Al nanoparticle melts before fracture of OS, producing high pressure. Then unloading wave after OS spallation disperses the liquid particle. For smaller particles, a larger portion of the Al melts and the pressure in the Al is higher (Fig. 1). The MDM

should not occur during slow heating⁹ and for large particles. For $\sigma_u = 9.0$ GPa and $R/\delta = 2000$, we obtain $p_0 < 9.0$ MPa which may not produce sufficient tensile stress in the unloading wave to disperse solid Al particles. For $T = T_m$, $\sigma_u = 9.0$ GPa, and $T_0 = 300$ K, we have $f_f = 0.21$ only.

Let a sphere of radius R be initially in equilibrium under applied external pressure p_0 . We found analytically pressure and velocity evolutions after reduction of the pressure at the boundary $r = R$ from p_0 to the final value p_f during the time of OS spallation, t_s , after which it remains constant. We define an acoustic time $t_p = R/c$ during which boundary regime propagates to the center of the sphere, where $c = \sqrt{K_1^m/\rho_1^m} = 4166$ m/s is the sound velocity in Al. Thus, for $R = 41.66$ nm we have $t_p = 10$ ps. We consider unloading and the first reflected wave only. For fast heating of an Al nanoparticle with a thin OS when the OS fractures at reaching its theoretical strength, $t_s \ll t_p$; we take $t_s = 0.2t_p$. For $R = 40$ nm, we take $p_0 = 1$ GPa and $p_f = 2\Gamma/R \approx 0.05$ GPa.

The results shown in Fig. 2 are typical for the regime with $t_s \ll t_p$ and $p_f \ll p_0$. For $t_s < t < t_p$, a region with tensile pressure appears and propagates to the center of the sphere; the magnitude of tensile pressure grows drastically (Fig. 2). Maximum values of tensile pressure and velocity in the central part of the sphere reach $|p| = 3.75$ GPa and $v = 240$ m/s, respectively.

Fracture of liquid (cavitation) criterion is $p < p_c < 0$, where p_c is the critical tensile pressure; we can estimate p_c

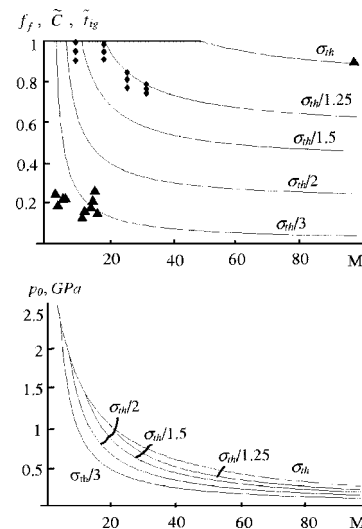


FIG. 1. Volume fraction of melt necessary for fracture of OS f_f (a) and pressure p_0 in Al particle at the fracture of OS (b) as a function of $M = R/\delta$ for various values of ultimate strength σ_u (shown near the curves); $\delta = 3$ nm, $T_0 = 600$ K. The upper line, common for all curves, corresponds to complete melting. Diamonds represent dimensionless flame velocity $\tilde{C} = C/(1 \text{ km/s})$ vs M from experiments (Ref. 2). Triangles are the relative ignition time $\tilde{t}_{ig}/(100 \text{ ms})$ from experiments (Ref. 5).

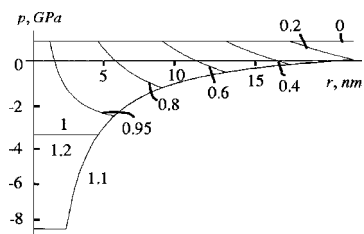


FIG. 2. Pressure (GPa) distribution in Al sphere after spallation of OS at different instances of propagation of the unloading wave (shown near the curves as a fraction of t_p). The upper line ($t=0, p=1$ GPa) and the lowest curve are common for all curves for $t \leq t_p$, i.e., at each time the pressure profile consists of straight horizontal line (1 GPa) in the central part, the line connecting to the lowest curve and part of the lowest curve.

$= -(0.01 - 1)$ GPa. For $p_c = -0.1$ GPa, the cavitation criterion is satisfied at radius $r_f = 0.85R$ (Fig. 2). Cavitation results in separation of a thin liquid outer shell from the main droplet. The thickness of this shell during flight will get smaller down to atomic size then it will break into small sectors. Since tensile pressure without fracture exceeds significantly $|p_c|$ (Fig. 2), the thickness of each subsequent separated liquid shell is of order of several atomic sizes. Al clusters may react with oxygen and nitrogen in the air, gaseous MoO_3 which starts to sublime at 973 K, with Teflon which decomposes at 825 K, or solid oxidizers.

For the same conditions but for $t_s = t_p$, there is no tensile pressure in Al for $t < t_p$. For $t > t_p$ cavitation is in principle possible but not very probable. For $t_s = 2t_p$, tensile pressure does not appear in the unloading and first reflected waves, i.e., dispersion cannot occur.

If, e.g., $\sigma_u = 9$ GPa, then for $M \leq 19$ the entire particle melts before OS fracture [Fig. 1(a)] (pressure $p_0 = 1.0$ GPa for $\delta = 3$) and disperses providing maximum possible mixing, oxidation rate, energy release, and flame front velocity. According to the MDM, there should not be an essential difference in the mixing of components and oxidation rate of particles with $R/\delta \leq 19$. In our experiments,² flame velocity is approximately the same (950 m/s) for the Al particles with an averaged $R = 20$ and 38 nm and reduces for particles with $R = 53$ and 58.5 nm ($\delta \approx 2$ nm) [Fig. 1(a)]. In our recent experiments,⁵ laser induced ignition times are in the range of 12.4–25.5 ms for $R = 6.7$ –99 nm and grows to 6.039 s for $R = 20 \mu\text{m}$ [Fig. 1(a)]. These results support our theory.

Damage of the OS should suppress the MDM but promote traditional diffusion oxidation mechanism. Pressing thermite pellets in a die potentially damages the OS. That is why C is expected to decrease with sample density for nanothermites and grow for microthermites. That is exactly what was observed in our experiments.⁶ On the other hand, vibrational compaction without pressure should not damage OS and suppress the MDM. Indeed, vibrational change in density in the range of 5%–10% of TMD did not change C .²

Consider an ellipsoidal penny-shaped particle with semi-axes b and $R \gg b$ that mimics a nanoflake. The hoop stress in the circular section of radius R is $\sigma_h^{\text{max}} \approx -pR/(2\delta)$, while the hoop stress in the elliptic section, $\sigma_h^{\text{min}} \approx -p\pi b/(4\delta)$, is much smaller. Simultaneous spallation of the entire OS cannot occur. Fracture occurs along the circular section of radius R when $\sigma_h^{\text{max}} = -\sigma_u$, i.e., for $p = 2\sigma_u\delta/R$ [like for large spherical particles]. For nanoflakes, $b = 10$ –100 nm, while $R = 10$ –100 μm .¹⁰ Thus, fracture of a flake occurs at a similar pressure as a micro particle. Thus, according to the MDM,

nanoflakes would tend to react like micro particles, while traditional arguments (higher reactivity and large surface to volume ratio) suggest similar reactivity for nanoflake and nanospherical particles. In experiments,¹⁰ nanoflakes did not produce higher flame rate than micro spherical particles.

The main conditions for the dispersion of the spherical Al particle are that it completely melts before OS spallation, that OS fracture time is smaller than wave propagation time ($t_s < t_p$), and that pressure in the particle before OS spallation is approximately equal to the cavitation pressure. These conditions can be fulfilled for a thin amorphous defect-free or nanocrystalline OS, and only during fast heating. We can predict several ways to increase particle radius for which the MDM is operative.

- (1) Our analysis predicts that increasing T_0 may extend the MDM for large particles. If we increase T_0 up to 900 K, then for $\sigma_u = 9.0$ GPa almost the whole particle melts for any particle radius. If we assume that $p_0 = 20$ MPa is sufficient for fracture and dispersion of Al particle, then for $\sigma_u = 9.0$ GPa and $\delta = 8$ nm, we obtain $R = 7.2 \mu\text{m}$.
- (2) To create the condition for fast heating of large particles, they have to be mixed with nanosized particles ($R < 40$ –100 nm) which have much higher probability to be dispersed in a liquid state. Indeed, a mixture of 70% of Al particles with $R = 38$ nm ($R/\delta = 8.3$) and with 30% particles with $R = 2$ and 10 μm provided practically the same flame velocity as 100% of Al nanoparticles.¹¹
- (3) An alternative way to decrease internal stresses before melting and to ensure complete melting before OS fracture is to introduce initial porosity in the Al particles.
- (4) Alloying the fuel particle may reduce its local surface energy and cavitation pressure.

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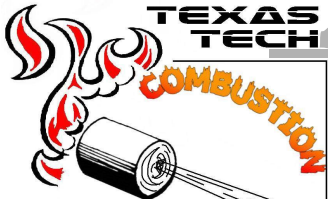
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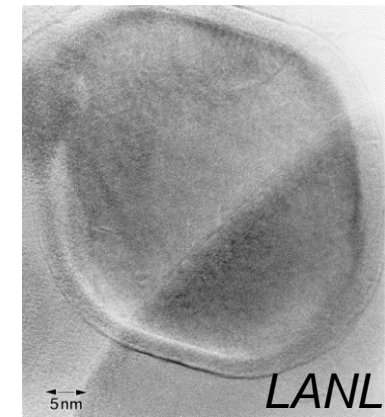
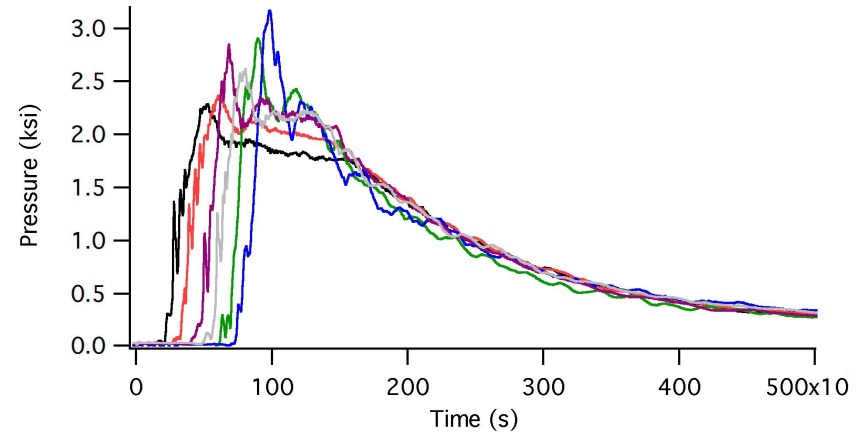
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Fast oxidation of Al nanoparticles: diffusion models do not describe this process

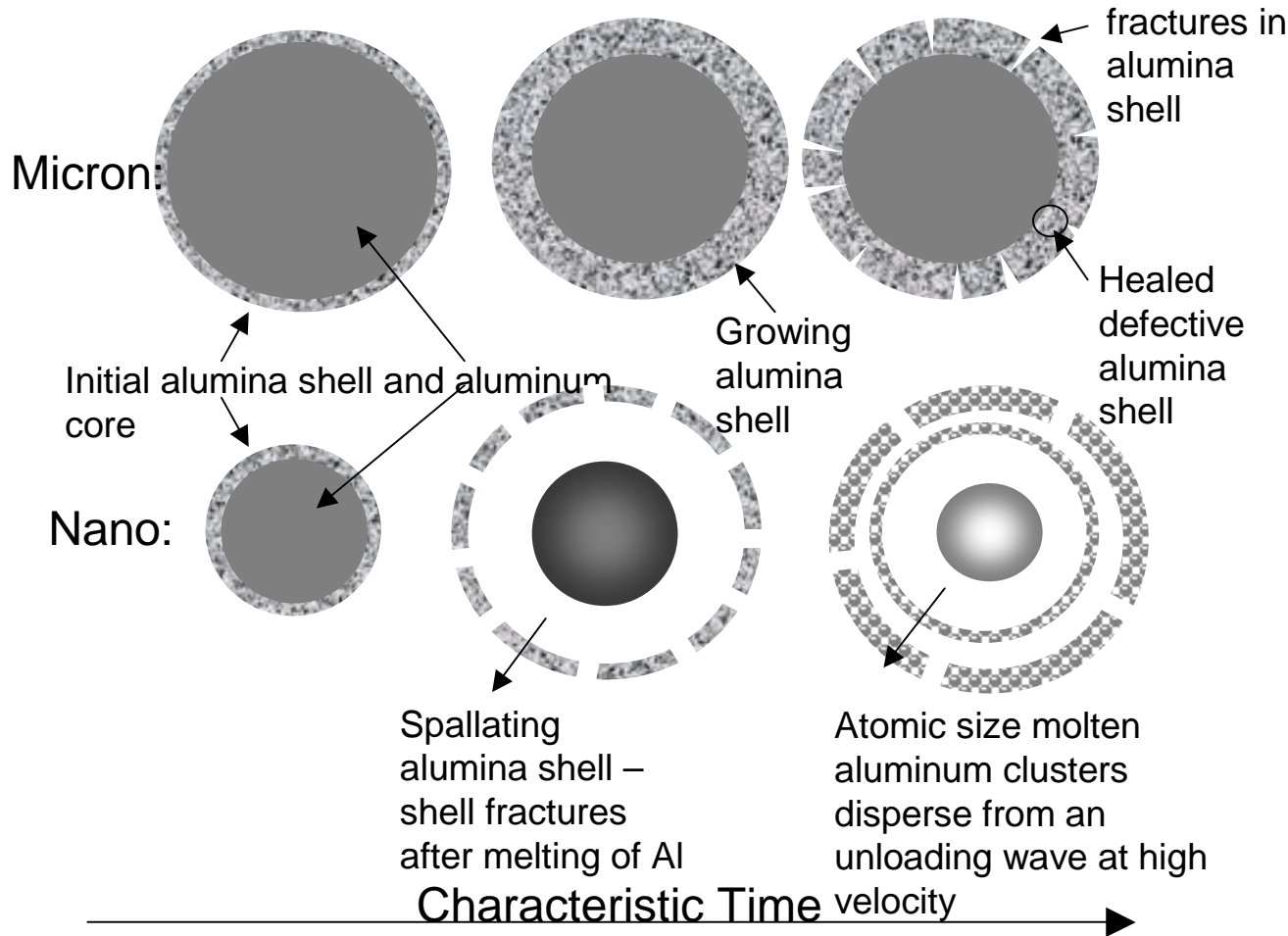
- Rise time for T and P $\tau \sim 10 \mu\text{s}$
 - characterizes the reaction time
 - Diffusion coefficient of Oxygen & Al in α -alumina at 800 °C is $D \sim 10^{-19} \text{ cm}^2/\text{s}$
 - Diffusion length $l_d = 2\sqrt{D\tau} = (2-6)10^{-5} \text{ nm}$
 10^5 times smaller than oxide shell thickness!!
 - Diffusion time would need to be increased by a factor of 10^{10} to obtain a reasonable diffusion length of 2-6nm.
- Burn rate is independent of particle size for $d_{\text{Al}} < 100 \text{ nm}$, while for diffusion controlled oxidation $\sim 1/r$
- Ignition delay time independent of particle size for $d_{\text{Al}} < 100 \text{ nm}$, while for diffusion controlled oxidation $t \sim r^n$
- Nanoflakes react like micron size particles
- Previous diffusion models do not explain experimental observations!



A TEM of an Al particle



New Mechanism for Fast Reactions



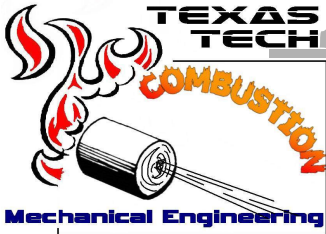
- Due to small size of the amorphous shell and very fast strain and stress growth ($\sim 10^6$ GPa) strength approaches theoretical strength.

- Pressure in the melt is very high (0.1-4 GPa)

- Dynamic spallation of shell results in complete exposure of the liquid Al droplet and creates a tensile pressure (0.1-4 GPa)

- The tensile pressure in an unloading wave disperses the Al droplet into small clusters which fly with high velocity

- Oxidation is not limited by diffusion



Exploit the melt dispersion mechanism

- Suggest methods that will improve the performance of nano and micron size composites by exploiting this new mechanism for fast oxidation.
- Reduction of sensitivity to the ignition in combination with high performance (flame rate) is the main concern.

Controlling parameters

- (1) the temperature at which the oxide shell is formed determines initial stresses in the particle and core;
- (2) the porosity of the Al core;
- (3) ratio of oxide thickness to particle diameter;
- (4) defects of the oxide shell;
- (5) particle size distribution, including combination of nano- and micron size particles.

