ABSTRACT
Aerogels have been of interest to the aerospace community primarily for their thermal properties, notably their low thermal conductivities. While such gels are typically fragile, recent advances in the application of conformal polymer layer deposition [15] have allowed the development of aerogel−alcohol composites with excellent mechanical properties. We have performed computer simulations of aerogel thermal conductivity and tensile and compressive failures, with results that are in qualitative, and sometimes quantitative, agreement with experiment. However, recent experiments in our laboratory suggest that gels with similar densities may exhibit substantially different properties. In the work, we extend our original fracture limited diffusion cluster aggregation (DLCA) model for gel structures to incorporate additional variations in simulation parameters, with the aim of producing DLCA clusters of similar densities that nevertheless have different fractal dimension and secondary particle coordination. We perform particle statics simulations of gel strain on these clusters, and consider the effects of differing DLCA simulation conditions, and the resultant differences in fractal dimension and coordination, on gel strain properties.

INTRODUCTION
Silica aerogels are low-density materials whose thermal properties have made them of ongoing interest for a wide variety of applications [1−5]. While pristine gels are fragile, polymer coating gels can greatly improve their strength while minimally impacting their insulating properties [4].

To provide a microscopic understanding of the mechanical behavior of the gels, and to provide a predictive tool for the development of a multimodal model for the behavior of silica aerogels, we have performed computer simulations for a range of densities (0.0377, 0.0651, 0.127, and 0.302 g/cm

AEROGEL STRUCTURE
Aerogels exhibit a ropelike morphology that displays three levels of structure. X-ray and neutron probes suggest that the gels consist of distributed aggregates of connected fractal clusters, with fractal behavior evident over a limited range of length scales [3−6]. Fractal dimensions of 1.7 to at least 1.6 can have been observed in low-density colloidal materials [10].

The smallest structural detail is the so-called “primary” particle, a few nanometers in diameter, consisting of approximately 1000 silica units. Primary secondary (or “secondary”) particles are an order of magnitude larger, and typically somewhat lower in density. Secondary particles, in turn, are connected via interparticle bridges, and form the observed porous structure that may range from a “non-bridgeable” network, to a denser, more highly coordinated structure. It has been reported that gels having the same density may nevertheless exhibit differing properties.

STRUCTURAL MODEL
Our structural model consists only secondary particles, which are assumed to be homogeneous; their detailed structure and any internal degrees of freedom are ignored. These particles are united by very low-density, linear chains of atoms, with an average density of 2 g/cm

Model gel structures are produced by DLCA computer simulation [11].

• 5000 single-particle clusters (primary+secondary particles) are distributed at random without overlap in a computational cell whose size determines the density. Cluster aggregation proceeds via repeated collisions of primary particles.

• A cluster to be moved is chosen at random with probability \( P = 0.1 \) in three-dimensional space. If the move is successful, the cluster and the closest cluster in the cell, respectively, is a scaling variable. The closer cluster is moved to the center of the cell, respectively, and is calculated. The closer cluster is moved to the center of the cell, respectively, and is allowed to relax.

The simulation terminates when only a single cluster remains.

The following DLCA parameters are varied in an attempt to determine the sensitivity of fractal dimension, coordination, and strain behavior on simulation conditions.

• Cluster diffusion step size (2, 3, 5, 10, and 15 nm)
• Particle size distribution method and width (uniform size, log normal, or truncated Gaussian distributions with widths \( \sigma = 0.4, 0.75, 1.0 \) and 1.5)
• Initial relative porosity (0.20, 0.40, 0.60)
• Number of clusters in the initial distribution (1000, 2000, 4000, 8000)

These parameters are varied in an attempt to determine the sensitivity of fractal dimension, coordination, and strain behavior on simulation conditions.

STRUCTURAL MODEL RESULTS—FRAC TAL DIMENSION
DLCA simulations were performed for a range of densities (0.0377, 0.0651, 0.127, and 0.302 g/cm

The fractal dimension of secondary particles is determined using simulation parameters that are shown in figure 3 (a) and (b). The density of clusters (\( \rho_g \)) is an important factor in determining the fractal dimension. The fractal dimension of the clusters is determined by the following equation:

\[ D_f = \frac{\log N}{\log \alpha} \]

where \( D_f \) is the fractal dimension, \( N \) is the number of clusters, and \( \alpha \) is the mass scaling exponent.

Because we do not expect fracture behavior to be correct in detail due to the absence of the angular term, we instead consider the fractal dimension as the most important parameter in determining the strain behavior. The fractal dimension is determined using the following equation:

\[ D_f = \frac{\log \gamma_{\text{in}}}{\log \gamma_{\text{out}}} \]

where \( D_f \) is the fractal dimension, \( \gamma_{\text{in}} \) is the tensile strain at the surface, and \( \gamma_{\text{out}} \) is the tensile strain at the core of the gel. The fractal dimension of the clusters is determined by the following equation:

\[ D_f = \frac{\log \gamma_{\text{in}}}{\log \gamma_{\text{out}}} \]

where \( D_f \) is the fractal dimension, \( \gamma_{\text{in}} \) is the tensile strain at the surface, and \( \gamma_{\text{out}} \) is the tensile strain at the core of the gel.

CONCLUSIONS
We have performed a computer simulations of the tensile strain and failure of silica aerogels, based on a multimodal model for cluster interactions, and a diffusion-limited cluster aggregation model for the gel structure. The simulations were carried out at a level of density equal to that of the aerogel−alcohol composites. We find that the DLCA algorithm is sufficiently robust with respect to variation in simulation parameters. Over the range of parameters studied, only changes in density causes significant differences in cluster fractal dimension and aerogel. Therefore, we do not have a model that successfully explains differences in properties among clusters having the same density. It has been suggested that single DLCA clusters exhibit a stiff wall structure that is too brittle, and that these are an important structural feature that is lost in the aerogel−alcohol composite model. Because we do not expect fracture behavior to be correct in detail due to the absence of the angular term, we instead consider the fractal dimension as the most important parameter in determining the strain behavior.

REFERENCES