The Fabrication of Functionally Graded Energetic Materials Using Twin_Screw Extrusion

Frederick Gallant
NSWCIHDIV-Indian Head,MD
& Dept. of Mechanical Engineering
University of Maryland

Hugh A. Bruck
Dept. of Mechanical Engineering
University of Maryland

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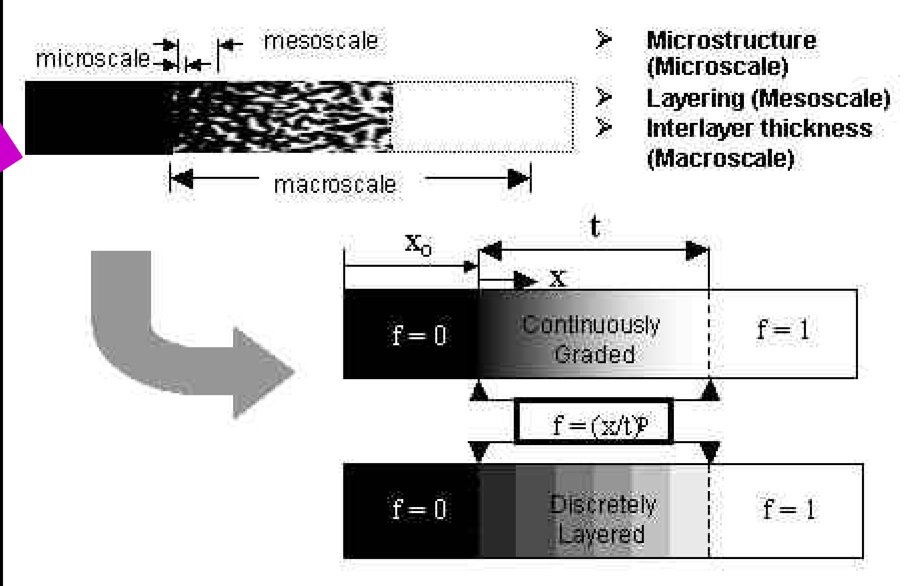
Technical Objective

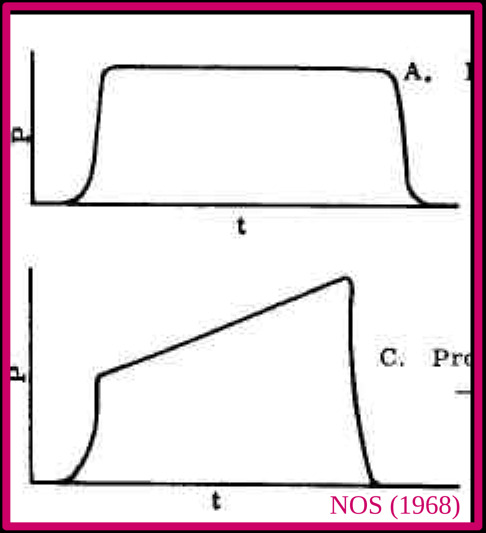
Tailor Burn Rate Performance in a Monolithic Rocket Motor Utilizing New Design and Control Schemes for Twin Screw Extrusion based on Functionally Graded Material (FGM) Architectures

Propellant Continuously Extruding from Die of TSE

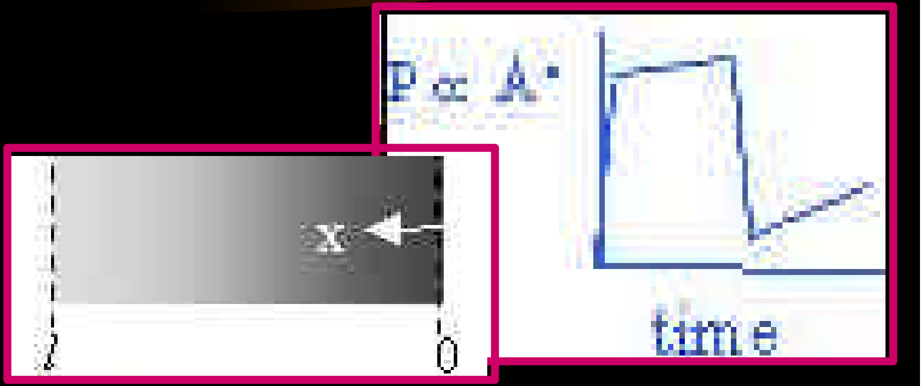


FGM Architecture

Rocket Motors with Two Burning Rates



Examples of Conventional Rocket Motor Pressure-Time Traces



Functionally Graded Rocket Motor Pressure-Time Trace

$$P \propto \dot{r} S r = \dot{m}$$

$$\text{where, } \dot{r} = b\left(\frac{P}{1000}\right)^{n}$$

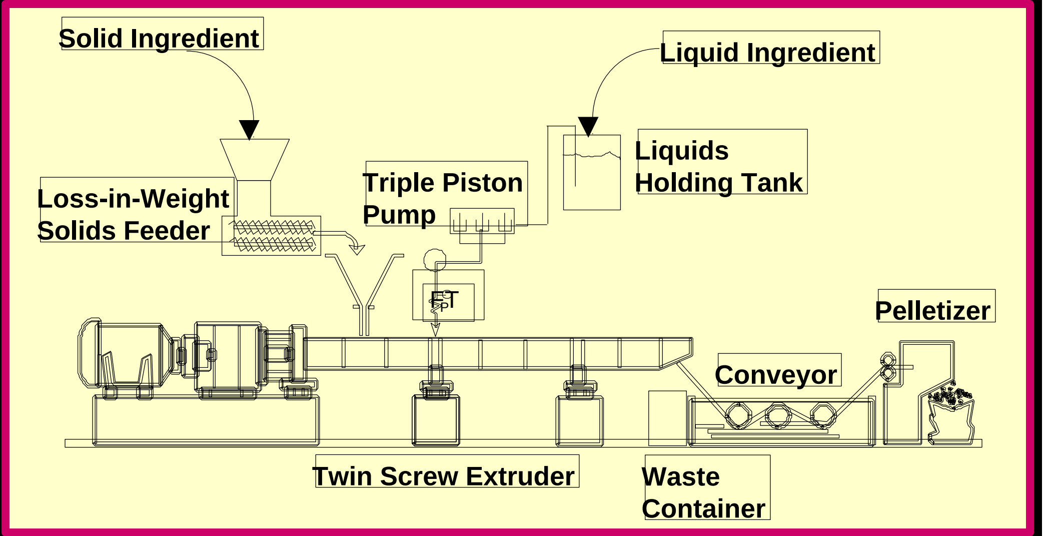
$$P \propto \dot{r}_x S r_x = \dot{m}$$

$$\text{where, } \dot{r}_x = b_x\left(\frac{P}{1000}\right)^{n_x}$$

NOS (1968)

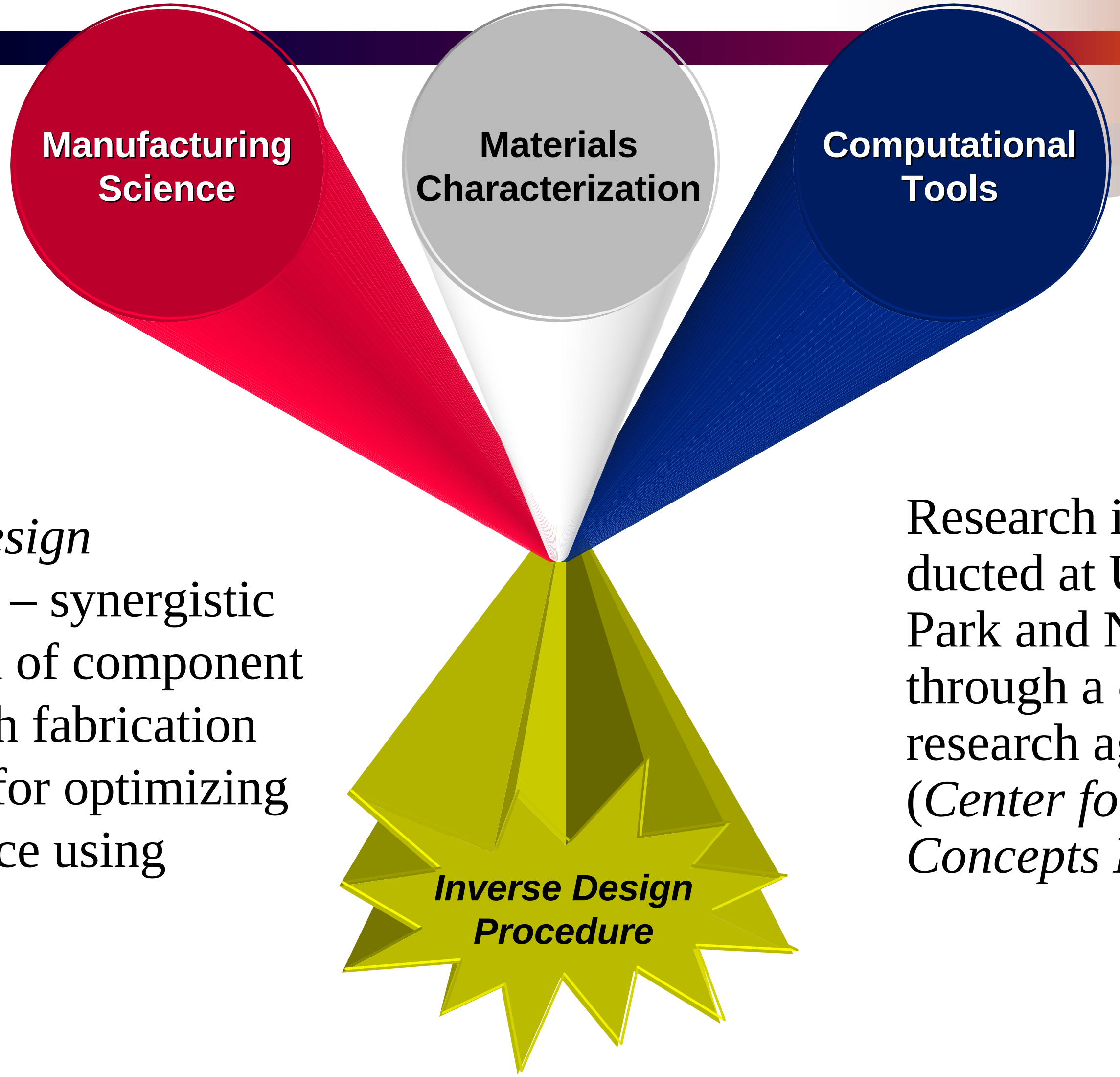
Research Objectives

- How do dynamic variations in process conditions or ingredient addition during TSE affect the evolving microstructure of the extruded composite?
- Can the architectures be predicted by newly-developed residence distribution (RD) models?
- What characterization techniques have to be developed to adequately quantify microstructural variations in extruded material?
- How are these related to burning rate performance?



Twin Screw Extrusion Process

Research Approach



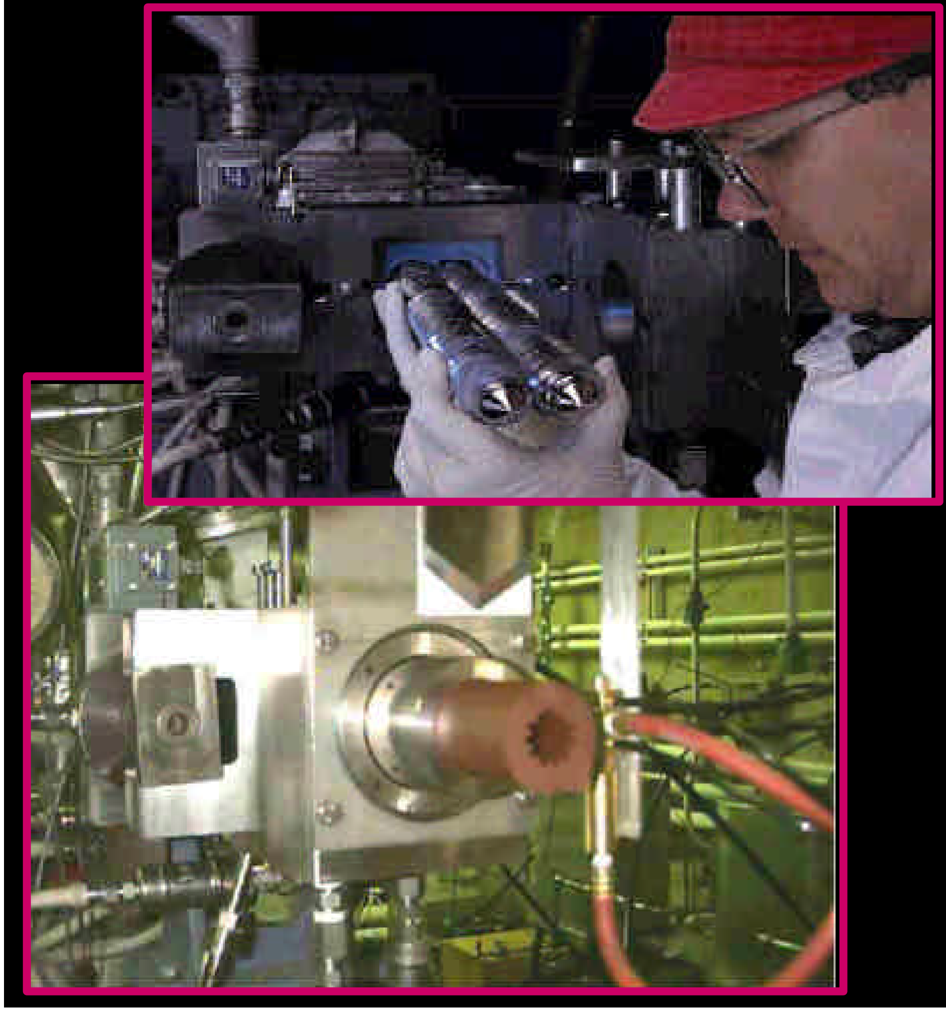
Inverse Design Procedure – synergistic integration of component design with fabrication processes for optimizing performance using FGMs

Research is being conducted at UMD/College Park and NAVSEA-IH through a collaborative research agreement (*Center for Energetic Concepts Development*)

Advance State of the Art

- Composite Energetic Materials have been traditionally manufactured using batch processing
- Current manufacturing of composite energetic materials is focused on homogeneous formulations
- New continuous manufacturing technology known as Twin Screw Extrusion (TSE) is being used to produce higher quality composite energetic materials with more flexibility and control
- The continuous nature of the TSE process is ideally suited for the manufacture of functionally graded materials (not restricted to energetic material)

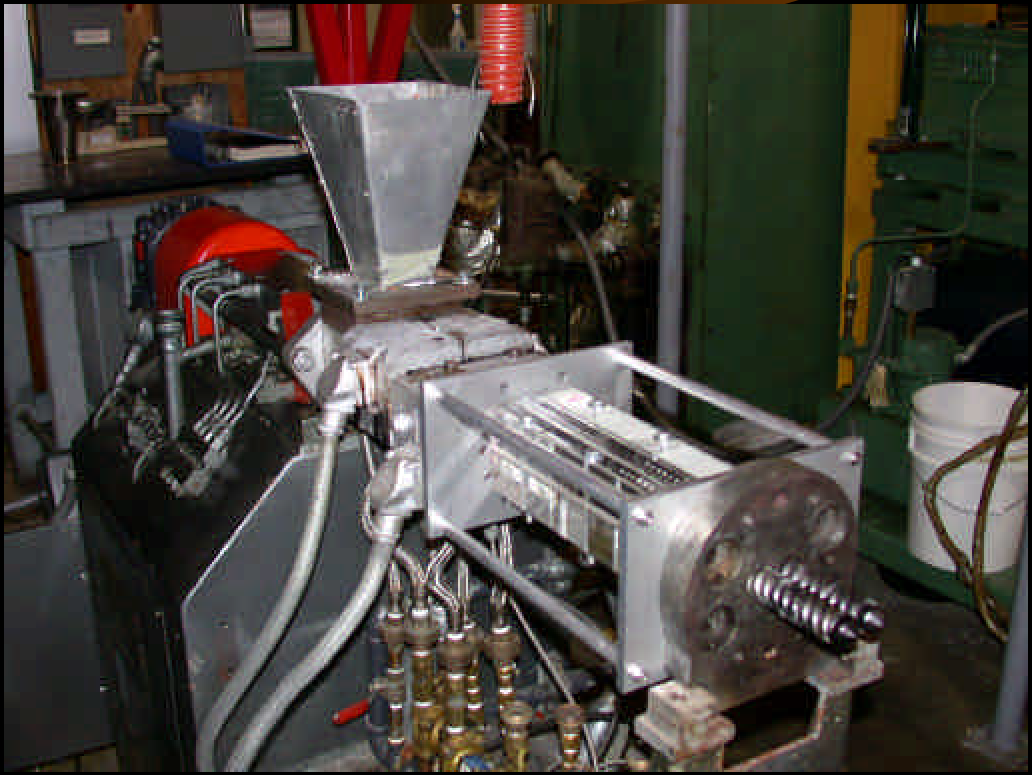
40 mm TSE at Indian Head Process Technology Division



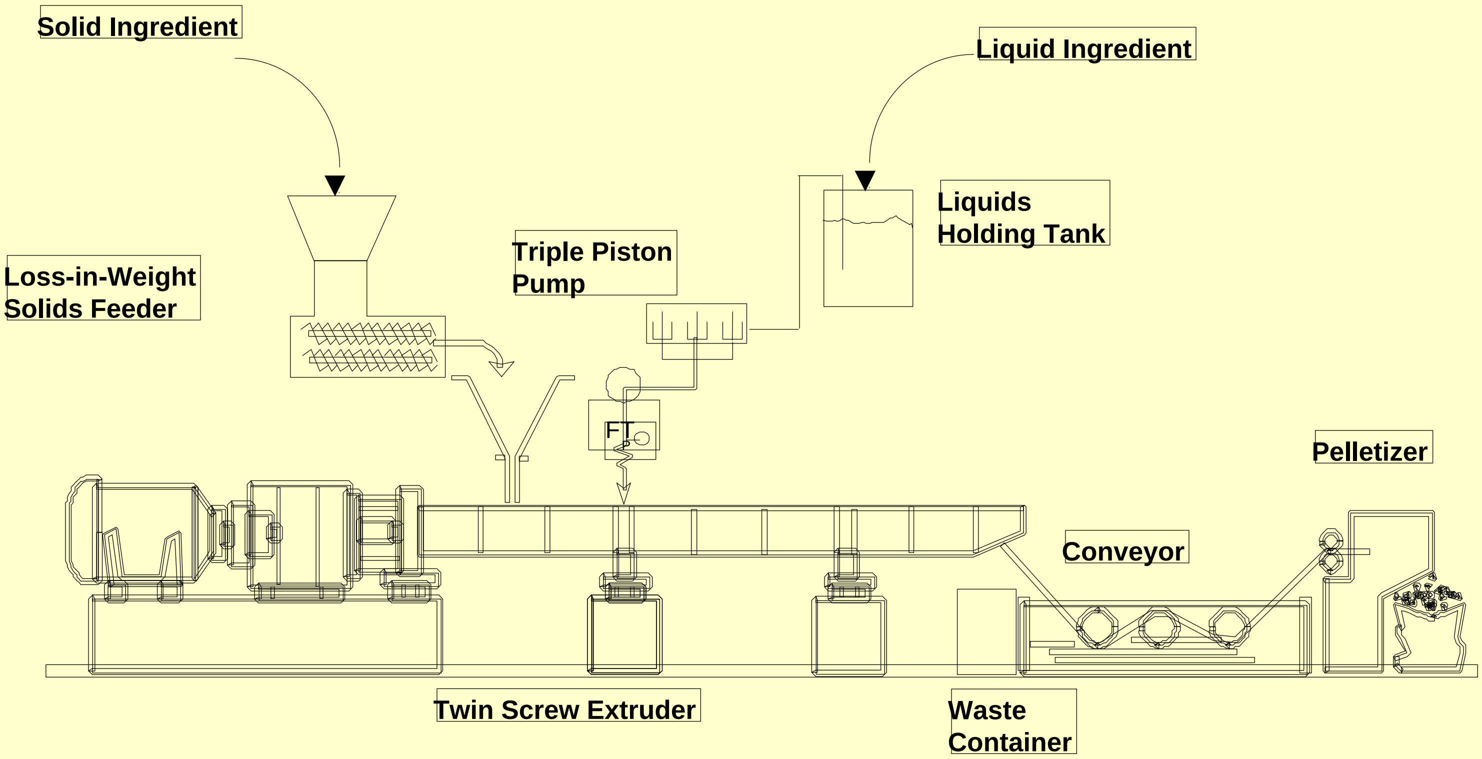
- Pilot Scale, 15 - 100 #/hr
- World-class research facility for energetics
- New and existing propellants and explosives
- Refill capability
- Four LIW feeders, gear & triple-piston pumps
- On-line QA
- Ingredients to grains in one facility in one processing step

28 mm TSE at UMD

- Polymer Processing Laboratory - Dr. Bigio
- Corotating, fully intermeshing
- Research scale, < 10 #/hr
- Available torque is low
- Highly flexible process section
- Large screw inventory
- TSE Installed in 2001
- Feeders Upgraded to Loss-in-Weight Control 2002



Continuous Processing using a Twin-Screw Extruder

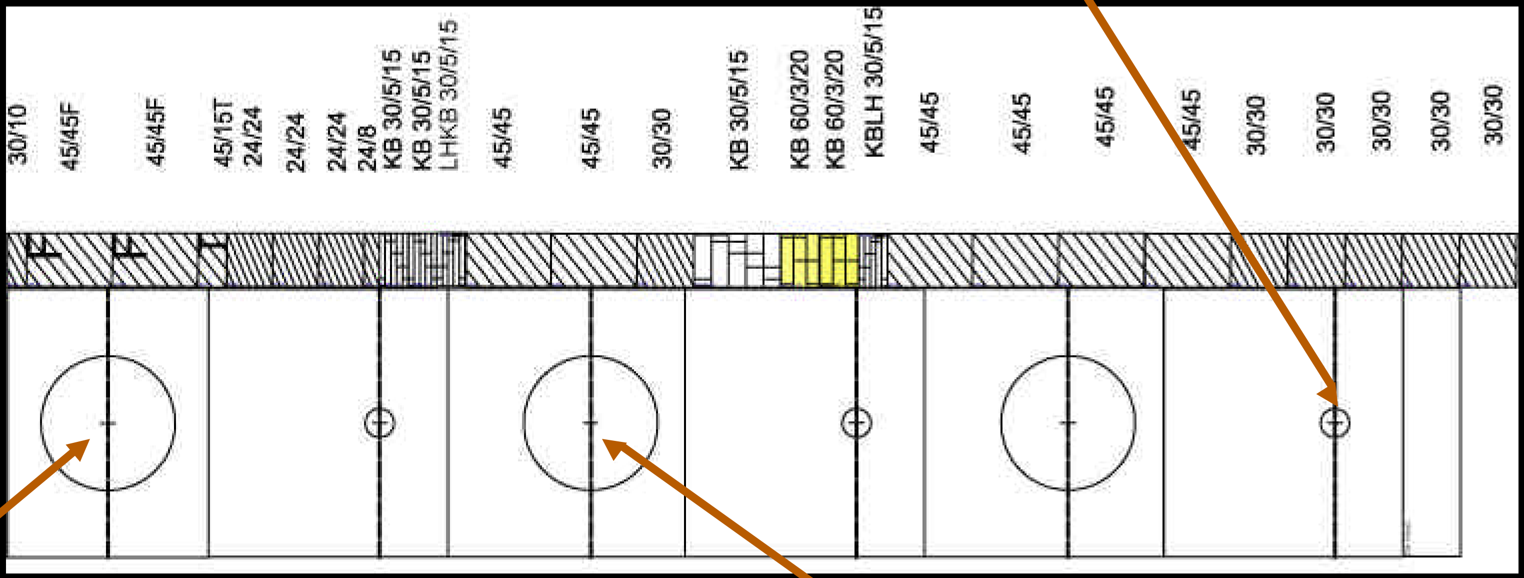


Residence Distribution (RD) Modeling

- Description of flow through stirred tank reactors (Danckwerts, 1953)
  - Age distributions
  - Distribution functions
  - Experimental determination with tracers
- Characteristic description of dampening due to backmixing (Rauwendaal, 1986)
- Characterize the ability of the process to dampen disturbances (Gao, Walsh *et al*, 1999)
- Experimental method: impulse addition of tracer
- Concentration of tracer at exit (or other location)
  - Function of time (Danckwerts)
  - Extended to Volume and RPM domains (Gasner, Bigio *et al*, 1999)

RTD Experiments (UMD)

Fiber Optic Probe

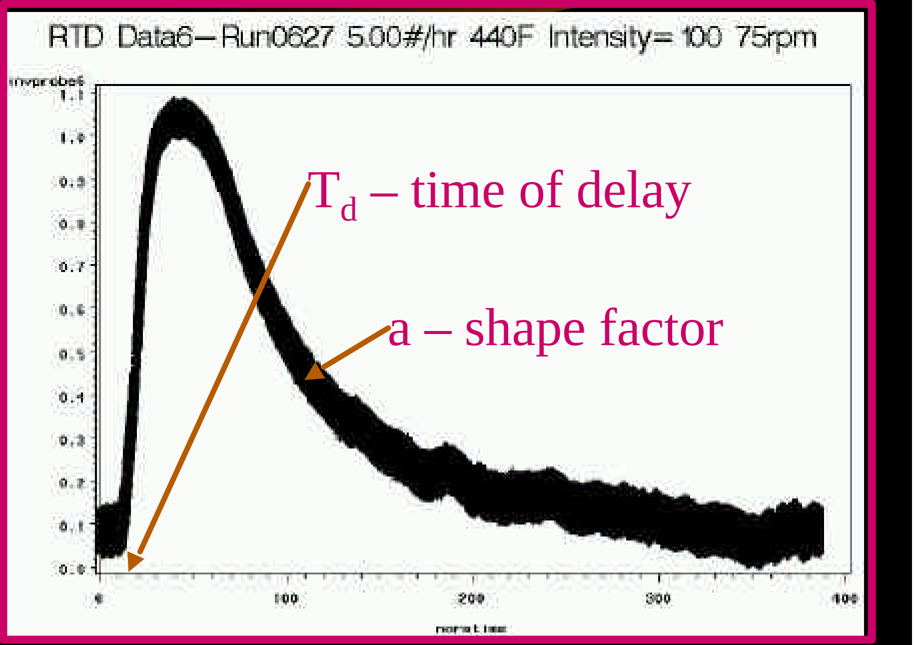


1) Engage® POE 8401 (white)
2) Glass-filled polypropylene (black)
3) Tracer, black color concentrate, 0.50g

KCl (filler)

Gao's Residence Time Distribution (RTD) Model

- Physically-based model
- Screw geometry
- Experimentally derived constant
- Quantifies the effect of a disturbance (e.g., ingredient change to make a FGM)



$$f(t)=\frac{a^3}{2}(t-t_d)^2 e^{-a(t-t_d)}$$

Gao, Walsh *et al* (2000)

Material Transport in TSE

Change in Filled Region Length with Respect to Time

$$\frac{dL_f}{dt} = \frac{(Q_{in} - Q_{out})}{\overline{HW}\,(1-\Phi)}$$

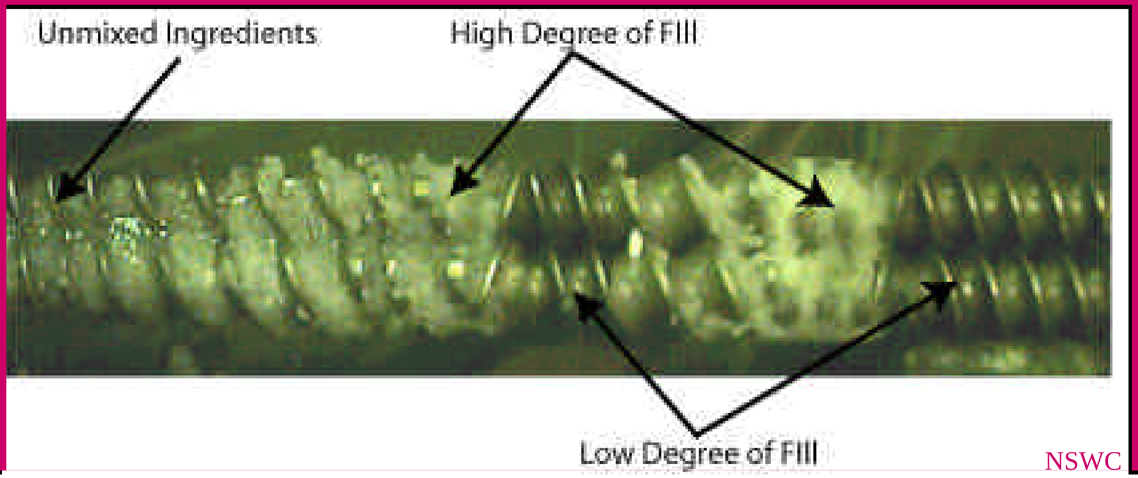
Flow into a Filled Region Described by Starved Flow

$$Q_{in} = Q_{st}(L_{st}, t) = Q_{feed}\left(t - \frac{2(L - L_f)}{V_{bz}}\right)$$

Flow out of a Filled Region Described by Die Flow

$$Q_{out} = Q_{die}\left[\text{die geom., fluid props.,} \left(\frac{dP}{dx}\right)_{die}\right]$$

Mudalamane (2002)



NSWC

Residence Volume Distribution (RVD)

Delay Volume (v_d)

$$v_d = t_d \times Q = A - \frac{3}{C} + B\frac{Q}{N}$$

$$g(v) = \frac{c\left(\frac{v}{Q}\right)}{\int_0^\infty \left| c\left(\frac{v}{Q}\right) \right| dv} = \frac{e\left(\frac{v}{Q}\right)}{Q} = \frac{a_v^3}{2}(v - v_d)e^{-a_v(v-v_d)}$$

Residence *Volume* Distribution

RVD Curve
Shape Parameter

$$a_v = \frac{a}{Q} = C$$

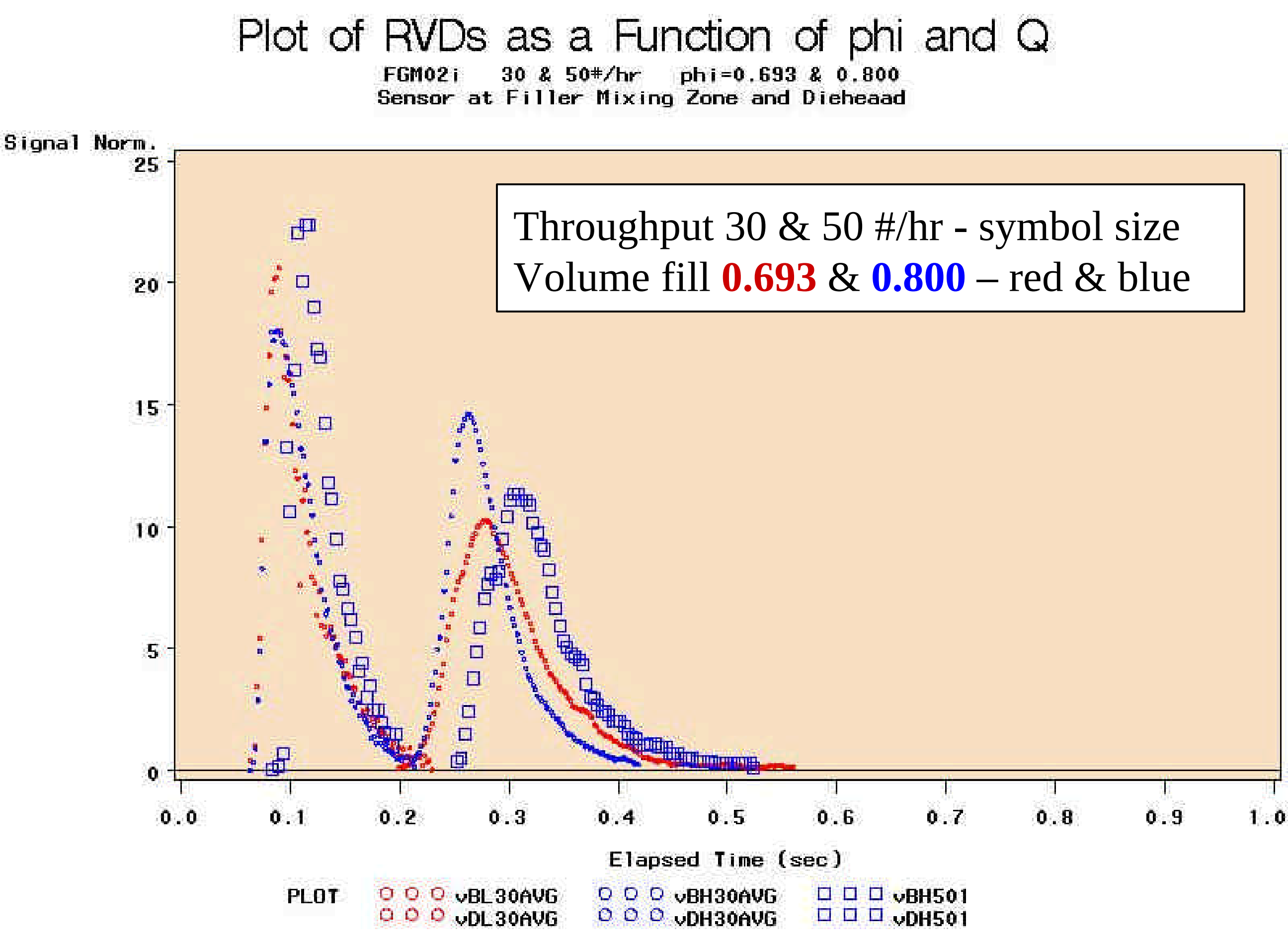
Gao, Walsh *et al* (2000)

RVDs of Inert Composite Propellant using 40mm TSE

-RTDs for Inert Composite as Measured at Second Mixing Zone and the Diehead.

-RVDs not created yet.

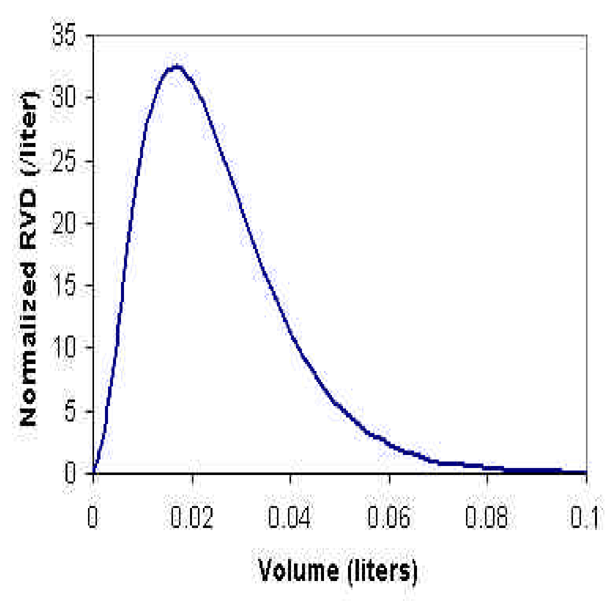
-Similar measurements with live propellant conducted. (Data not analyzed yet.)



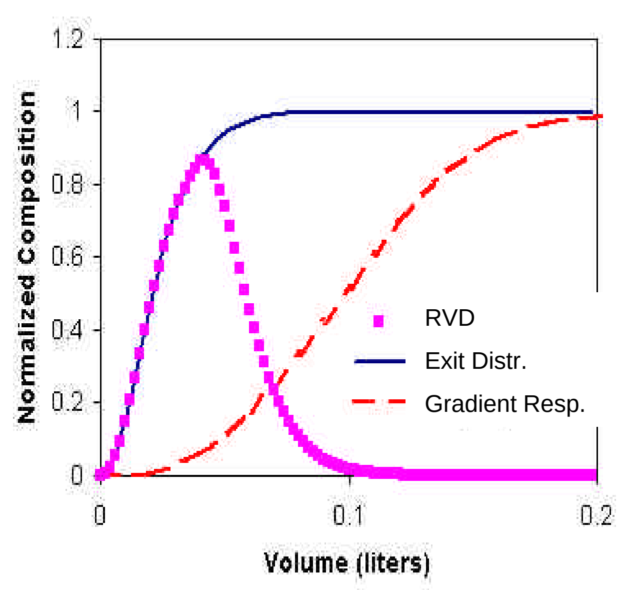
Throughput 30 & 50 #/hr - symbol size
Volume fill **0.693** & **0.800** – red & blue

Prediction of Gradient Architecture

Convolution of RVD to predict responses to step and ramp inputs

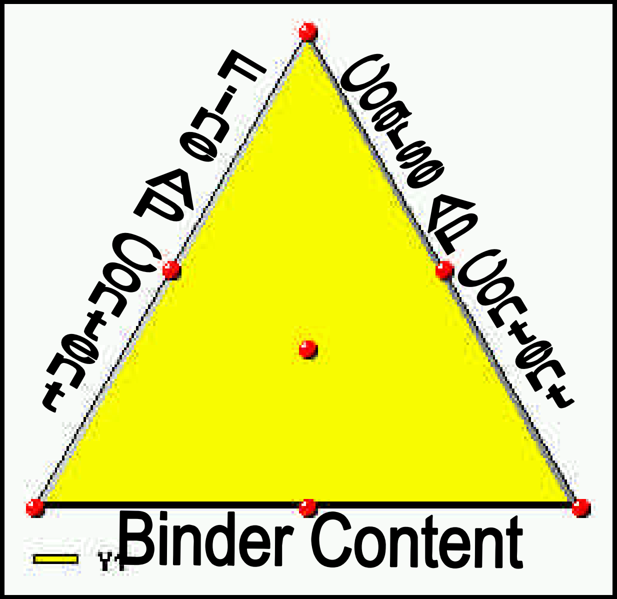


$$f[z(v)] = \int_o^v g(v-v')h(v')dv'$$



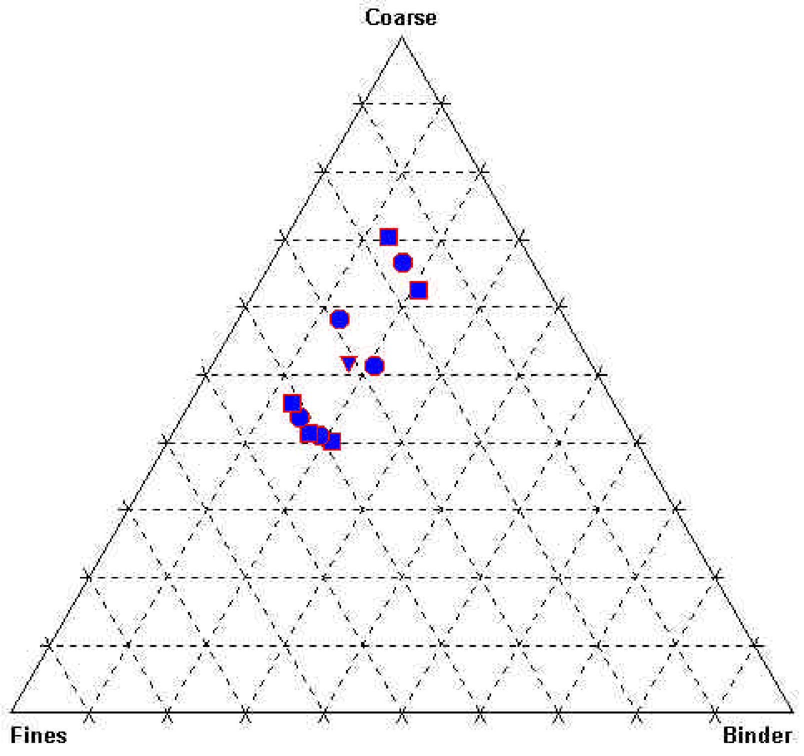
Potential for Creating Gradient using IH-AC3

- Process Constraints
  - Two mixing stages process
  - Available solids feeders
  - Powder properties
- Safety Constraints
  - Ultra high fill
  - Too far from optimum ratio of coarse/fine
- Time Constraints
  - Limited manuf. budget
  - Multiple samples per extruder



Mixture Experiments

Extreme Vertices Design



- Effects of individual ingredients on burning rate
- Three constituents for IH-AC3
- Combined effects of constituents
- Analysis by response surface methods
- Constrained region, Snee (1975)
- Cornell (1990)

Microstructural Characterization

- Techniques Common to Metals, Ceramics, and Propellants
- *Functionally Graded Materials* are Highly Nonhomogeneous:
  - ➢ Structure is Designed
  - ➢ Structure Varies with Position
- Stereological Methods for Grain Size and Topological Distributions
  Liu (2000)
  - ➢ Scanning Electron Microscopy
  - ➢ Optical Stereoscope
- Transmission Electron Microscope, Energy Dispersive X-ray Spectroscopy
- Physical Properties
  - Stress-strain relationships
  - Micro-indentation

Versamet Stereoscope and Optics System

Stereological Analysis

- Average Individual Particle Size

$$d_{av} = \frac{2}{p}\int_0^{p/2}\left(\frac{\cos^2 q}{d_{\max}^2}+\frac{\sin^2 q}{d_{\min}^2}\right)^{-1/2} dq$$

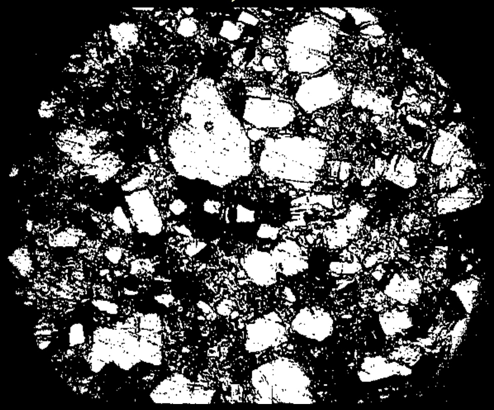
- Average Particle Size for the Distribution of Particles

$$\bar{d}_{av} = \sum g(d_{av})d_{av}$$

- Similar Treatment for Shape Factor Analysis

$$s_f = \frac{d_{\min}}{d_{\max}} \qquad \bar{s}_f = \sum g(s_f)s_f$$

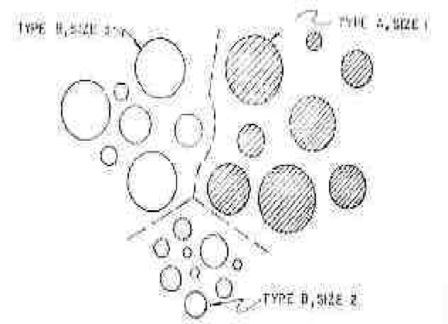
- Texture Analysis of Binary Images, Ohser (1998)
  - Linear filtering
  - Specific line length (volume fraction)
  - Length of total projection (specific surface area)
  - Integral of curvature (specific mean curvature)



Binary Image of IH-AC3 Simulant

Petite Ensemble Model

- Statistically-based combustion model
- Combines Beckstead, Derr, and Price (BDP) model with Glick's statistical formalism
- Models composite propellant as a random arrangement of polydispersed pseudopropellants



Polydispersed pseudopropellants

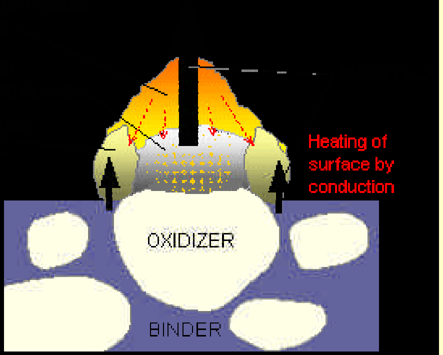
$$F_d = \frac{1}{(2\pi \ln \sigma)^{1/2}} \exp\left[-\frac{1}{2}\left(\frac{\ln D_o - \ln \bar{D}_o}{\ln \sigma}\right)^2\right]$$

$$\bar{r} = \int_{D_o} \frac{r_d F_d}{\alpha_d} d(\ln D_o)$$

$$R_p = \frac{1}{\bar{r}} \int_{D_o} \frac{R_{p,d} r_d F_d}{\alpha_d} d(\ln D_o)$$

$$R_v = \frac{1}{\bar{r}} \int_{D_o} \frac{R_{v,d} r_d F_d}{\alpha_d} d(\ln D_o)$$

Steady–State PEM Calculations (w/o fuel)



$$\bar{r} = m_{ox}^{p} / r_{p}$$

$$m_{ox}^{p} = m_{ox}^{T}\left[3\left(\frac{h}{D_o}\right)_{+}^{2} + 3\left(\frac{h}{D_o}\right)_{-} + 1\right]$$

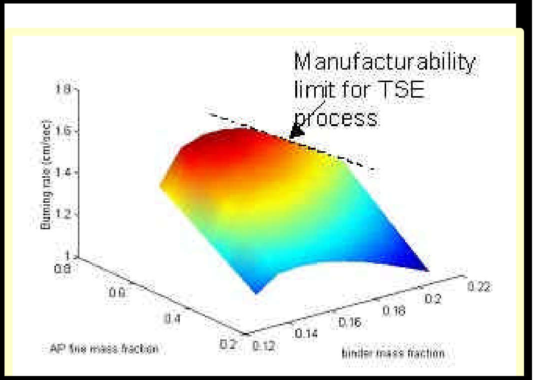
$$m_{ox}^{T} = A_{ox}\exp\left[-\frac{E_{ox}}{RT_{s,ox}}\right]$$

$$\left(\frac{h}{D_o}\right)_{\pm} = f(r_{ox}, t_{ign}, D_o)$$

$$r_{ox} = m_{ox}^{T} / r_{ox}$$

$$t_{ign} = \frac{C_{ign} D_o^{\,d_D+1}}{p d_p}$$

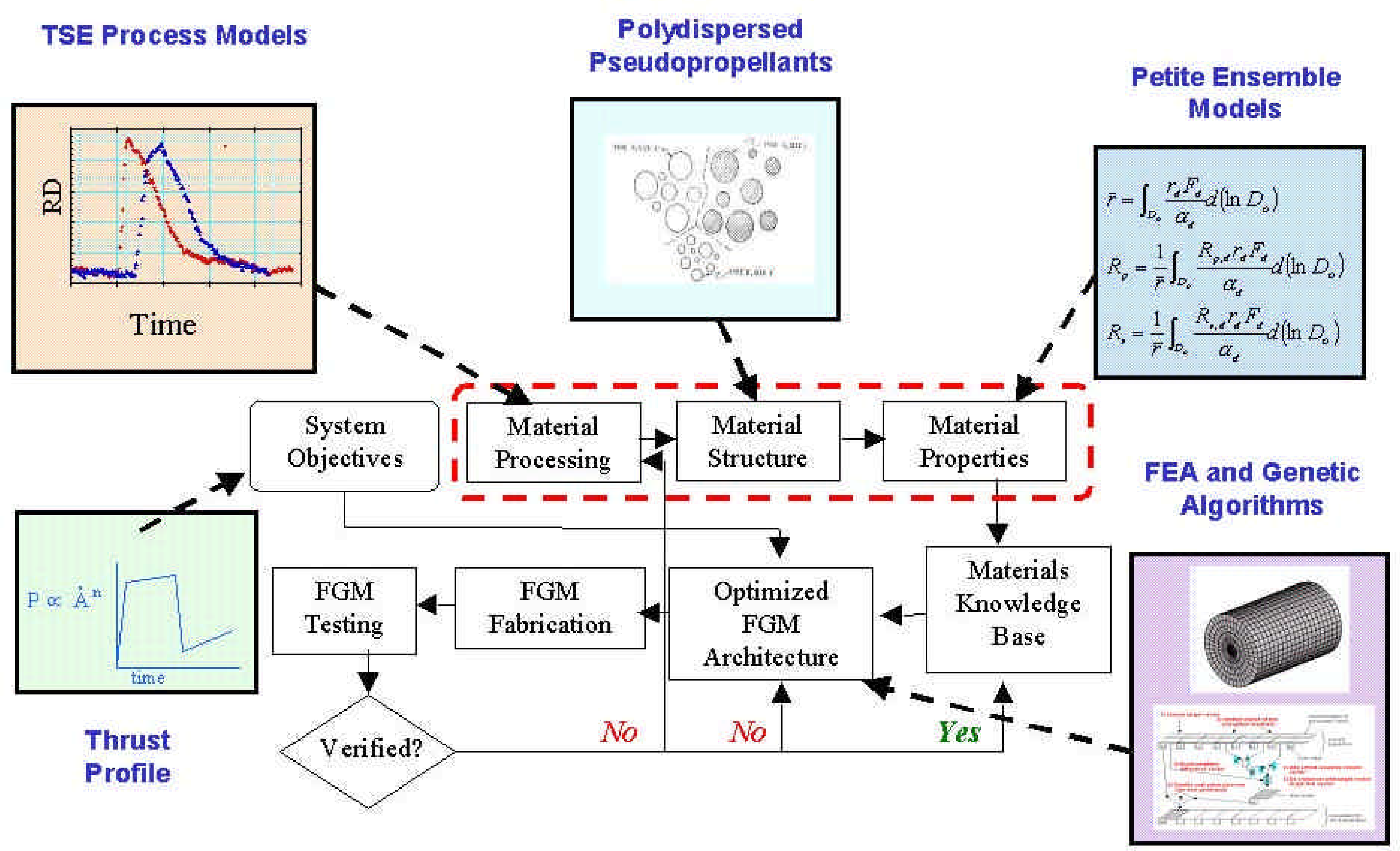
Gradient effects?



Burn rate variation w/ composition

- Use COE to determine $T_{s,ox}$ from adiabatic flame temperature calculations (PEP, NASA SP-273)

Inverse Design Procedure



TSE Process Models

Polydispersed Pseudopropellants

Petite Ensemble Models

$$\bar{r} = \int_{D_o} \frac{r_d F_d}{\alpha_d} d(\ln D_o)$$

$$R_p = \frac{1}{\bar{r}} \int_{D_o} \frac{R_{p,d} r_d F_d}{\alpha_d} d(\ln D_o)$$

$$R_s = \frac{1}{\bar{r}} \int_{D_o} \frac{R_{s,d} r_d F_d}{\alpha_d} d(\ln D_o)$$

System Objectives

Material Processing

Material Structure

Material Properties

FEA and Genetic Algorithms

Materials Knowledge Base

Optimized FGM Architecture

FGM Fabrication

FGM Testing

Verified?

No

No

Yes

Thrust Profile

Conclusions

- Described techniques to quantify dynamic variations in ingredient addition during TSE
- Discussed methods to relate process dynamics to the evolved microstructure of the extruded composite
- Shown how residence distribution (RD) models may be used to predict architectures
- Presented characterization techniques that should quantify microstructural variations in extruded material
- Researching to relate microstructural variations to burning rate performance