Final Report

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Highly Insulating Windows with a U-value less than 0.6 W/m²K

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List of Acronyms

BET	Brunauer, Emmett, and Teller – surface area of materials by gas adsorption
BTMSPA	Bis [3-trimethoxysilylpropyl] amine
BTU	British thermal unit
CR	Condensation resistance
EG	Ethylene glycol
FTIR	Fourier transform infrared
GC	Gas chromatography
GLY	Glycerol
HCl	Hydrochloric acid
HMDS	Hexamethyldisilazane
IGU	Insulated glazing unit
LBNL	Lawrence Berkeley National Laboratory
NREL	National Renewable Energy Laboratory
NFRC	National Fenestration Research Council
NH ₄ OH	Ammonium hydroxide
Ormosils	organically-modified silica aerogels
PDMOS	Polydimethoxysiloxane
SHGC	Solar heat gain coefficient
TC	Thermal conductivity
TEOS	Tetraethylorthosilicate
TMOS	Tetramethylorthosilicate
TMS-OEt	Trimethylethoxysilane
TMS-OH	Trimethylsilanol
TMSPM	Trimethoxysilylpropyl methacrylate
TR	Transparency ratio
UV-VIS-NIR	Ultraviolet-visible-near infrared
VT	Visible transmittance
W	Watt
Tsol	Solar transmittance
Rsol1	Solar Reflectance –exterior
Rsol2	Solar Reflectance –interior
Tvis1	Visible light transmittance - exterior
Tvis2	Visible light transmittance - interior
Rvis1	Visible light reflectance – exterior
Rvis2	Visible light reflectance – interior
Tir	Infrared light transmittance
E1	Infrared emittance – exterior
E2	Infrared emittance – interior
k-eff	Effective thermal conductivity

ABSTRACT

U.S. households rely primarily on three sources of energy: natural gas, electricity, and fuel oil. In the past several decades, electricity consumption by households has grown dramatically, and a significant portion of electricity used in homes is for lighting. Lighting includes both indoor and outdoor lighting and is found in virtually every household in the United States. In 2001, according to the US Energy Information Administration, lighting accounted for 101 billion kWh (8.8 percent) of U.S. household electricity use. Incandescent lamps, which are commonly found in households, are highly inefficient sources of light because about 90 percent of the energy used is lost as heat. For that reason, lighting has been one focus area to increase the efficiency of household electricity consumption.

Windows have several functions, and one of the main functions is to provide a view to the outside. Daylighting is another one of windows' main functions and determines the distribution of daylight to a space. Daylighting windows do not need to be transparent, and a translucent daylighting window is sufficient, and often desired, to diffuse the light and make the space more environmentally pleasing. In homes, skylights are one source of daylighting, but skylights are not very energy efficient and are inseparably linked to solar heat gain. In some climates, added solar heat gains from daylighting may be welcome; but in other climates, heat gain must be controlled. More energy efficient skylights and daylighting solutions, in general, are desired and can be designed by insulating them with aerogels.

Aerogels are a highly insulating and transparent material in its pure form. The overall objective for this project was to prepare an economical, translucent, fiber-reinforced aerogel insulation material for daylighting applications that is durable for manufacturing purposes. This advanced insulation material will increase the thermal performance of daylighting windows, while satisfying constraints such as durability, cost, user acceptance, size limits, and environmental safety concerns. The energy efficient daylighting window will consist of a translucent and resilient aerogel panel sandwiched between glass panes in double glazed windows. Compared to the best windows available today, the double glazed translucent windows with 1/2" aerogel inserts will have a U-value of 1.2 W/m² K (0.211 BTU/ft² h °F) without any coating or low conductivity fill gases. These windows will be more effective than the windows with an *Energy Star* rating of U-2 W/m² K and could be made even more efficient by using low-e coated glass glazings and inert gas fills.

This report summarizes the work accomplished on Cooperative Agreement DE-FC26-03NT41950. During this project, Aspen Aerogels made transparent and translucent aerogels from TMOS and TEOS. We characterized the transparency of the aerogels, reinforced the transparent aerogels with fibers and prepared large translucent aerogel panels and blankets. We also conducted an initial market study for energy efficient translucent windows. A lab-scale process was developed that could be scaled-up to manufacture blankets of these translucent aerogels. The large blankets prepared were used to fabricate prototype translucent windows and skylights.

1. Executive Summary

The primary goal of this project was to develop transparent, resilient, hydrophobic silica aerogels that have low thermal conductivities ($R=10/\text{inch}$) to be used to produce aerogel insulated double-glazing windows with a U value of $0.6 \text{ W/m}^2\text{K}$. To meet this objective we developed a process and equipment to produce blankets of translucent, hydrophobic aerogel. We focused on silica, organically-modified silica aerogels (Ormosils), and fiber reinforced silica aerogels due to the appreciable expertise in silica sol-gel processing available with the personnel at Aspen Aerogels, and also due to the quantity of knowledge available in the scientific literature. The project was conducted in three budget periods, herein called BP1, BP2 and BP3.

Work began on Budget Period 1 for Contract No DE-FC26-03NT41950 in October, 2003, on Budget Period 2 in March, 2006, and on Budget Period 3 in August, 2007. The original scope of the effort was to develop transparent aerogels for windows, but the scope was revised to take into account comments received during the Department of Energy (DOE) Peer Review meeting held in May, 2005. The overall objective for the remainder of this project was to prepare translucent aerogel insulation materials for daylighting, roof windows, and/or skylights. The advanced insulation materials developed will increase the thermal performance, while satisfying constraints such as durability, cost, user acceptance, size limits, and environmental, safety, and health concerns for daylighting, roof windows, and/or skylights.

During BP1, Aspen Aerogels set out to optimize silica aerogel formulations, building upon previous formulation technologies developed at Aspen during a previous DOE contract, DE-FC26-00NT40998. The success criteria were established by the Department of Energy (National Energy Technology Laboratory) to include 85-90%/cm visible light transmittance while maintaining durable hydrophobicity. In order to make hydrophobic aerogels during BP1, we investigated treating the aerogels with additives. To maximize the transparency and monolithicity we used specific precursors and used drying additives to improve monolithicity.

During BP2, the project plan was modified, as mentioned above, to focus on translucent aerogels. Aspen defined, reviewed, and determined the constraints and requirements for the translucent aerogel concepts. The goals for thermal performance versus thickness and density, detailed environmental condition parameters, vacuum containment, flexibility criteria, acceptable thermal and mechanical testing methods, potential costs of production and secondary applications for the most promising insulation candidates were determined. Aspen Aerogels conducted a preliminary market study, which outlined the market for an aerogel based daylighting solution, and computer simulations of the performance of a window containing aerogel insulation.

During BP2 we began investigating using a specific precursor and reinforced these aerogels with fibers to lower the cost of manufacturing translucent aerogels. The effects of target density and component ratio on transparency, final density, and thermal conductivity were investigated. The results were encouraging and monolithic aerogels were prepared that had excellent transparency. Polyester and glass fiber reinforcements were identified that could be used to prepare a fiber reinforced aerogel blanket that allowed 70% transmittance of visible light. Prototype gel-forming and scale-up strategies were investigated at the laboratory level to select the best

approaches for making large blankets. A pre-formed roll process was investigated which gave promising results.

During BP3 we continued to improve the preformed roll process for producing aerogel blankets. The process was scaled-up systematically to eventually produce blankets that were 2 ft wide by 11 ft long from both polyester and glass fiber reinforcements. Samples from these blankets were sent to DOE laboratories to test transmittance and UV stability. NREL tested the UV stability of the hydrophobic aerogel by exposing the samples to UV radiation for 3874 hours. After UV exposure, the hydrophobicity decreased and the samples turned slightly yellow. These tests indicate that a better hydrophobicity reagent will be required to make aerogels that will have the necessary lifetimes for use as insulating windows. Blankets were also sent to window manufacturers who used the blankets to make prototype fenestration products.

To make superinsulating, translucent insulated glazing units (IGUs) a reality, Aspen Aerogels successfully developed a low cost method for producing fiber reinforced translucent aerogels with improved mechanical properties. Aspen also developed a process that could be scaled up to make production quantities to manufacture translucent aerogels for the production of superinsulated windows.

2. Background

The United States consumes roughly 97 quadrillion BTUs (quads) of primary energy per year. The Nation's 87 million homes and commercial buildings consume 36%, or 34.2 quads, of this total. Buildings also use two-thirds of all electricity generated nationally. More than \$230 billion is spent each year in the U.S. to provide heating, cooling, lighting and related energy services for buildings. Even if the energy intensity of buildings remains constant, as more buildings are constructed, energy consumption and associated economic and environmental costs will continue to escalate. Energy consumption in buildings is a major cause of acid rain, smog, and greenhouse gas emissions in the United States, representing 35% of carbon dioxide emissions, 48% of sulfur dioxide emissions, and 21% of nitrogen oxide emissions.

U.S. households rely primarily on three sources of energy: natural gas, electricity, and fuel oil. In the past several decades, electricity consumption by households has grown dramatically, and a significant portion of electricity used in homes is for lighting. Lighting includes both indoor and outdoor lighting and is found in virtually every household in the United States. In 2001, according to the US Energy Information Administration,¹ lighting accounted for 101 billion kWh (8.8 percent) of U.S. household electricity use. Incandescent lamps, which are commonly found in households, are highly inefficient sources of light because about 90 percent of the energy used is lost as heat. For that reason, lighting has been one focus area to increase the efficiency of household electricity consumption.

One of the main functions of windows is to provide a view to the outside. Daylighting is another one of windows' main functions and determines the distribution of daylight to a space. Daylighting windows do not need to be transparent, and a translucent daylighting window is sufficient, and often desired, to diffuse the light and make the space more environmentally pleasing. In homes, skylights are one source of daylighting, but skylights are not very energy efficient and are inseparably linked to solar heat gain. In some climates, added solar heat gains

from daylighting may be welcome; but in other climates, heat gain must be controlled. More energy efficient skylights and daylighting solutions, in general, are desired.

Technologies to reduce energy loss or amount of unwanted solar energy allowed to pass through windows were developed and commercialized in the late 1970s. Market adoption of these innovations has already led to halving the energy requirements of new windows sold today compared to those sold in the 1970s. To reduce energy losses further and make zero energy buildings a reality, superinsulating windows are needed.

Superinsulating fenestration products with U-values below $0.6 \text{ W/m}^2 \text{ K}$ ($0.106 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$) can be constructed in several ways. Most common is triple or quadruple glazed systems using coatings with low emissivity and noble gas filling of the gap between the transparent glazings. However, the price paid to obtain the very low U-values will be a low transmission of solar energy and, to a lesser extent, daylight, both of which will have a negative impact on the total energy balance of the window, especially in heating dominated climates. By applying more advanced coatings it may be possible to obtain overall U-values of about $1.0 \text{ W/m}^2 \text{ K}$ ($0.176 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$), but again the cost for this improvement will be a decrease in the solar gain. So far it has not been possible to construct a glazing that has both a very high thermal resistance and a high transmittance of light. Therefore, to improve energy efficiency of windows further, new approaches are needed.

The ideal material needed to make a highly insulated skylight would be one that has a high transmittance of visible radiation, a low transmittance of infrared radiation, and a high R-factor. If such materials were used to make superinsulated daylighting windows with U values less than $0.6 \text{ W/m}^2 \cdot \text{K}$ ($0.106 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$), then buildings would be highly energy efficient and electricity consumption for lighting would decrease. The magnitude of energy savings would be very significant if these superinsulated daylighting fenestration products were widely available and used in new and existing residential and commercial buildings.

Silica aerogels have the desired transparency and insulating capabilities to be a candidate for use in super insulated glazing units (IGUs). Aerogels have thermal insulation values between R-10 and R-15/inch (R-12 on the average) and a transparency to visible light of ~90% or greater. A combination of an aerogel insulated window with spectrally selective coatings, low-e coatings or electrochromic coatings could be fabricated to make ultra efficient windows with U values $< 0.6 \text{ W/m}^2 \text{ K}$ ($0.106 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$).

The energy efficient superinsulating fenestration products could consist of a translucent, fiber reinforced resilient aerogel sheet sandwiched between glass panes, as illustrated in Figure 1. Compared to the best windows available today, the double glazing windows with a 1/2" aerogel insert will have a U-value of $1.2 \text{ W/m}^2 \text{ K}$ ($0.211 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$) and with 1" aerogel inserts will have U-values of less than $0.6 \text{ W/m}^2 \text{ K}$ ($0.106 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$) without any coating or low conductivity fill gases. These windows will be more than twice as effective as the *Energy Star* rating of $U-2 \text{ W/m}^2 \text{ K}$ ($0.35 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$). With an argon gas fill, the overall U-value could be reduced to as low as $0.5 \text{ W/m}^2 \text{ K}$ for an aerogel sheet sandwiched between double glazing windows.

Figure 1. Aerogel insulated double glazed fenestration product.

To meet the project objectives, Aspen Aerogels identified a low-cost precursor and developed a manufacturing method to reduce the cost of translucent aerogels. To improve the properties of translucent aerogels, the approach was to optimize the optical properties of the aerogels and then reinforce these transparent aerogels with low-density, lofty, fibrous battings.

2.1. Market Study

The market study conducted for this effort included research and analysis to understand manufacturing and process constraints, the market potential, and a value proposition for a translucent fenestration product that uses aerogel material as a thermal insulator.

Aspen Aerogels worked with Tim Adams of GTP Associates (Midland, MI) to conduct the initial market study, which focused on replacement of transparent windows with translucent solutions. Additional market evaluation has been performed to evaluate the marketplace for utilization of aerogel materials in daylighting solutions. The results of the study are discussed in the next section to generate a picture of the overall market for translucent aerogel insulated fenestration products.

2.1.1. Purpose and Scope

The purpose of this market study was to gain a better understanding of the market needs, requirements, and specifications, for improved insulation for super-insulating skylights and daylight fenestrations, quantify the need, and estimate the value added by aerogels. This study was approached as described below:

- Segment and characterize the “daylight fenestration” market; quantify performance and other requirements for each segment.
- Identify the requirements of insulated fenestration products for acceptance in the marketplace.
- Identify and quantify the important aerogel properties such as weight, size (thickness, length, and width), strength, transparency and degree of light scattering, and thermal conductivity (U-Value) needed to satisfy identified performance criteria and markets.
- Quantify the value added by the inclusion of aerogel into fenestration products and estimate cost of production and production rate needed to satisfy identified markets.

The desired outcome is a value proposition to create a knowledge-base and provide insight into an optimal aerogel fenestration product that can be introduced into the marketplace. The market study has provided the following:

- A clear assessment of the developments needed to produce marketable aerogel sheets for the translucent fenestration industry.
- The ability to quantify success criteria for aerogel weight, size (thickness, length, and width), strength, transparency and degree of light scattering, thermal conductivity, cost of production and production rate needed to satisfy identified markets.
- The knowledge that assisted DOE and Aspen in determining whether they should continue research to develop an aerogel product for the translucent fenestration market.

The work consisted of internet based secondary market research, supplemented by approximately 30 telephone interviews with experts in glazing technology, energy efficiency, and people in the fenestration industry, as well as an on-site visit to a leading daylighting manufacturing facility to evaluate the needs of the daylighting industry, familiarize Aspen with the manufacturing process, and evaluate the overall feasibility of an aerogel based daylighting solution.

Based on the results of the study, Aspen's aerogel technology should increase market sizes of some particular markets, with skylights being a good example. It is difficult to access growth potential in these areas, because no parallel market growth technology exists. Certainly the opportunity to purchase a skylight that has the same insulating value as a wall or roof will persuade owners/builders to utilize such solutions, but predicting growth potential in this area would be nearly impossible without accomplishing the work set forth in this program, defining performance, lifetime, and cost.

2.1.2. Opportunity Analysis

2.1.2.1. Translucent Fenestration

Translucent fenestration insulation has a variety of functions that most vision windows do not, including the following:

- Privacy
- Security
- Light diffusion
 - Reduce direct light
 - Reduce glare
- Block undesirable views, but allow light
- Prevent visual distractions (schools, businesses)
- Aesthetically pleasing architectural feature

Some of the translucent products made for the above reasons are as follows:

- Daylighting Insulation
 - Sidelights (beside main entry doors often)
 - Stained glass windows
 - Clerestory windows

- Decorative or frosted windows (in front entry doors for example)
- FRP Insulation (glass fiber reinforced insulation, primarily used for siding and roofing)
- Skylights
- Monumental skylights (atria)

Because aerogel materials offer thermal insulation properties *and* the ability to transmit light, aerogel offers value in applications where heat loss is associated with daylight. There are existing markets, such as skylights, where the market is underserved because of unsatisfactory product forms in terms of heat loss, and other markets that are small because of the lack of an effective heat-loss solution, i.e., sidelights and clerestories.

As the cost of energy rises and the science of daylighting matures, the functions of “windows” will be separated for optimization. There are vision windows, but as long as they are energy-negative, they will decline in number and size. Since consumers desire windows, daylighting solutions will increase as the number of vision windows decrease if they are more energy efficient. With the right product and appropriate placement within a building envelope, daylighting insulation has the potential to be energy positive. The sections below describe the benefit that aerogels could have in a number of daylighting applications.

2.1.2.2. Daylighting Panels

Aerogel insulated daylighting products could be used for sidelights, skylights, clerestories and styles yet to be designed and would combine the functions of energy efficient daylighting with any combination of privacy, security, aesthetics, or light diffusion.

The daylighting panel market could be significantly enhanced by utilization of the proper aerogel product form. Design trade-offs are being made between daylighting and energy consumption whose relationships will change by insulating products with aerogels and increasing energy cost. When daylighting is part of the energy strategy in a new building design, every effort is made to keep initial costs to a minimum and payback times as short as possible. Aerogel insulation increases the energy efficiency of these daylighting products and will reduce the building’s energy requirements; furthermore, the rising price of energy results in payback at a faster rate than originally anticipated. So, aerogels look more and more attractive in comparison with other solutions as energy prices increase.

Aerogel granules are already being used by the daylighting industry made by another aerogel producer. This product form has been used by multiple daylighting producers to provide a highly insulating product form (up to R-20). There are, however, several difficulties associated with the use of aerogel granules in daylighting applications. These difficulties include settling, compaction, use in hybrid systems, weight, color variation, thermal settling, dust generation, electrostatic charging, and cost. Aspen’s aerogel blanket offers a superior product for all of these areas with a flexible or semi-rigid aerogel “blanket” solution.

The granular aerogel product currently available for use in translucent fenestration could be replaced with a more viable alternative product form at a reduced cost per insulating value. The granular product available in the market today provides an R value of R-8. An aerogel blanket

solution, as currently produced by Aspen could provide up to an R-12. This is a 50% increase in the insulating value versus the loose fill material. Additionally, a flexible or semi-rigid solution could allow for integration with existing manufacture of daylighting, by integrating an aerogel blanket with the fiber currently used in many daylighting applications, generating a sandwich composite, allowing for the “tuning” of light transmission, insulating value, and cost.

2.1.2.3. Daylighting Panel Applications

Windows in Northern Climates

An aerogel filled daylighting IGU will allow light and solar energy in the form of heat into the structure and then retain that heat presumably better than any other solution. These properties are valued in northern climates. This market requires no blocking of solar heat gain with maximized U-value keeping the gained heat inside the building where it is needed. This application may also be a good place to test monolithic aerogel in clerestory windows to take best advantage of the solar heat gain.

Sidelights in Mixed and Northern Climates

Sidelighting has been popular for a long time, especially in commercial buildings because a floor to ceiling “side” window can allow light to get much deeper into the building. Most of these are vision windows, but many can be converted to translucent, especially in rooms where there are other vision windows, and in Northern Climates.

Residential entry doors are another common application of sidelights. Many are frosted or etched with a decorative pattern to obscure the view for privacy. A high percentage of these can be converted to translucent panels for the improved insulation properties. In the U.S., where U-values are assigned to the whole component, but not the whole building envelope, door manufacturers will improve overall component U-values (the door unit) by improving the U-value of the sidelight(s).

Monumental Skylights

A large and more easily accessed market is available in commercial skylights. Most commercial skylights are built on site for several reasons. Primarily, many are custom designed to meet the requirements of the building. They may also be too large to transport to the building site. Most manufacturers of “monumental” skylights have their own proprietary designs for large area skylights, the likes of which can be seen in an airport concourse or shopping mall for example. The manufacturers often use their designs as a means of differentiation from their competitors.

Most monumental skylights are translucent. Even in Northern colder climates, direct sunlight can have an overheating effect and increase cooling loads. The use of translucent panels diffuses the light and provides some measure of resistance to solar radiation and subsequent heat gain. The degree of solar heat gain control will be higher in cooling dominated climates and lesser in the central and Northern zones. Monumental skylights are also favorable to an aerogel product because the requirement for light transmittance can be lower than for other skylights. Right-sizing the amount of light for the space should be done in the design stage. In many cases, there can be so much translucent ceiling in square feet, that even low transmittance can add up to enough lumens for the buildings’ requirements.

Unit Skylights

Skylights represent an easily penetrable main stream market for several reasons. A skylight is more likely to be acceptable as translucent than a vertical window, because the primary function of a skylight is light, not vision. Currently, approximately 15% of shop built or “Unit” skylights are translucent.² Skylights have terrible thermal properties compared to vertical windows. They have up to 3 times the solar heat gain problems, and U-values are generally not as good as vertical windows. Generally, skylights are in a predominantly horizontal position and take the sun’s energy in a more direct fashion than vertical windows. At the very least, because of their location on the building, skylights have more time of day in direct sunlight than vertical windows.

Clear skylights which allow direct sunlight are almost always aided by blinds or other means to block the sun. Diffuse light is desired, and translucent glazings are better at diffusing light than transparent glazings. Translucent glazings have a significant share of the market with room for growth, especially if they can reduce solar heat gain. Aerogel sandwiched between glazings will offer reduced summer solar heat gain and prevent heat loss in colder northern climates.

There is an opportunity to convert transparent glass skylights to translucent ones. The market study highlighted that glass is the favored glazing material for skylights, particularly tempered and laminated glass. Glass was chosen for its perceived higher quality than plastic, but not necessarily for its transparent properties. Accordingly, there are no translucent glass glazings available, so the decision for glass automatically carried the decision for transparency. Likewise, if a user wanted translucency, the decision for plastic was automatic. Three market segments are enabled or extended by the properties of aerogels including:

- Currently translucent unit skylights which could benefit from better thermal properties
- Transparent unit skylights which could be specified translucent
- Translucent unit skylights where there were none before, but would now be considered because of the energy efficiency of aerogel

Unit skylights are predominantly used in residential applications; unfortunately a breakdown between commercial and residential was not possible. In total, 1,750,000 skylights were sold domestically in 2005,³ with an average of 8 square feet of glazing area (Wallin, AFG),⁴ for a total of approximately 14 million square feet as indicated by the data tabulated in Table 1.

Table 1. North American Unit Skylight Market 2005.

Units Manufactured (1)	Units	Sq. Ft (Clear) (2)	Sq. Ft. (Translucent)
Glass (66%)	1,155,000	9,240,000	0
Plastic (34%)	595,000	2,660,000	2,100,000
Total	1,750,000	11,900,000	2,100,000
Total Sq. Ft.			14,000,000

(1) Data source; Nick Limb, Ducker Research 4-28-06

(2) Square feet of glazing area per skylight = 8 sq. ft. (Fred Wallin, AFG Glass)

2.1.2.4. Market Validation

Gary Curtis, President of West Wall Group, NFRC board member, and former program manager for Energy Star, discussed daylighting panels with Tim Adams (Market Analyzer for this market study for Aspen) in regard to the utility and market potential for translucent, above eye-level, daylighting panels for applications like clerestories and sidelights for daylighting. In particular, the discussion focused on an insulated glass unit with frosted glass, aerogel, light transmittance at ~ 0.40 and a U-value of 0.2, or $R-5 \text{ ft}^2 \text{ h } ^\circ\text{F}/\text{BTU}$.

Mr. Curtis indicated that if the material was really expensive there could still be a market for high end panels that could be a showcase for the technology in special circumstances (such as zero energy homes), but for the mainstream market, Mr. Curtis thought that the cost of aerogel window would have to be in the range of \$25 to \$40 per square foot, depending on insulating value. This pricing estimate has been additionally validated in discussions with one of the premier daylighting manufacturers in the market, Major Industries. He saw a good market for a product as described, and that it would fit well in the latest trend in daylighting; for example, using very low light transmission windows for vision at eye level, then using daylighting windows or panels above them to light the room, “exactly like the Harmony Library.”⁵

The Harmony Library in Fort Collins, Colorado is an example of managing the amount of daylight to that which is needed, and providing the illusion that it is lighter inside the building than outside. The Harmony Library is projected to use about 40% less energy than a comparable new building plus it has the inherent associated daylighting benefits.

With more energy efficient panels available, more will be used, especially in situations where the entire building envelope is considered in the building codes, not just the U-value of the component.⁶ Daylighting specialist George Kruggel of Radiant Design, Inc. confirmed this. “If I could get daylight for free (without the loss of energy), I’d use as much as I could.” This means that energy negative vision glass could be reduced to that which needs to be seen through, and lighting would or could be provided by more efficient daylighting panels.

2.1.3. Aerogel Property Requirements

The requirements for an Aspen Aerogels product offering in translucent fenestrations include the following:

- 1 Physical and mechanical requirements for surviving the manufacture of windows.
- 2 Physical and mechanical properties to survive a typical window or skylight lifespan.
- 3 Functional requirements containing the added aerogel value.

If the market appears attractive to Aspen and a decision is made to invest in a product offering, a more thorough requirements analysis including more depth into quantification of specific property requirements can be done. Hopefully this analysis uncovers any “showstoppers.” Aspen’s product offering for fenestration systems will most likely be an aerogel-fiber reinforced blanket.

Physical and mechanical requirements to make it through the manufacturing environment in an insulated glass manufacturing plant include:

- Aerogel blanket will be required in various lengths and widths to meet the size of the

IGU in which it will be inserted. There are many sizes in the market, but a large portion will be standard sizes, for example, the daylighting manufacturer visited indicated that >90% of the insulation installed in their solution was cut to 1'x 2' panels.

- Aerogel can be supplied in both rolls and in pre-cut, sized panels offering an excellent variety for the IGU manufacturer.
- Aerogel panels must be undamaged by exposure to common sealants used in the manufacture of IGU's such as polyisobutylene and silicone.
- Aerogel panels must be and remain in a desiccated state prior to insertion into an IGU.

Physical and mechanical properties required to survive a typical window or skylight lifespan include:

- Aerogel panels will be expected to be functional throughout the life of the fenestration which is nominally 20 years for windows and 15 years for skylights, although longer lifetimes are desirable. (The shorter lifespan of the skylight is attributed to the data that shows that skylights are typically replaced in conjunction with a roof.) In discussions with daylighting providers, it was indicated that a 25 year guarantee is the "gold standard" and would be desirable.
- The aerogel panel should be stable to radiation and not discolor during its life.
- The aerogel panel should stay in place without settling or "powdering" during its useful life.
- Depending on methods of attachment in various IGU's, the thickness of the panel will have specifications to allow for thermal expansion.
- The aerogel panel will be subjected to thermal expansion within the normal temperature range, between -50 and +150 °F. The panel must survive 20 years of thermal cycling through that temperature range without crumbling or losing its functional properties.

Functional requirements which contain the aerogel value include:

- A width of 4 feet will be large enough to supply all but very large custom units. Rarely does any fenestration exceed 4 feet in width. Ability for manufacturer to cut panel to desired width could be beneficial for custom products. Aspen's current roll-to-roll process allows manufacture of blankets up to 57" in width.
- A range of thicknesses may be needed to stratify the market in terms of cost and performance; however stacking layers is acceptable.
- The thermal performance of the entire finished panel should have a U-value of at least 0.2 or R-5, with a range of products incrementally spaced in U-value up to the maximum insulation value achievable (at least $U = 0.1$ or R-10) while keeping the visible transmittance (VT) value greater than 0.35.
- The visible light transmittance for a finished aerogel panel should be approximately 0.35 or 35%. The product offering should be a palette of VT values in spaced increments up to the maximum VT achievable.
- The finished aerogel insulated window must offer a range of Solar Heat Gain resistances. Combining a low-e or an electrochromic coated glass with the aerogel insulation could control the SHGC of the fenestration system.
- Thickness may be dependent on the width of space in the IGU, but more importantly, the thickness of the aerogel panel will be dependent on the specific thermal properties required.

Customer Acceptance:

- Uniform light distribution – no visible pattern that would annoy consumers.

A summation of some of the key properties that aerogel based fenestration solutions may be measured against are listed in Table 2. This list of properties was provided to daylighting manufacturers to determine the properties required for an aerogel fenestration solution. The key findings and results are summarized in the section below.

Table 2. Target properties of aerogels for fenestration products.

Property	Desired Targets	Property	Desired Targets
		Ridged Monolith	Composite
Transparency (monolith)	>90%	90%	
Transparency (composite)	>20% /2.5 inches thick		~17%
Compression Modulus	Application Dependent	30-50 psi	50-1000 psi
Flexural Strength	Application Dependent	2-8 psi	unknown
Flexural Modulus	Application Dependent	unknown	unknown
Shear Strength	Application Dependent	unknown	unknown
Vibration Resistance	Same as glass	unknown	unknown
Impact Strength	NA	unknown	unknown
Coefficient of Thermal Expansion (CTE)	Compatible with window	unknown	35×10^{-6}
Effect of thermal cycling	none	Cracks develop	unknown
Hydrophobicity	yes	yes	yes
Thickness	0.25 to 2.5 inches	<0.5-1 inch	<0.25 – 1 inch
Width	variable	Max = 3 ft	5 ft
Height	variable	Max = 8 ft	variable
Cut-able	yes	no	yes
Durability	>20 year lifetime	unknown	unknown
Manufacturing Cost	<\$25-40/ft ² for R10	\$50-100/ft ² for R10	<\$15/ft ² for R10 (expected)

2.1.3.1. Key Findings and Results

In summary, the following key findings have resulted from Aspen's investigation into the translucent fenestration market:

- Energy is expensive and will only continue to increase in cost.
- There is a global trend toward energy efficiency and sustainability in building design, which is fueled by energy prices and the desire for a cleaner environment.
- Private citizens, public and private companies, special interest groups, industry consortia, and governments of all sizes are vocal and supportive of energy saving efforts.
- Green building trends are highlighting the need for energy efficiency and causing a renewed interest in daylighting technology to reduce the need for non-renewable energy.
- Daylighting saves both the electricity to run electric lights and the cost of cooling load caused by all the lights. Windows allow solar heat into buildings. Lighting electricity and solar heat gain through windows account for approximately 74% of the cooling load in a typical commercial building.

- The proper use of daylighting techniques and modern window technology can reduce energy costs easily by 40% and possibly up to 90% compared to similar buildings without daylighting designs.
- Minimizing the heating effect the sun has on buildings also saves energy. Fenestration products need to include approaches such as low-e coatings to minimize or control solar heat gain.
- Since aerogels scatter light and do not currently meet requirements for transparent window acceptance; the market for aerogel insulated fenestration products should focus on non-vision applications.
- Aerogel can be used in translucent fenestrations which allow light to enter the space but control both the heat gain and loss depending on need.
- Market requirements for an aerogel product offering are not beyond the scope of existing technology and should present no prohibitive barriers in final product development efforts for this marketplace.
- A flexible or semi-rigid solution offers the easiest entry into the daylighting market, especially when hybridized with existing insulating materials.

2.1.3.2. Value Propositions

A translucent panel insulated with aerogel can bring daylight into a building more effectively and at a lower cost than traditional windows. It does so by diffusing the light over a wider area, eliminating glare, and allowing the correct amount and light-intensity in the lighted space. For example, in Northern climates, translucent aerogel fenestration products can allow the warmth of the sun in during the day and prevent its escape at night.

Aerogels have properties that allow the architect, the daylighting specialist, and the fenestration designer the freedom to be creative in discovering how aerogels can be used to decrease buildings' reliance on non-renewable energy sources, while delighting the occupants with greater physical and psychological comfort at the lower, stable cost of sunlight.

2.1.3.3. Recommendations

The size of the identified available market could range between 123 and 371 million square feet of available glazing in North America. Capturing that available market depends on many factors including energy economics (which should always be favorable for aerogel) and the ability of a final product to pass light at an acceptable rate which has been identified at or above 0.35 VT.

Based on these results, there is an obvious need to develop and market translucent daylighting products for this market. There is a decent sized early entry niche market, a larger available market, and a short, responsive path to market.

2.2. Simulation of Aerogel Insulated Window's Thermal Performance

2.2.1. Introduction

Aspen Aerogels used WINDOW 5 and THERM 5 to simulate the performance of aerogel insulated fenestration products, developing models predicting the performance of fenestration products integrating aerogel materials for additional insulating value. To validate the results generated in the modeling exercise, Lawrence Berkeley National Laboratory (LBNL) has been

contacted to evaluate the validity of the assumptions utilized to generate the window model. A general description of the system being evaluated, as well as the general assumptions being made, was included in the information sent to LBNL.

The *NFRC Simulation Manual* states that the program calculates the overall energy flow through a fenestration product as a function of:⁷

- **Temperature Driven Heat Transfer:** When there is a temperature difference between inside and outside, heat is lost or gained through the fenestration product frame and glazing by the combined effects of conduction, convection, and radiation. This is indicated in terms of the U-factor of a fenestration assembly.
- **Solar Gain:** Regardless of outside temperature, heat can be gained through fenestration products by direct or indirect solar radiation. The amount of heat gain through products is measured in terms of the solar heat gain coefficient (SHGC) of the glazing.
- **Infiltration:** Heat loss and gain also occur by infiltration through cracks in the fenestration assembly. This effect is measured in terms of the amount of air (cubic feet or meters per minute) that passes through a unit area of fenestration product (square foot or meter) under given pressure conditions.

The *2001 ASHRAE Handbook of Fundamentals* contains the following equation for calculating the energy flow through a fenestration product (assuming no humidity difference):

$$q = U_t \times A_{pf} \times (t_{out} - t_{in}) + (SHGC_t \times A_{pf} \times E_t) \quad (1)$$

Where:

q = instantaneous energy flow, W (Btu/h)

U_t = overall coefficient of heat transfer (U-factor), W/m²-°K (Btu/h-ft²-°F)

t_{in} = interior air temperature, °C (°F)

t_{out} = exterior air temperature, °C (°F)

A_{pf} = Total projected area of fenestration, m² (ft²)

$SHGC_t$ = overall solar heat gain coefficient, non-dimensional

E_t = incident total irradiance, W/m²-°K (Btu/h-ft²-°F)

This equation shows that the properties of U-factor and SHGC are major factors which determine the energy flow through a fenestration product. The NFRC also rates fenestration products for air infiltration and the *NFRC 100*, *NFRC 200*, and *NFRC 400* documents define the procedures for calculating U-factor, SHGC and air infiltration for the total product.

2.2.2. Temperature Driven Heat Transfer

Fenestration product heat loss/gain due to temperature is a combination of three modes of heat transfer:

1. Conduction through glazing, frame elements, and air or other gases in the glazing gap
2. Convection through air layers on the exterior and interior fenestration product surfaces and between glazing layers

3. Radiative heat transfer between glazing layers, or between IG units and interior or exterior spaces.

Solar radiation absorbed by glazing layers will influence these processes, while solar radiation transmitted through the glazing system serves to add heat to the space. The modes of heat transfer are shown schematically in Figure 2. Heat flows from warmer to cooler bodies, thus from inside to outside in winter, and reverse directions in summer during periods when the outside temperature is greater than indoors.

Figure 2. Mechanisms of heat transfer.

The amount of heat transfer due to these three processes is quantified by its U-factor ($\text{W/m}^2\text{-}^\circ\text{C}$ or $\text{Btu/h-ft}^2\text{-}^\circ\text{F}$). The inverse of heat flow, or resistance to heat transfer, is expressed as an R-value. NFRC's rating system quantifies and predicts U-factors.

2.2.3. U-factor

The U-factor (also referred to as U-value) is the standard way to quantify insulating value. It indicates the rate of heat flow through the fenestration product. The U-factor is the total heat transfer coefficient of the fenestration system, in $\text{W/m}^2\text{-}^\circ\text{C}$ ($\text{Btu/hr-ft}^2\text{-}^\circ\text{F}$), which includes conductive, convective, and radiative heat transfer for a given set of environmental conditions. It therefore represents the heat flow per hour, in Watts (Btu per hour) through each square meter (square foot) of fenestration product for a $1\text{ }^\circ\text{C}$ ($1\text{ }^\circ\text{F}$) temperature difference between the indoor and outdoor air temperature: the smaller the U-factor of a material, the lower the rate of heat flow. The total R-value, which measures thermal resistance, is the reciprocal of the total U factor ($R=1/U$).

The U-factor depends on the thermal properties of the materials in the fenestration product assembly, as well as the weather conditions, such as the temperature differential between indoors and outside, and wind speed. NFRC has standardized the exterior conditions (called environmental conditions) for U-factor calculations for product ratings using the following temperatures and wind speeds:

- Wind Speed: 5.5 km/hr (12.3 mph)
- Indoor air temperature: 21 °C (70 °F)
- Outdoor air temperature: -18 °C (0 °F)

Skylights and roof windows are simulated at a 20 degree slope from the horizontal.

2.2.4. Solar Heat Gain Coefficient (SHGC)

The total solar heat gain coefficient is determined by an area-weighted average of contributions from the transparent and the opaque elements in the fenestration product. The SHGC is a function of the solar transmittance, the absorbed solar radiation of each layer and the inward conduction of thermal energy. The SHGC is calculated for each component of the product separately.

2.2.5. WINDOW Computational Method

Heat transfer across a fenestration product is a function of both the temperature difference between the inside and outside and the incident solar radiation on the product. In order to evaluate heat transfer through a specific product, its configuration and physical dimensions must be specified. This includes the glazing properties (visible, total solar and infrared optical properties, and thermal conductivity), the cavity fill (air, low-conductivity gas, vacuum, aerogel) thermophysical properties, spacer and frame characteristics, and environmental conditions.

Fenestration product heat transfer through the center-of-glazing area is primarily a one-dimensional process. It is analyzed by breaking down the glazing system cross section into an assembly of nodes and calculating the heat transfer between each node. Under steady-state conditions, the temperatures of the nodes are such that the net energy flux entering each node is equal to that leaving each node. To perform the energy balance, WINDOW models the user-defined glazing system as a one-dimensional, steady-state resistance network, shown in Figure 3. An iterative solution method is then used to converge upon the correct temperature distribution. From this temperature distribution, any desired performance index can be calculated.⁷

Figure 3. Resistance network used to model center-of-glazing heat transfer in WINDOW.⁷

Two temperature nodes are assigned to each glazing layer (exterior and interior surface), along with outside air and inside air temperature nodes. The resistance between each node equals the inverse of the sum of the radiative and conductive/convective heat transfer coefficients. The temperature-dependent conduction/convection and effective-radiation heat transfer coefficients for the outward-facing and inward-facing surfaces and for the gas-filled gaps are calculated from the temperature distribution. The heat transfer coefficients between the nodes within the solid materials simply depend on the conductivity of the materials.

Conductive/convective heat transfer coefficients are calculated based on empirical relationships. The outside film coefficient depends on the windspeed and the direction from which the wind is blowing. The inside film coefficient is a function of the difference between the inward-facing surface temperature, the inside temperature, and the height of the fenestration product. Gap heat transfer coefficients are computed from empirical equations for the Nusselt number. The Nusselt number relates the temperature difference between the surfaces bounding the gap, the width, the height and the thermophysical properties of the gap. Fenestration product tilt is also accounted for in all conductive/convective correlations.

The radiative energy flux leaving each surface is calculated from the Stephan-Boltzmann law using the surface infrared hemispherical emissivity and temperature. The net radiative flux between radiating nodes divided by the associated temperature difference gives an effective radiation heat transfer coefficient.

To model glazing systems with multiple spectrally selective glazings (i.e., glazings with solar/optical properties varying by wavelength, such as many low-emissivity coatings), a multi-band model is used in WINDOW. In this model, WINDOW calculates the transmittance and reflectance for the glazing layer or the glazing system wavelength by wavelength, and then weights the properties by the appropriate weighting functions to obtain the total solar, visible, thermal infrared properties, as well as the damage weighted transmittance and the transmittance between 0.30 and 0.38 microns. To use the multi-band model, WINDOW needs a spectral data file for each glazing layer. These data files are updated and maintained by LBNL and available from NFRC. If some of the glazing layers in a glazing system do not have a spectral data file, WINDOW assumes a flat spectral behavior of the glazings, based on their stated visible and solar properties. For NFRC certification simulation, the NFRC-approved spectral data files must be used (indicated by a # symbol in the WINDOW Glass Library).

2.2.5.1. Total Product Calculations

The total fenestration product properties for U-factor, SHGC and VT are based on an area-weighted average of the product's component properties which are:

- center-of-glazing
- frame
- edge-of-glazing
- divider
- edge-of-divider

The frame and edge-of-divider properties depend on the center-of-glazing properties. This area-weighted total product value can be calculated using the WINDOW program or another calculation tool, such as a spreadsheet. The procedure for this area-weighted calculation is:

1. Multiply the component property by the component area
2. Sum these area-weighted component properties
3. Divide the area-weighted sum by the total projected area of the product

The operator types (fixed, vertical slider, horizontal slider, casement) determine which components (head, jamb, sill and meeting rail) are required to calculate the whole product area-weighted values.

2.2.5.2. Condensation Resistance (CR)

The whole-product Condensation Resistance (CR) calculation is implemented in WINDOW according to *NFRC 500: Procedure for Determining Fenestration Product Condensation Resistance Values*.

2.2.6. Results of Simulations

The first strategy to model an aerogel filled window was to define the properties of a fill that could be inserted between glazings. Table 3 lists the properties of the gases typically used as fills plus new fills defined for a vacuum and an aerogel. Vacuum was defined as having 0% concentration of any gases and an aerogel was defined as a fill with a thermal conductivity of 14 mW/m•K, a heat capacity of 691 J/kgK, and a density of 116 kg/m³. WINDOW 5 calculates the Prandtl number as a function of the viscosity, the thermal conductivity, and the heat capacity. The viscosity and Prandtl numbers for aerogels are not universally known, but a large Prandtl number indicates that heat diffuses slowly. An aerogel cavity fill is expected to decrease convective heat flow compared to gas fills by reducing or eliminating gas convection because the aerogel's nanosized pores are smaller than the mean free path of gas molecules. This approach is only a first approximation and does not account for the optical properties of aerogels.

Table 3. Properties of cavity fills.

ID	Name	Type	Conductivity W/m K	Viscosity Kg/m s	Cp J/Kg K	Density Kg/m ³	Prandtl
1	Air	Pure	0.024069	0.000017	1006.103	1.292	0.7179
2	Argon	Pure	0.016349	0.000021	521.929	1.781	0.6704
3	Krypton	Pure	0.008664	0.000023	248.091	3.738	0.6717
4	Xenon	Pure	0.005160	0.000021	158.340	5.856	0.6542
5	Vacuum	Mix	0	0	0	0	0
6	Aerogel	Pure	0.014143	0.67	691.07	115.97	32738

Glazing systems were defined in WINDOW5 and input into THERM5 using the standard sample frame supplied with THERM5. A cross section of the frame is shown in Figure 4. The sample frame was made of wood, a silica desiccant, a flexible vinyl/metallic spacer and is sealed with butyl rubber. Standard NFRC 100-2001 exterior boundary conditions were used. Since the main

application is daylighting, the windows were modeled in the vertical position. The overall thickness of the glazing was 25.92mm. The windows in Figure 5, Figure 6, and Figure 7 use the frame in Figure 4. The windows in Figure 5, Figure 6, and Figure 7 have an air fill, aerogel fill, and a vacuum respectively.

Figure 4. Cross section of frame (Sample-Sill with Sample GlzSys in THERM5).

Figure 5. Double glazed window with air fill and low-e coating on surface 3 ($U = 2.0$ W/m^2K (0.35 $BTU/ft^2 h ^\circ F$) $CR = 49$, $SHGC = 0.602$).

Figure 6. Sample frame with aerogel fill, low-e coating on surface 3 ($U = 1.408 \text{ W/m}^2\text{K}$ ($0.248 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$), $\text{CR} = 57$, $\text{SHGC} = 0.602$).

Figure 7. Sample frame with evacuated glazing, low e coating on surface 3 ($U = 0.936 \text{ W/m}^2\text{K}$ ($0.165 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$), $\text{CR} = 61$, $\text{SHGC} = 0.609$).

To model an aerogel insulated window, the optical properties of a hydrophilic aerogel were input into OPTICS5. Vis-NIR spectral data was collected using an integration sphere to measure the

direct transmitted radiation and the diffuse or scattered transmitted radiation (Figure 8). The data that was input into OPTICS5 for the aerogel was the total transmitted light which is the sum of the diffuse and direct components (Figure 9). The total transmitted radiation was used since OPTICS5 does not include algorithms to model scattered radiation. The total light transmitted was high for this transparent aerogel, and we assigned a low front and back reflectance following the rule that the sum of reflected and transmitted light is less than one for all wavelengths.

Figure 8. Spectrum of a transparent, hydrophilic aerogel between 300 and 2500 nm.

Figure 9. Spectral data used in OPTICS5 to model an aerogel window.

Since the WINDOW5 program has several constraints that must be followed, we used the main components of the sample frame and added an aerogel spacer (warm edge) so we could compare a double glazed window with an aerogel insulated window using clear glass. The frame used is not intended to represent an actual frame design, and its only purpose is to be able to compare a double glazed window with an aerogel insulated window. The cross section of the window is shown in Figure 10 and the temperature profile for this window is shown in Figure 11. The optical properties of the aerogel pane were calculated in OPTICS5 and the data input into WINDOW5 glazing database. The auto import function did not work with a message that said

“could not query “GlassQuery”. Therefore, the data calculated by OPTICS5 was manually input into the WINDOW program.

Figure 10. Cross section of double glazed window with air fill.

Figure 11. Double glazed window with air fill, clear CLEAR5.LOF glass ($U = 2.49 \text{ W/m}^2\text{K}$ ($0.438 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$), $CR = 48$, $SHGC = 0.633$).

The aerogel insulated glazing was defined as a 3 layer (triple glazed) system as shown in Figure 12. The aerogel pane could not be input into the 3 layer window with the aerogel right next the

glass panes since the program required that an air gap separate the layers. We used a 2mm gap between the aerogel and the Clear5.LOF panes. Figure 13 illustrates the results obtained using an air fill in the cavity and Figure 14 illustrates the results with the cavity evacuated.

Figure 12. Cross section of aerogel insulated window.

Figure 13. Aerogel insulated window with CLEAR5.LOF glazing ($U = 0.77 \text{ W/m}^2\text{K}$ ($0.136 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$), $CR = 90$, $SHGC = 0.618$).

Figure 14. Evacuated aerogel insulated window with CLEAR5.LOF glazing ($U = 0.512$ W/m^2K (0.09 BTU/ft² h °F), CR = 96, SHGC = .619).

Modeling an aerogel insulated window with a low-e coating on surface 2 resulted in a window with the same U value but with lower SHGC as shown in Figure 15. IGDB ID#926 CFTIR_5.AFG low-e coated glass was used as the 1st glass pane.

Figure 15. Aerogel insulated window with CMFTIR_5.AFG and CLEAR5.LOF glazing ($U = 0.77$ W/m^2K (0.136 BTU/ft² h °F) , CR = 89, SHGC = 0.365).

When the cavity of this window was evacuated, the U factor for the evacuated aerogel window with the same glazings decreased to 0.25. The cross section of this window is illustrated in Figure 16. The temperature profile for this window was unusual in that the temperature on the surface 3 increased to ~65 °C while it was only ~41 °C with air in the cavity. An evacuated window using the same CMFTIR_5.AFG and CLEAR5.LOF glazings gave a U factor of 0.292, CR = 97 and a SHGC of 0.387. It is interesting that the simulated evacuated aerogel insulated window had better properties than the non-aerogel insulated window. Evidently, the presence of the aerogel absorbs some of the radiation and decreases radiative heat transfer.

Figure 16. Evacuated aerogel insulated window with CMFTIR_5.AFG and CLEAR5.LOF glazing ($U = 0.25 \text{ W/m}^2\text{K}$ ($0.044 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$), $CR = 100$, $SHGC = 0.378$).

In the ideal aerogel insulated window, the aerogel would be inserted between the glass panes and there would be no air gap between the panes and the aerogels; however, the WINDOW5 program would not accept a window with no air gap between the glass and the aerogel. DOE requested that we try to determine the effect of the air gap thickness on the performance of an aerogel insulated window fabricated using low-e glass (CMFTIR_5 manufactured by AFG Industries). Therefore, the aerogel insulated glazing with input as a 3 layer system consisting of a low-e coated glass pane, an aerogel sheet, and a clear glass pane. The gap on each side of the aerogel was filled with air, and the window with a 0.01mm air gap is described in Table 4.

The sample wood frame was used for this window and an aerogel spacer was used for the aerogel insulated window. The center U-value calculated for this aerogel insulated window was $0.731 \text{ W/m}^2\text{K}$ and the U value for the total window was $0.78 \text{ W/m}^2\text{K}$. The solar heat gain coefficient was 0.375 and the VT was 0.559. The inside and outside temperatures of each pane (Layer 1 and Layer 2 and Layer 3) are listed in Table 5.

The U-value for a similar window with a 1.9 mm air gap and a 12.7 mm thick aerogel was $0.77 \text{ W/m}^2\text{K}$. Based on the calculated results from WINDOW5, the U value depended on the total

thickness of the insulated glazing unit, and the thickness of the air gap had little to no effect on the overall performance of the window. The simulated results represent a ‘best case’ scenario and will be used as target values for commercially viable aerogel insulated windows.

Table 4. Description for aerogel insulated glazing system with 0.01 mm air gaps.

ID	Name	D(mm)	Tsol	Rsol1	Rsol2	Tvis1	Rvis1	Rvis2	Tir	E1	E2	k-eff
Outside												
926	CMFTIR_5	4.6	0.471	0.297	0.391	0.767	0.150	0.115	000	0.840	0.035	1
	AIR	0.01										0.023
	AEROGEL	16.4	0.928	0.016	0.016	0.931	0.018	0.018	000	0.900	0.900	0.014
	AIR	0.01										0.025
9803	CLEAR5	4.7	0.796	0.074	0.074	0.888	0.082	0.082	000	0.840	0.840	1
Inside												

Table 5. Temperature distribution (°C) output from WINDOW5 for aerogel insulated window with low-e coating.

(Temperature: Winter – Outside: -18°C, Inside 21°C / Summer – Outside 32°C, Inside 24°C)

	Winter		Summer	
	Outside	Inside	Outside	Inside
Layer 1	-17.0	-16.9	41.1	41.5
Layer 2	16.9	16.5	41.5	31.4
Layer 3	16.6	16.7	31.4	31.2

2.2.6.1. Simulated Daylighting Product

We also simulated the performance of a daylighting product using an aluminum frame which is similar to the product offered by Major Industries. The center U value of the total window was 0.82 W/m²K (0.144 BTU/ft² h °F) and the center U-value for the daylighting product was 0.6 W/m²K (0.106 BTU/ft² h °F). The calculation predicts an R value of 6.9 ft² h °F/BTU. These values are quite good considering the frame is conductive and CR index was 33. With a better frame the performance would improve. The temperature profiles calculated are shown in Figure 17.

2.2.6.2. Thermal Analysis Summary of Simulations

The results are summarized in Table 6 and came out close to those expected. The first approximation indicated that an insulating transparent cavity fill and an evacuated cavity would improve window performance. Adding the optical properties to the model significantly improved the simulated results. An aerogel window is predicted to have a U value of 0.77 W/m²K (0.135 BTU/ft² h °F) with 12.7 mm thick pane of aerogel inserted between two panes of clear glass. When this glazing was evacuated, the U value decreased to 0.51 W/m²K (0.09 BTU/ft² h °F). When pane 1 was changed to glass with a low-e coating, the U value remained the same but the SHGC decreased; evacuating the glazing with the low-e coating, decreased the U value to 0.25 W/m²K (0.044 BTU/ft² h °F). Interestingly, this evacuated aerogel insulated glazing was predicted to have a lower U-value than one without the aerogel inserted between the panes.

Hartman, Rubin and Arasteh have simulated the thermal and optical properties of aerogel windows and calculated that an evacuated aerogel insulated window 12.5 mm thick would have a U-value of $\sim 0.5 \text{ W/m}^2\text{K}$ ($0.088 \text{ BTU/ft}^2 \text{ h } ^\circ\text{F}$).⁸ Aspen's results were in close agreement.

Table 6. Summary of properties of simulated windows.

	Description	Spacer	U factor	SHGC	T_{vis}	CR
1	Double glazed w low-e coating (3) Lite 1 = Clear5.LOF (IGDB = 9803) Air = glazing cavity fill Lite 2 = Low-e.LOF (IGDB = 9923)	metallic	2.001	0.602	0.648	49
2	Double glazed w Aerogel + low-e (3) Lite 1 = Clear5.LOF (IGDB = 9803) Aerogel = glazing cavity fill Lite 2 = Low-e.LOF (IGDB = 9923)	metallic	1.408	0.602	0.645	57
3	Double glazed w vacuum + low-e (3) Lite 1 = Clear5.LOF (IGDB = 9803) Vacuum = glazing cavity fill Lite 2 = Low-e.LOF (IGDB = 9923)	metallic	0.936	0.609	0.645	61
4	Double glazed Lite 1 = Clear5.LOF (IGDB = 9803) Air = glazing cavity fill Lite 2 = Clear5.LOF (IGDB = 9803)	aerogel	2.491	0.633	0.692	48
5	Double glazed w aerogel Lite 1 = Clear5.LOF (IGDB = 9803) Air = glazing cavity fill Lite 2 = aerogel Air = glazing cavity fill Lite 3 = Clear5.LOF (IGDB = 9803)	aerogel	0.776	0.618	0.646	90
6	Double glazed w aerogel + low-e coating (2) Lite 1 = CMFTIR_5.AFG (IGDB = 926) Air = glazing cavity fill Lite 2 = aerogel Air = glazing cavity fill Lite 3 = Clear5.LOF (IGDB = 9803)	aerogel	0.768	0.365	0.518	89
7	Double glazed w aerogel + vacuum Lite 1 = Clear5.LOF (IGDB = 9803) Vacuum = glazing cavity fill Lite 2 = aerogel Vacuum = glazing cavity fill Lite 3 = Clear5.LOF (IGDB = 9803)	aerogel	0.512	0.619	0.646	96
8	Double glazed w aerogel + vacuum + low-e (2) Lite 1 = CMFTIR_5.AFG (IGDB = 926) Vacuum = glazing cavity fill Lite 2 = aerogel Vacuum = glazing cavity fill Lite 3 = Clear5.LOF (IGDB = 9803)	aerogel	0.250	0.378	0.518	100

Figure 17. Aerogel insulated daylighting product with an aluminum frame.

2.3. Synthesis of Silica precursor from Si Powder

At the start of this effort, one of the major long term goals of Aspen Aerogels had been to eventually integrate the synthesis of silica precursors into the aerogel manufacturing. To identify possible synthesis methods, we conducted a patent search on the direct process to produce silicon alkoxides and identified 37 relevant patents. We selected two of these to investigate in more detail. These two patents were: U. Dittrich, F. Steding and R. Mueller “Continuous Method of Manufacturing Chlorine Free Tetraalkoxysilanes” US patent 6,191,296 B1 awarded to Hüls Aktiengesellschaft on Feb. 20, 2001 and W. Magee and J. Telschow, “Method for the Production of Tetraalkylsilicates,” US patent 4,288,604 awarded to Stauffer Chemical Company on Sept. 8, 1981. Both of these patents are similar and use 2-(2-butoxyethoxy)ethanol as a high temperature solvent for the reaction. Therefore, we investigated these two approaches to prepare the precursor from Si and alcohol. The results indicated that it is feasible to prepare tetraalkylsilicates by these approaches, but the lifetime of the catalysts was not very long and better catalysts would further improve these possible methods.

2.4. Development of Transparent/Resilient Aerogels

Previous results have shown that methoxy based silica precursors are the best precursors to use for synthesizing transparent silica aerogels. Therefore, we initially focused our research efforts on investigating and optimizing the conditions for making monolithic transparent silica aerogels from this precursor. The study also concentrated on making perfectly hydrophobic aerogels with good resiliency, transparency, and thermal performance. Nevertheless, improvements in monolithicity and increased sample size have not been easy to achieve.

During BP1 we concentrated our efforts on improving the chemistry and the sol-gel process to fabricate crack-free monolithic aerogels. The cracking of aerogels during supercritical drying

has always been considered as the main obstacle in producing crack-free, transparent aerogels reproducibly. There are several parameters that influence the monolithicity of the aerogel. These parameters include the pore size distribution, density and thickness of the gel, aging conditions, catalyst, nature of the solvent and additives, and supercritical drying conditions. By varying these processing parameters, aerogels with different microstructure and surface chemistry can be obtained, and the monolithicity can be considerably affected. Producing a well-organized silica network with a narrow pore size distribution will significantly improve the yield of monolithic, crack-free aerogels. Certain “drying” additives are known to alter the average pore size and pore size distribution of gels. The addition of these additives to the sol may result in more uniform pore and particle size distributions, thus reducing the differential drying stresses and preserving the monolithicity of the aerogel. Therefore, we studied the effect of these additives on the optical and physical properties of the aerogel. The aerogels produced were perfectly hydrophobic when aged in appropriate solvents.

2.4.1. Results and Discussion of Aerogel Synthesis

The results were very interesting since most of the gels were dried successfully without cracks. As we anticipated, the “drying” additives helped preserve the monolithicity of the gels. The final density of the aerogel varied as a function of the aging treatment and the target density. For samples with a target density 0.06 g/cc, the final density can be as high as 0.22 g/cc. The increased density arises from the silica that deposits in regions between particles (Figure 18). This phenomenon is known as “Ostwald Ripening”. This process increases the size of the particles, producing a stronger gel, but decreases the surface area.⁹

Figure 18. “Ostwald Ripening” phenomenon.

Also, the concentration of the “drying” additive strongly affects the transparency of the aerogels. The higher the concentration, the better the transparency. A specific solvent treatment is used to make hydrophobic aerogels, but at the same time this treatment decreases their transparency. The transparency of aerogels was improved by optimizing the two catalysts used in the sol-gel preparation. During Budget Period 1, we principally:

- Searched for alternative methods to produce hydrophobic aerogels without decreasing the transparency of the aerogel.
- Decreased Rayleigh scattering by optimizing the catalyst and silica ratios to produce highly transparent aerogels.
- Characterized the aerogels by BET analysis and pore size distribution, and determined the relationship between transparency and microstructure (or porosity) of the aerogel.

- Focused on the aging step by using a fluorinated silane to achieve durable and stable hydrophobicity for the aerogels.
- Scaled up the size of the aerogel to 25 cm x 16 cm x 1.3 cm.
- Investigated a cost-effective and less dangerous route to synthesize transparent silica aerogels.

It is interesting to note that some of the samples have low thermal conductivities (~ 11 mW/m-K) and also exhibit high transparency (evaluated visually). For the samples with target densities of 0.06 g/cc, the aging process does not seem to affect the thermal conductivity of the aerogel. However, the influence of target density on the transparency is strongly pronounced. With a 0.1 g/cc target density, samples demonstrated higher thermal conductivities. Based on this result, aerogels with a target density ranging between 0.06 and 0.08 g/cc have the best properties.

2.4.1.1. Optical Properties

In order to perform a quantitative characterization of the light transmittance behavior of a material receiving light or solar radiation, a spectrum must be obtained for the UV-VIS-NIR wavelength range using a spectrophotometer. In the case of aerogels, the expectation is not only high transmittance in this range, but also a glass-like (transparent) appearance. Physically, it means that the diffused part of the transmitted light must be as low as it is through two panes of glass. This is a very severe requirement, and to satisfy it, each improvement in the preparation of the aerogel is thus followed by optical measurements.

Considering the objective of the program, i.e. “transparent silica aerogels for double window glazing”, we planned to perform transmittance characterization on monolithic aerogels using Optical Data Associates facilities. The lab has a Cary 500E UV-VIS-NIR and a Nicolet 560 FTIR spectrometer, both equipped with a Labsphere Spectralon integrating sphere.

This equipment produces spectra of Normal/Hemispherical transmission (direct + diffuse). It is also possible to get the Total Diffuse transmission by subtraction of the direct part of the transmitted radiation as shown in Figure 19. By weighted integration, and for comparison purposes, we evaluated the transparency of the aerogels by determining the Direct-to-Hemispherical ratio, which we call the “transparency ratio” (TR). This parameter is defined in the visible range as:

$$TR = [(\tau_l^{nh} - \tau_l^{ndf}) / \tau_l^{nh}] \times 100 = \tau_l^{nd} / \tau_l^{nh} \times 100 \quad (2)$$

Figure 19. Different transmission spectral components collected by an integrating sphere.

The transmittance quantities are values integrated over the range 380-780 nm using a weighting function. The higher the TR, the better the transparency and clarity of the image as seen through the aerogel. The ratio does not take into account the influence of light absorption: it does not permit separation of the forward scattering contribution from the isotropic contribution. The TR values are between 0 (opaque) and 1 (transparent) and are expressed as a percentage. The values represent the influence of diffuse light on the transparency, and they only represent the necessary conditions (and not the sufficient ones) to be met, in order to obtain good imaging systems.

Rayleigh scattering effects, which depend on the thickness of the sample, strongly affect the transparency of the aerogel. A simple method can be used to quantitatively measure the relative contributions of Rayleigh scattering for silica aerogel prepared with different recipes. The extinction coefficient, E , of an aerogel slab of known thickness is measured for the Normal/Direct transmittance τ_l^{nd} (often, calculated at 550 nm due to the maximum sensitivity of the eye in the visible range): the smaller the value of E , the higher the clarity and transparency of the aerogel. The relation is given by:

$$E = -1/d * \ln (\tau_l^{nd}) \quad (3)$$

Where:

τ_l^{nd} = Normal/Direct transmittance

d = sample thickness.

Three samples were characterized at Optical Data Associates (ODA) and the results are given in Table 7. We selected the samples based on their visual transparency and their aging treatment. Samples 3 EG#3 and 3 EG#4 are the same material but they were aged differently. As indicated in Table 7, the hydrophobic sample (3 EG#4) is less transparent than the hydrophilic sample (3 EG#3) because the hydrophobe increases the silica particle size and affects the pore size distribution. Sample 7 EG#4, which is similar to sample 3 EG#4 but with higher target density (0.1 g/cc), has a lower transparency but has a similar thermal conductivity. It is important to note that the surface of the aerogel strongly affects TR and the extinction coefficient, E . For example, dust particles and irregularities on the surface might decrease the optical properties of the aerogels by 20 %.

Table 7. Optical properties of aerogels.

Samples	Density (g/cc)	Hydrophobic	TC (mW/m-K)	Thickness (cm)	TR (%)	E (m ⁻¹)
3 EG#3	0.083	No	11.2	1.50	85.7	18.93
3 EG#4	0.096	Yes	12.4	1.15	81.6	34.63
7 EG#4	0.18	Yes	13.6	1.35	63.7	83.19

Figure 20, Figure 21, and Figure 22 illustrate the transmission spectra of the three samples described above. In order to have high transparency in the visible domain (380 – 780 nm), it is preferable to have the spectra shifted as much as possible to the UV region, especially the

maxima of the diffusion spectra. Sample 3 EG#3 (Figure 20) shows good transparency up to 1250 nm. It is obvious why sample 7 EG#4 has low transparency (low TR), since the maximum of the diffusion spectrum is located entirely in the visible region (see Figure 22).

In Figure 23 the UV-VIS-NIR spectra of the three samples are compared over the entire wavelength range. The hydrophilic sample (3 EG #3) exhibits sharp absorption bands at 1365 and 1405 nm that are related to Si-OH groups. These bands are significantly weaker in the hydrophobic samples since most of them have reacted with HMDS and have been converted to SiOSiMe₃ groups. The hydrophobic aerogels have absorption bands at 1690 and 1735 nm that are not present in the hydrophilic aerogels, and these bands are probably related to the SiMe₃ groups.

Figure 20. Transmittance spectra of hydrophilic sample 3 EG#3.

Figure 21. Transmittance spectra of hydrophobic sample 3 EG#4.

Figure 22. Transmittance spectra of sample 7 EG#4.

Figure 23. Total direct spectra of the three samples: 3 EG#3, 3 EG#4 and 7 EG#4.

2.4.1.2. Effect of Gelation Process on Rayleigh Scattering

The two-step approach combined with the utilization of a drying additive improves the monolithicity of the gels and decreases the probability that cracks will develop during supercritical drying. However, the two-step approach has been shown to have a disadvantage in terms of transparency of the aerogel. Rayleigh scattering is much more pronounced when specific catalysts are used.

The scattering is undoubtedly governed by the aerogel microstructure, particle size, and pore size of the silica particles forming the aerogel. The catalysts principally control the microstructure of the gel. Therefore, their concentrations were investigated to optimize aerogel transparency. No special aging treatment was performed on these gels; therefore, they are hydrophilic.

With just the right concentrations, the gels exhibited high transparency and clarity after supercritical drying. It is important to note that this study was only designed to determine the influence of the catalysts on the transparency of the aerogels, so the aerogels produced were hydrophilic. The surface area and the pore size of the different samples were evaluated to try to establish a relationship between the transparency and the microstructure of the aerogel. The surface area does not vary too much ($1024 - 1132 \text{ m}^2/\text{g}$). However, the pore volume and pore size definitely depend on the catalysts' ratio and water content.

Also, it's interesting to note that the transparency of the aerogels does not depend on the surface area. For instance, when we compare samples 3 (high transparency) and 4 (low transparency), as shown in Figure 24, we can immediately see that their corresponding surface areas are almost identical. However the pore volume and the average pore diameter of sample 3 are two times larger than those of sample 4.

Figure 24. Two monolith aerogels prepared by two-step catalysis. (a) Sample 3, and (b) sample 4.

Figure 25 clearly shows the pore size distribution of the samples prepared. Samples 4GLY#3 and 3EG#3 possess good transparency, were synthesized by the two-step approach, and used a drying additive.

Some samples have similar pore size distributions and pore volumes. However, when additional water is used the pore structure of the aerogels is undoubtedly affected. The evidence indicates that: the higher the basic catalyst concentration, the higher the pore volume, which produces aerogels with better transparency.

Figure 25. Pore size distribution versus chemistry of the gels.

Comparing 4GLY#3 and 3EG#3 with samples 1-6 indicates that lowering the concentration of acid appears to shift the maximum of the pore size distribution curve to smaller pore sizes (below 10 nm). Still, the average pore diameter for good transparent aerogel is nearly around 16 nm and for translucent aerogel in the range 7 – 10 nm.

The thermal conductivity of samples produced was determined to evaluate their thermal insulation performance. The TCs (Thermal Conductivities) are listed in Table 8, and were lower for the samples prepared with higher water content. There is no obvious relationship with the pore structure or surface area of the material. In this study, the goal was to prepare aerogels with the lowest TC and the highest optical transparency possible.

Table 8. Thermal conductivities of aerogel samples.

Samples	Density (g/cc)	S _{BET} (m ² /g)	TC (mW/m•K)
Sample 1	0.085	-	12.2
Sample 2	0.092	1123	11.9
Sample 3	0.100	1122	12.0
Sample 4	0.089	1132	13.2
Sample 5	0.095	1024	14.3
Sample 6	0.110	1025	13.5

If we look at all the results collected, the formulation used for sample 3 seems optimum to produce high quality transparent silica aerogels.

2.4.2. Hybrid Aerogels

Inorganic-organic hybrid polymers and nanocomposites have been the subject of research by many scientists. Numerous papers have been published, but to our knowledge, no one has succeeded in producing highly transparent/resilient aerogel panels that are better than Aspen's amine/TMOS derived aerogels. Further improvements in strength and transparency are still desired, so we investigated and tested some new hybrid systems, which were expected to lead to an improved resilient aerogel.

We focused on introducing new crosslinking agents, into the silica matrix and then studied the aging conditions and physical state of the gels during supercritical drying. For comparison purposes, in one case a drying additive was not used.

2.4.2.1.1. Results and Discussion

The synthesis of the hybrid gels was affected by fast gelation for systems with a crosslinker/silica precursor molar ratio higher than 0.1. Despite the fast gelation time, the gels were more translucent than transparent. On the other hand, another hybrid system led to a highly transparent hybrid gel, but these gels didn't shrink enough to be easily removed from the mold for drying with CO₂. Some of the gels are more rubbery and elastic (and stick to the mold). Some pieces of these gels (we had to break the gels to take them out of the mold) were dried at supercritical conditions of CO₂, and it was difficult to evaluate the quality (transparency and resiliency) of the aerogels because of the small size of the aerogel pieces.

2.4.2.2. Strong Hybrid Aerogels

We also investigated another crosslinking agent to make stronger, transparent hybrid aerogels. The procedure for making these gels are based on the methods reported by Cuney *et al.*¹ The hydrolyzed-Si appears to yield the most transparent gels so far; however, some of the gels were not dried due to low shrinkage and the associated difficulty of removing the gels from the mold. Of the hydrolyzed-Si gels, the most transparent aerogel is one that is 0.05g/cc in target density and low loading of crosslinker. A higher loading of the crosslinker in the same formulation was less transparent.

The gels were a little cloudy when they were cast, which suggests that the cloudiness is more likely related to the hydrolysis step. They were hydrolyzed in water using an acidic catalyst. When the base is added for the condensation step the gels turn cloudy. Some gels were made without using the acid, which significantly increased hydrolysis time, but the gels were not cloudy.

A series of hydrolyzed-Si gels with lower loading levels of the crosslinker as well as different target densities were synthesized (in rectangular shape) for mechanical testing. We also conducted a study on the relationships between optical and mechanical properties and porosity of the silica/urea aerogels. Influence of crosslinker content and target density of the aerogel on porosity of the aerogel were examined. As listed in Table 9, the transparency of the aerogels was evaluated (visually). For preparation of the aerogels, water content and hydrolysis times were

constant. More gels with different water content and hydrolysis time were also prepared. Also, the flexural strength of the aerogels was measured.

Table 9. Transparency of the hybrid aerogels versus target density and crosslinker content.

ID sample	Monoliths							
	TM1	TM2	TM3	Sil4	Sil5	Sil6	Sil7	Sil8
Water ratio	8	8	8	8	8	8	8	8
Crosslinker	5%	5%	5%	5%	5%	5%	20%	30%
Target density (g/cc)	0.05	0.08	0.10	0.05	0.08	0.10	0.05	0.10
Transparency*	3	3	3	4	4	4	3	1

* 5-Good transparency, 1- bad transparency

2.4.2.2.1. Porosimetry and BET Surface Area of Hybrid Aerogels

The porosimetry data are tabulated in Table 10 and illustrated in Figure 26, Figure 27, Figure 28, and Figure 29. The specific surface area of the silica/urea aerogels determined from the BET method is quite high and ranges between 730 and 1045 m²/g. The pore volume of these aerogels is also high (2.33 – 4.18 cm³/g). These hybrid aerogels possess relatively high surface areas and pore volumes compared to regular transparent silica aerogels. For a better comparison, we also list the characteristics of the highly transparent silica aerogel in Table 10. Surprisingly, this material exhibits a lower total pore volume and lower surface area than the hybrid aerogels.

The transparency of these aerogels (except the silica aerogel) was not quantitatively evaluated. We estimated their transparency visually and gave them a value of between 1 and 5, with 5 being the most transparent. The low density (target density = 0.05 g/cc) hydrolyzed hybrid aerogel appears to be the most transparent aerogel among these hybrid aerogels. Although all aerogels possess high surface areas, the transparency does not seem to be directly related to this property. However, the density of the material does affect the average pore diameter of the material. When we compare the average pore diameters for samples Sil4, Sil5 and Sil6, we can clearly see the average pore diameter increases as the density increases (Table 9 and Figure 27).

Table 10. BET surface, pore volume and average pore diameter of the hybrid aerogels.

ID sample	PDMOS.	TM1	TM2	TM3	Sil4	Sil5	Sil6	Sil7	Sil8
S _{BET} (m ² /g)	616	961	904	950	1045	917	816	730	957
Water Ratio	8	8	8	8	8	8	8	8	8
Total Pore Volume (cm ³ /g)	1.79	3.34	3.19	3.68	3.33	4.18	3.93	2.33	3.1
Average Pore Diameter (nm)	10.96	13.9	14.13	15.48	12.77	18.23	19.28	12.75	12.95
Transparency*	5	3	3	3	4	4	4	3	1

Figure 26. Adsorption-desorption isotherms of hydrolyzed hybrid aerogels.

Figure 27. Pore size distribution of hydrolyzed hybrid aerogels.

However, samples Sil4, Sil5 and Sil6 have similar transparencies so the average pore diameter does not seem to be a property related to transparency either. The influence of the urea content is clearly illustrated in Figure 27 by samples Sil7 and Sil8. Higher concentrations of crosslinker noticeably cause the pore size distribution to become broader. Even higher concentrations of this urea crosslinker would definitely be too high and decrease the transparency of aerogels.

For hybrid samples, the pore size distribution (Figure 28 and Figure 29) is narrow and the transparency for all three samples is the same. Again, the target density (for fixed crosslinker loading) does not appear to affect the transparency of the aerogel.

Figure 28. Adsorption-desorption isotherms of hybrid aerogels.

Figure 29. Pore size distribution of hybrid aerogels.

2.5. Fiber Reinforced Translucent Aerogel Panels and Blankets

2.5.1. Selection of Silica Precursors

Aerogels are a microporous silica material which is transparent because the pores and particles are smaller than the wavelength of visible light. Since TMOS, the usual precursor for transparent silica aerogels, is toxic we used alternative precursors so that these materials can be commercialized. As described by Tewari *et al.*,¹⁰ TEOS is a logical choice. However, the aerogels prepared from TEOS are not as clear as those prepared from TMOS and require further optimization of synthesis conditions. Tewari *et al.* studied the hydrolysis and condensation of TEOS to prepare transparent aerogels and showed that it is possible to prepare transparent aerogels from TEOS that are almost as transparent as those prepared from TMOS.

The classical sol-gel process for synthesizing silica aerogels involves the hydrolysis / condensation of TEOS or TMOS to produce a gel which can be supercritically dried. Controlled

hydrolysis of TEOS or TMOS leads to partially hydrolyzed precursors that can subsequently be gelled. The controlled hydrolysis of TEOS and TMOS has been studied by Tillotson *et al.*,^{11 12} and they first hydrolyzed TEOS or TMOS under acidic conditions using less than stoichiometric amounts of water to produce a partially condensed silica precursor. This precursor is then gelled by adding a condensation catalyst.

We used this two step approach to prepare transparent aerogels from two precursors, TEOS and another precursor that is a complex mixture of silicate molecules and contains ethanol, monomer, and various oligomers ranging from dimers to octamers.¹³

2.5.2. Selection of Fiber Reinforcements

Aspen Aerogels considered two types of fibers as reinforcements for aerogels: polyester and glass fibers. Polyester fibers have almost ideal properties for manufacturing aerogel blankets and would be our first choice for highest scale-up potential. However, the density of the usual polyester batting is too high and it is not very transparent. Therefore, we searched for less dense and more transparent alternative polyester battings. These battings are referred to as G polyester, S polyester and polyester P. The transmittance for these fibers was determined and the results are listed in Table 11. The S polyester has the best transparency of those tested and will be used to make fiber reinforced aerogels. Since UV stability is important for daylighting applications other fibers were also considered. This is because polyester fibers may discolor with extended exposure to UV rays.

Glass fibers were also considered since these fibers are currently used in daylighting products and are accepted by the daylighting industry and are expected to have better UV stability than the polyester fibers. Discussions were held with the manufacturer of the glass fiber materials that are used in Major Industry's daylighting products. These materials are available in a range of densities and thicknesses, and are sold as rolled goods. Sample rolls of these materials were received for evaluation, including 5 mm and 12 mm thick rolls. These glass fiber rolls are available in widths up to 90" and lengths up to 600 feet, suitable for integration in Aspen's manufacturing process. A photograph of the glass fiber batting is shown in Figure 30.

Figure 30. Glass fiber Reinforcement.

This glass fiber is available with 3 different fiber diameters, and we determined the transmittance of the glass fiber reinforcements made with different diameter glass fibers (glass fiber, and glass fiber A and B). These glass fibers were evaluated for transmittance using a light transmission box equipped with a light transmission digital meter. The results are listed in Table 11 and shown in Figure 31.

Table 11. Transmittance of fiber reinforcements.

Fiber	Thickness (mm)	Transmittance (%)
S polyester	7	75
	19	67
G polyester	13	54
Polyester P	18	64
Quartzel	12	62
Glass fiber/ binder	3	80
	20	69
	73	49
Glass fiber/no binder	13	88
Glass fiber A/ binder	16	75
Glass fiber B/ binder	15	69

Figure 31. Transmittance versus thickness of candidate fiber reinforcements.

The glass fiber is coated with an organic binder which accounts for about 20% of the batting's weight. This coating is fairly thick and decreases the transparency of the batting. The binder was removed by heating the fiber; burning off the binder led to a clear glass fiber batting that is more flexible but weaker. After removing the binder the fiber is much softer and more transparent as indicated by the data listed in Table 11.

Glass fiber A had better transmittance than the glass fiber previously received. Polyester batting generally showed lower transmittance than glass fiber, but they are more feasible reinforcements for manufacturing a translucent aerogel using Aspen's manufacturing process.

2.5.3. Transparent Aerogels Prepared from TEOS

Transparent aerogels were prepared from TEOS using a similar strategy as reported by Tillotson *et al.*¹² In our case, we used an acid as the catalyst with varying water content. To hydrolyze TEOS, TEOS and water and ethanol were mixed together and stirred. These mixtures are not homogeneous since water and TEOS are not miscible. Therefore, the solutions consist of two phases and are cloudy when initially mixed. When the TEOS begins to hydrolyze, alcohol and heat are generated and the solutions turn clear. The silica ‘sol clear time’ was determined as an indicator of reaction kinetics and hydrolysis was typically complete within 2 hours. The heat generated upon the onset of hydrolysis can be felt by touching the reaction flask. We used these silica sols to make gels after they were hydrolyzed for 3 hours.

All the hydrolyzed silica sols prepared had different ‘sol clear times’, which is an indicator for the hydrolysis rate of silica precursor. The hydrolysis reaction rate is expected to increase as the acid concentration increases. The most dramatic example is a ‘sol clear time’ of 8.5 min. compared to 135 min., which had a 10x lower acid concentration. The reaction rate is also dependent on the stirring speed. Sols had longer sol clear times if the stirring speeds were slower. Since the hydrolysis rates were slower, the exotherm associated with the reaction was also lower. Since the stirring speed and acid concentration control the hydrolysis reaction rate, they can be adjusted to control the temperature rise associated with the reaction. The polydispersity of the precursors prepared at the slower reaction rate did have higher PDIs which may be an affect on the slower reaction rate.

2.5.3.1. Condensation and Gelation of the Sols

The first sets of samples were prepared by diluting sols with ethanol and water to a target density (TD) of 0.0475 g/cc. To gel these sols, a range of dilute basic solutions were added. Two samples were prepared for each composition and one was aged in base followed by aging in a hydrophobic solution. The other sample was aged in EtOH, followed by aging in the same hydrophobic solution. Figure 32 and Figure 33 show photographs of typical samples prepared by this method. Figure 33 shows the monolithic aerogel with the best transparency.

Figure 32. Pictures of aerogel monoliths. Ruler in inches.

Figure 33. Photo of transparent aerogel monolith. Ruler in inches.

The following conclusions can be drawn from these results:

- Higher water to silica ratios in the condensation step results in transparent silica aerogels.
- The two different aging methods did not have a significant effect on the transparency of the aerogels. Aging in ethanol caused less shrinkage but the gels aged in base solution were stronger.
- The sol with the lowest acid concentration produced aerogels with the highest transparency.

In the second batch of aerogels prepared, the hydrolyzed precursors were diluted with ethanol to the desired target density of 0.05 g/cc. Gelation was initiated by adding base as the condensation catalyst. The gels were aged in base and then treated with a hydrophobic solution. After hydrophobic treatment, the gels were supercritically dried.

Figure 34 summarizes the gel times observed for gels prepared and are plotted versus the total amount of water used to make the gels. The gel times become shorter for gels made from the sol where the total amount of water used increased from 8 to 16. The amount of scatter in this data indicates that gel times can be affected by the amount of water used in the hydrolysis step.

Figure 34. Plot of gel time versus amount of water in the total reaction system for gels.

Another observation was that the amount of water used in the hydrolysis step has an effect on the transparency. The dependence of the transparency of the silica aerogel on the different amount of water in the hydrolysis step can be seen from Figure 35. The photographs show that the transparency of the aerogel decreases as the water amount increases to 16 in the hydrolysis step. This result indicates that the transparency of the aerogels could be increased by decreasing the amount of water used to hydrolyze TEOS.

Figure 35. Photographs of aerogel monoliths. The total amount of water to silica ratio is 16 for all samples.

In summary, lower amounts of water in the hydrolysis step produces gels with higher transparency and shorter gel times. Using a lower amount of water in the hydrolysis step and keeping the total water ratio less than 16 also helps to achieve better transparency and uniformity of the aerogel because adding a large amount of water for the condensation step causes bubbles to form in the gel (probably due to condensation reaction continuing after gelation) which decreases the transparency of the aerogel.

2.5.3.1.1. The effect of acid concentration

In order to determine the effect of acid concentration on the transparency of the silica aerogel, TEOS was hydrolyzed with different acid amounts. The influence of acid amount on gel time can also be seen from the data shown in Figure 36. The basic trend is that under the same reaction conditions, the higher the acid concentration, the longer the gel time for total water to silica molar ratio lower than 16 and for those higher than 16, the gel time seems longer for lower acid amount in the hydrolysis step. The gel times are more influenced by the base concentrations and for these experiments, the amount of base added was the same for both sols.

Figure 36. The influence of acid amount on gel time.

The transparency of the aerogel can also be easily seen from Figure 37, which shows photographs of aerogel monoliths made with these high and low acid concentrations. Basically, the results indicate that the lower the acid concentration, the higher the transparency.

Figure 37. Photographs of WD-35-14a, 20a (upper panel, lower acid concentration) and WD-36-14a, and 20a (lower panel, higher acid concentration).

Figure 38 shows the aerogels made using fresh WD-43 silica sol and using 1-day-old WD-43 silica sol. For a total water to silica molar ratio of 8, a decrease in the transparency can be seen for aerogels prepared using fresh and 1 day old sols (upper left panel and lower left panel), but for a water to silica molar ratio of 12 and 16, there is no visible degradation of the transparency, as we can see from the middle and right panels. This result indicates this silica sol is very stable and one day storage of the silica sol will not make the transparency decrease dramatically especially with higher water to silica molar ratios.

Figure 38. Photographs of aerogels prepared from freshly prepared and 1 day old WD-43.

2.5.4. Fiber Reinforced Silica Aerogels Prepared from TEOS

To reduce the manufacturing cost of aerogels we conducted experiments on fiber reinforced aerogels. We used two different types of fibers as the reinforcements for the casting of the transparent aerogel coupons: polyester fibers and glass fibers. Samples were prepared using both types of fibers and experiments were conducted to maximize the transparency of the samples.

2.5.4.1. Glass Fiber Reinforced Silica Aerogels Prepared from TEOS

In order to accurately determine the transparency of the silica aerogel, we made 8" x 8" coupons using glass fiber as the reinforcement material.

Table 12 and Table 13 summarize the properties of the glass fiber reinforced aerogel coupons made from silica sols WD-45 (lower acid concentration) and WD-44 (higher acid concentration). The first notable difference between these two series of samples is that the thickness of samples prepared with WD-45 are ~ 1 mm higher than those for samples prepared from WD-44, but the densities were similar. Also, the thermal conductivities for these two series of aerogels made with different acid amounts are very similar. The TCs (thermal conductivities) are all in the range of 14.5 – 16.5 mW/mK. Accordingly, the R value normalized to 1 inch for all the coupons are in the range of 9-10. We can also see from Table 4, that by using lower acid amounts, used to make WD-44, we obtained glass fiber reinforced aerogel materials with very high transmittance. The transmittance reached 69%, which is just 1% lower than that of the glass fiber and indicates that the aerogel is highly transparent.

Table 12. Properties of aerogels prepared from WD-45.

Sample ID	Total water ratio	Thickness (mm)	Density (g/cc)	TC (mW/mK)	R value /inch	%T
WD-45-4B	4:1	11.78	0.090	15.9	9.1	65
45-6A	6:1	12.87	0.083	15.6	9.2	63
45-6B	6:1	12.45	0.089	16.3	8.8	68
45-8A	8:1	12.81	0.082	15.7	9.2	69
45-12A	12:1	12.33	0.083	15.8	9.1	66
45-16A	16:1	12.75	0.080	15.3	9.4	65
45-20A	20:1	12.81	0.099	15.6	9.2	68

Table 13. Properties of aerogels prepared from WD-44.

Sample ID	Total water ratio	Thickness (mm)	Density (g/cc)	TC (mW/mK)	R value /inch	%T
WD-44-4A	4:1	11.8	0.083	16.4	8.8	53
44-8A	8:1	11.5	0.081	15.3	9.4	56
44-12A	12:1	11.8	0.081	15.0	9.6	64
44-16A	16:1	11.5	0.086	15.4	9.4	56
44-16A1	16:1	11.6	0.083	15.6	9.2	58
44-16A2	16:1	11.8	0.083	14.7	9.8	46
44-20A	20:1	11.1	0.089	15.6	9.2	57

The best transparency for aerogels prepared using WD-44 was 64% transmission for an aerogel prepared using a water ratio of 12:1 and all the others show transmission between 46-60 percent.

2.5.4.2. Polyester S and G Polyester Fiber Reinforced Silica Aerogels

2.5.4.2.1. Condensation of the Silica Sol WD-68

A silica sol was reinforced with two different polyester fibers (8" x 8") as the batting materials for the casting of the transparent aerogel coupons: G and S, which have different densities (the density of S is half that of G). The thickness of G varied from 10 to 19 mm and thickness of S polyester from ~7-10 mm. The transmittance of the fiber increased slightly after being lofted, as can be seen from the transmittance of the fibers in Table 14. The transmission of the aerogel coupons decreased slightly for G-unlofted, which are 48.5% or 49.5%. For the lofted G polyester coupons, the transmittance is lower than it is for the unlofted G polyester batting materials. The TC data of both lofted and unlofted fiber batting materials are 13.5 – 15.5 mW/m-K. The TC of WD-68-12C gives the lowest TC value for G polyester reinforced aerogels.

Table 14. Properties of fiber reinforced aerogels prepared using WD-68.

Sample ID- WD-	Fiber	Thickness	Density	TC (mW/m•K)	R/inch	%T
WD-68-12A	G-unlofted, ~10 mm thick	8.60	0.0836	15.3	9.4	49.5 /fiber: 50
68-12B	G-lofted, ~17mm thick	11.63	0.0809	14.5	9.9	48.5 /fiber: 51
68-12C	G-lofted, ~12 mm thick	8.64	0.0839	13.8	10.4	45 /fiber: 49
68-12D	G-lofted, ~19mm thick	11.96	0.0816	14.9	9.7	45 /fiber: 52
68-12E	S-unlofted, ~7mm thick	6.98	0.0787	13.2	10.9	64 /fiber: 64
68-12F	S-lofted, ~17mm thick	9.16	0.0791	14.6	9.9	63 /fiber: 65

There is only one type of S fiber available, and it can be lofted to attain a thickness of ~ 17 mm. The TC of both coupons using lofted and unlofted S fiber give TC of 14.6 and 13.2 mW/mK, respectively, while the transmittances were similar, 64 vs. 63 %. There is one disadvantage of the S fiber: the fiber distribution is not perfectly uniform in the batting. Therefore, the transmittance can be 70 % for one area of batting but 65 or 64 % for another area of the batting. This non-uniform distribution of fibers may be due to the low density and high loftiness of the batting.

Comparing the TC data and transmission data of G, S polyester and glass fiber reinforced aerogel coupons, we can conclude that S and glass fiber both give high light transmission while G is obviously not a good choice in terms of transmittance. However, the TCs of aerogels reinforced with G and S are 13-14 mW/mK, which corresponds to a R/inch value of 10-11. The TCs for polyester reinforced aerogels are lower than those for aerogels reinforced with glass fiber battings.

Photographs of the coupons prepared are shown in Figure 39.

Figure 39. Photographs of the aerogel coupons with different batting materials. (a) S polyester as batting material; (b) G polyester as batting material; (c) S polyester as batting material; (d) Glass fiber as batting material.

In order to optimize the TC and transmission data, we made aerogel coupons using S-lofted fiber as batting material and tried different water to silica ratios in the silica sol gelation step. In both cases investigated, clear gels were obtained before extraction.

Table 15 summarizes the TC and transmission data for the aerogel coupons. One can see that a water to silica ratio of 4 and 6 resulted in lower TCs, i.e. 13.6 mW/m-K, while water to silica ratios larger than 8 showed TCs of 14 - 14.6 mW/m-K. This result means that the water to silica ratio did increase the TCs slightly. The transmittance of the aerogel coupons can be maximized by using higher water to silica ratios, which again proves results obtained for aerogel monoliths. The photographs of the aerogel coupons can be seen in Figure 40.

Figure 40. Photographs of aerogel coupons made by S polyester reinforcement. Left pane: WD-68-16A and right panel: WD-68- 20A.

Table 15. Properties of fiber reinforced aerogels prepared from WD-68.

Sample ID-WD-	Total water ratio	Thickness	Density g/cc	TC (mW/mK)	R/inch	%T
68-4B	4:1	8.26	0.0909	13.6	10.6	49.5
68-6B	6:1	7.98	0.0905	13.6	10.6	48.5
68-8B	8:1	8.06	0.0836	14.5	9.9	45
68-12A1	12:1	8.20	0.0860	14.0	10.3	45
68-16A	16:1	8.27	0.0787	14.4	10.0	64
68-20A	20:1	8.27	0.0790	14.6	9.9	63

Therefore, the advantage of using S polyester over glass fiber is the possibility to lower the TC of the aerogel coupons to 13-14 mW/m-K without decreasing the transmittance of the material.

2.5.4.2.2. Condensation of the Silica Sol WD-69

From the above discussion, aerogel reinforced with glass fiber batting normally have higher TC values than aerogel reinforced with S fibers. Since glass fiber is more transparent and is currently used by the translucent windows industry, we tried to optimize the TC data by tuning the target density of the aerogel coupons in order to lower the TC value.

A downselected silica sol was used to prepare aerogels with different densities. The size of the glass fiber reinforced aerogel coupons was 8" x 8". The excellent transparency of these monolithic aerogels is illustrated in Figure 41.

Figure 41. Photographs of WD 69- 5a, 6a and photographs of WD 69-7a, 8b.

Fiber reinforced aerogel coupons were made by using the glass fiber as reinforcement. The photographs of the coupons are shown in Figure 42. Target densities were 0.05, 0.06 g/cc for 69-5a, 6a respectively and 0.07, 0.08 g/cc for 69-7a, 8a respectively. The properties of these aerogels are shown in Table 16.

Figure 42. Photographs of the WD-69-5A (left panel) with target density of 0.05 g/cc and WD-69-6A (right panel) with target density of 0.06g/cc.

Table 16. Properties of aerogels prepared using WD-69.

Sample ID- WD-	TD (g/cc)	Final Density (g/cc)	% T	Thickness (mm)	TC (mW/mK)	R/inch
69-2A	0.02	0.0746	57, bad aerogel	6.27	19.92	7.2
69-3A	0.03	0.0665	55, bad aerogel	9.19	22.88	6.3
69-4A	0.04	0.0730	62	11.62	16.38	8.8
69-5A	0.05	0.0790	64	13.04	16.23	8.9
69-6A	0.06	0.0952	63	12.78	16.12	8.9
69-7A	0.07	0.1052	64	13.47	16.32	8.8
69-8A	0.08	0.1217	64.5	13.16	16.72	8.6

One can see from the above discussion that the transparency of the aerogel coupons can reach 69% using glass fiber as reinforcement, which is only 1% less than the glass fiber. The glass fiber reinforcement gives the best transmittance of the aerogel coupons, which is comparable to the S-polyester reinforcement, but G gives transmittance of ~ 50 % and that transmittance is limited by that of the G polyester fiber.

Our results also indicate that the type of fiber affects the thermal conductivity. The TC data for samples using the glass fiber reinforcement (15-17 mW/mK) is higher than those using S and G polyesters (14-16 mW/m-K), as indicated by the results shown in Figure 43; however, there is very little correlation with aerogel density. The aerogels reinforced with glass fiber usually have a few % higher transmittances, but they have more particle shedding than those reinforced with the polyester. Overall, the polyester is a better reinforcement. UV stability of the aerogel panels reinforced with polyester fiber will be discussed later.

Figure 43. Thermal conductivity versus density for fiber reinforcements.

2.6. Development of Process to Manufacture Fiber Reinforced Translucent Aerogels

2.6.1. Manufacturing Challenges

A review of the manufacturing costs associated with aerogel daylighting panels was undertaken. Alternative production methods that were considered are:

- 1) **Panel Process:** This process consists of producing discrete 12" x 24" panels to meet standard size requirements of domestic daylighting product manufacturers.
- 2) **Roll Process:** This process consists of casting the sol into a fiber batting that is approximately 60" wide. The sol gels as the batting is rolled on a mandrel and then supercritically dried.
- 3) **Pre-formed Roll Process:** This process consists of performing the fiber batting into a roll before the sol is cast. The preformed roll is impregnated with the sol and after gelation the roll is supercritically dried. This process avoids rolling the batting filled with wet gel.

The panel process would be the scale-up approach currently used by Aspen Aerogels to produce transparent and translucent aerogel-based materials in our research laboratory, while the roll process is similar to the approach used to manufacture flexible aerogel blanket products currently being manufactured by Aspen Aerogels. The pre-formed roll process is a hybrid of the two methods and allows us to prepare translucent aerogels with the same throughput as the blanket product. All of the methods have a number of challenges; however, a preliminary comparison indicated that the panel process would be more than twice as expensive as a roll process.

Challenges to cost effective production which were studied include:

- **Panel Production:** The panel process requires a large number of individual tray-like molds which require specialized handling, and the panels do not pack very tightly in typical production equipment.
- **Roll Process:** The roll process assumes that the wet gel can be bent around the mandrels and the aerogel unbent from the roll without suffering significant damage. Tests have indicated that the aerogels made by this process have decreased transparency.
- **Preformed Roll Process:** In the preformed roll process, the sol is gelled within a batting that has been previously bent around the mandrel. Although the wet gel does not need to be bent, the dried aerogel must be removed from the roll (unbent) before it can be installed in a fenestration product. Positive results have been obtained in a lab setting, and the aerogels had similar transparency as samples that had not been rolled.
- **Alcohol costs:** At the present time a significant volume of clean, fresh alcohol is required to produce good quality translucent, aerogels; i.e., using pure alcohol increases the production costs. The added cost of using pure alcohol does not appear to be prohibitive to manufacturing the translucent aerogel at our target price. Developing methods to use recycled alcohol will be the first step undertaken to reduce our manufacturing costs.

2.6.2. Fiber Reinforcements

Three different fiber battings were studied as reinforcements for translucent aerogel rolls which will be referred to as 1) glass fiber, binderless glass fiber, and polyester fibers. The glass fiber

batting contains about 20-25% organic binder and is a somewhat rigid, yet still roll-able, and has good transparency (Figure 44). The binderless glass fiber was prepared by heating the glass fiber batting in a furnace to thermally decompose the binder. The resulting fiber batting is very transparent but weak, and it sheds small fibers (Figure 44).

Figure 44. Glass fiber A) with binder and B) after removal of binder

The polyester has the lowest transparency of all fiber reinforcements being tested, but has the highest flexibility and strength (Figure 45).

Figure 45. Polyester fiber batting.

2.6.3. Thermal Stability of Glass Fiber

The thermal behavior of the material needs to be understood, to ensure suitability for use in Aspen's manufacturing process. To determine the fibers thermal stability and the amount of

binder coating the fibers, the fiber was evaluated by TGA/DSC (Figure 46).

Figure 46. TGA/DSC of candidate glass fibers coated with binder.

A sample was placed into the TGA/DSC, and ramped from ambient temperature to 400 °C. The material showed some mass loss (~1.3%) between 100 and 150 °C, likely due to water loss. Additional mass loss began at ~200 °C, losing 22.45 % of its total mass by 400 °C. This mass loss is associated with the decomposition of the polymeric binder that is used in the product, and correlates to the mass percentage range of binder that the manufacturer said the product contained. According to the TGA, the binder begins to decompose at ~200 °C, which is a lower temperature than that used for the final drying temperature. Therefore, the oven temperature would need to be reduced when this product is heat treated at the end of the manufacturing process.

The fibers were tested for compatibility with the processing fluids used in Aspen's aerogel blanket manufacturing process. To demonstrate that the binder and fiber will not dissolve in the current processing media, a sample of the fiber was weighed and then placed in a solution that mimics the fluids used in Aspen's aging process. The measured mass loss was only 0.7%.

Since the transparency of the glass fiber after calcination (without binder) is much higher than that of the glass fiber (with binder), we tried to use the calcined fiber as the batting material. Removing the binder can have several advantages: the fiber flexibility increases, it can be filled better with sol during casting, soaks the sol better and it is much more transparent.

After calcination the fiber is much softer and its chemical stability also needed to be tested. The stability of this fiber in the solvents used to prepare translucent aerogels were tested. After calcination this fiber is not very stable in the required solvents.

The thermal behavior of the aerogel reinforced material also needs to be understood, to ensure that it would be suitable for use in Aspen's manufacturing process. The stability of the aerogel-

glass fiber composite was investigated by TG-DSC, as shown in Figure 47. As can be seen from Figure 46 and Figure 47, both fiber reinforced silica aerogel and the fiberglass lose the binder at ~ 400 °C (~ 23% wt). However, there was some weight gain after 400 °C which has not yet been explained.

Figure 47. TG-DSC data for the glass fiber reinforced aerogels.

Aspen evaluated the glass fiber batting in terms of its ability to withstand a roll to roll processing technology. A 30 foot long sample was used to make an aerogel blanket. The standard formulation was used to make a blanket. After supercritical drying, the blanket was heated to remove residual solvent at a lower temperature than normally used to ensure no oxidation of the glass fiber's polymeric binder. This test was conducted only to evaluate the ability of the fiber material to withstand typical processing conditions in our manufacturing facility, and was not intended to produce materials that are suitable for integration into daylighting products.

The glass fiber batting with organic binder was too rigid, has too many voids and did not produce a good product in the roll process. Problems arose when working with it from the casting step (the sol easily flows out of the fiber since the fiber doesn't soak well) to rolling of the gel (fiber is too rigid, may "cut" through the gel and crack it), and finally to handling of the aerogel (voids in the fiber promote aerogel shedding).

As discussed later, the final fiber selection was also dependent on additional testing involving the UV stability of the batting. The glass fiber batting is currently used in daylighting applications and is expected to pass UV tests. The polyester fiber had not previously been tested for UV stability.

2.6.4. Preparation of a Lab Scale Pre-formed Aerogel Roll

As opposed to aerogel manufacturing casting on the conveyor and by rolling of gel sheets, the pre-formed method is based on casting the catalyzed sol in a rolled fiber-pre-form. This method

simplifies and accelerates the processing of gels and leads to higher quality aerogel by eliminating rolling steps. Rolling can damage the aerogel and its light transmittance performance, since it puts a mechanical stress on the frail gel and introduces cracks which scatter light.

A glass fiber reinforced aerogel roll of 7ft long (Figure 48) was produced by this method using a 7.5 in. diameter mold in less than 24 hours. The flexible aerogel showed a thermal conductivity of 14.6 mW/m-K, at 8.9 ± 0.2 mm thickness and 0.080 g/cc density (D).

Figure 48. Example of aerogel blanket prepared by pre-formed roll process.

2.6.4.1. Preparation of the Pre-formed Aerogel Roll

To prepare the pre-formed roll, the fiber batting was co-rolled with a plastic separator over a 2" diameter mandrel as shown in Figure 49.

Figure 49. Co-rolling of binderless glass fiber batting and plastic separator.

The fiber roll was placed in a mold that was 5 inches high so that it completely filled the mold (Figure 50).

Figure 50. Rolled fiber and separator in mold ready for sol casting.

A sol formulation utilizing a partially hydrolyzed silica precursor was scaled up to 3L volume in order to prepare the transparent aerogel roll. The rolled fiber was infiltrated with the catalyzed sol which gelled quickly. Although there was enough time to completely infiltrate the fiber roll with the sol, the gelation time may have been too rapid for complete elimination of air bubbles. The gelled roll was kept in the closed mold for another 3 hours after gelation, and then it was transferred into a larger diameter vessel for aging (Figure 51). The monolithic aerogel roll was loosened by partial unrolling so that the aging solution could penetrate all the layers of the gel blanket (Figure 51).

Figure 51. Gelled “monolithic” roll in aging bath

Aging of the roll was carried out to make the aerogel hydrophobic and strengthen the gel. The solvent was exchanged after aging to remove the excess hydrophobe reagent and the base used to strengthen the gel. At this point, the gel is strong enough to be handled and transferred to the extractor for supercritical drying (Figure 52).

Figure 52. Pre-formed gelled roll ready for extraction.

Solvent extraction was performed under subcritical liquid CO₂ conditions for 3 hours to extract the majority of the solvent. After solvent removal, the roll was dried under supercritical CO₂ for 3 hours followed by a 2 hour depressurization period to obtain the aerogel blanket.

2.6.4.2. Evaluation of Glass Fiber Reinforced Aerogel Blanket

The sheets of fiber reinforced aerogel of 5" width and 2 ft length were unrolled, dried for several hours, then analyzed visually (Figure 53).

A. Aerogel- binderless glass fiber

B. Aerogel- glass fiber

C. Aerogel- polyester

Figure 53. Fiber reinforced aerogel from pre-formed rolls of A) binderless glass fiber, B) glass fiber and C) polyester battings.

Defects were found mainly in the aerogel reinforced with glass fiber without binder, in the form of cracks (Figure 54, A) and uneven thickness/transparency (Figure 54, B).

Figure 54. A) Cracks and B) thickness variations in aerogel blanket reinforced with the binderless glass fiber batting.

The glass fiber reinforced aerogel, which is the thickest and most rigid of all the fiber reinforced aerogels studied shows some cracks and creases, especially in the region of tighter rolling (Figure 55).

Figure 55. Defects in the glass fiber reinforced aerogel.

By contrast, the polyester reinforced aerogel shows the highest homogeneity and flexibility, virtually free of defects (Figure 56).

Figure 56. Polyester reinforced aerogel sheet (left) and glass fiber reinforced aerogel (right).

The fiber reinforced aerogels were tested for thermal performance and transmittance (Table 17). The average thickness and the standard deviation were reported. Twenty data points were taken to measure the thickness of the aerogel sheets.

Table 17. Performance of fiber reinforced aerogel from small-scale pre-formed roll.

Fiber	Average thick/ sheet (mm)	T C (mW/mK)	Density (g/cc)	Thickness/ Coupon (mm)	Transmittance coupon %	R- value/ inch
polyester	4.0±0.7	11.6	0.114	4.8	58	12.4
Glass fiber without binder	5.1±1.2	11.7	0.118	6.5	65	12.3
Glass fiber with binder	9.8±0.9	13.5	0.120	8.8	61	10.7

The highest standard deviation in thickness was for the blanket prepared using the binderless glass fiber, which, as mentioned before, is not uniform after binder removal. The best thickness

uniformity was obtained for the resilient polyester reinforcement. This fiber, however, gave aerogel blankets with the lowest thickness and transmittance. The binderless glass fiber reinforced aerogel blanket sheds aerogel particles and is not uniform, but it has the highest transmittance, while the glass fiber reinforced aerogel blanket has the highest thickness and TC. All of the reinforced aerogels had R/inch values of greater than 10/inch.

2.6.5. 2 ft Wide Glass Fiber Reinforced Aerogel Roll

A larger pre-formed roll was prepared by casting silica sol onto a roll of glass fiber reinforcement. The fiber was co-rolled with a separator and was placed on a mandrel that had a perforated plate welded at the bottom. A perforated top plate secured to the mandrel kept the roll in place during casting of the sol.

The glass fiber/HDPE roll was placed in a stainless steel canister and the sol was cast inside the canister. This approach eliminated sol migration outside of the fiber roll which previously occurred when plastic bags were used. The plastic bags were unable to properly act as secondary containment of the sol.

Gelation of the silica-based sol occurred in enough time to allow all air trapped within the roll to escape. After processing and drying the resulting aerogel sheet measured 13 ft long by 23.5" wide. The semi-flexible aerogel material showed good transparency but contained some cracks on the inner face of the roll (Figure 57). The average thickness of the blanket was 5.9 ± 2.0 mm. A plot of blanket thickness versus length is shown in Figure 58.

Figure 57. Glass fiber reinforced aerogel roll.

Figure 58. Mapping of the glass fiber aerogel roll (0 = outside of roll).

Coupons were cut from the roll and tested for TC and light transmittance, both as single coupons or a stack of two coupons to double the thickness. The results are listed in Table 18.

Table 18. TC and transmittance of the glass fiber reinforced blanket.

Coupon	TC (mW/mK)	Thickness (mm)	Thickness (inch)	R- value	R/inch	Transmittance (%)
1	13.1	8.825	0.35	3.82	10.99	64
2	13.8	10.325	0.41	4.24	10.43	54
stack	16.9	19.15	0.75	6.42	8.51	37

The data indicates that the transmittance decreases with increasing thickness and R-value as would be expected (Figure 59). To have an R-value of 4, the glass fiber reinforced aerogel would have a transmittance of about 60% which is similar to that obtained for the polyester reinforced blanket.

Figure 59. Glass fiber aerogel transmittance as a function of thickness.

2.6.6. 2 ft Wide Polyester Fiber Reinforced Roll

A preformed roll was prepared by casting a silica sol onto a polyester reinforcement. The fiber was co-rolled with a separator horizontally at medium tensioning on top of a core rolled around a mandrel fitted with a perforated bottom plate. A perforated top plate secured to the mandrel kept the fiber/separator roll in place during casting of the sol. After processing the aerogel sheet was 13 ft long and 23.5" wide. The flexible aerogel material showed good transparency and was crack-free (Figure 60).

Figure 60. Polyester reinforced aerogel roll

The aerogel sheet thickness was mapped according to the following diagram (Figure 61).

Figure 61. Aerogel sheet – positioning for thickness mapping.

Thickness data is given in Table 19.

Table 19. Thickness of the polyester reinforced aerogel

Position	Length (in)	Thickness top (mm)	Thickness bottom (mm)
Outer	5	7.3	7.7
	10	6.4	7.5
	15	7.3	6.6
	20	6.8	7.4
	25	6.6	7.4
	30	6.5	6.3
	35	7.0	7.1
	40	7.7	6.6
	45	6.4	7.3
	50	6.2	6.8
	55	6.2	6.5
	60	6.7	6.3
	65	7.2	7.0
	70	6.7	6.5
	75	6.7	6.6
	80	5.8	6.3
	85	6.6	6.6
	90	7.2	7.1
	95	6.9	7.9
	100	6.9	7.5
	105	6.4	6.2
	110	6.5	5.9
	115	5.9	6.3
	120	6.6	4.6
	125	5.6	4.7
	130	4.6	5.4
	135	4.7	5.6
	140	5.3	5.1
Inner	145	5	4.9
Average (mm)		6.4	6.5
St. dev.		0.77	0.90

The total average thickness was 6.4 ± 0.8 mm. This is the lowest standard deviation we have obtained so far. Mapping of the roll thickness is shown in Figure 62. The thickness was relatively uniform for the most part and it decreases toward the inner portion (side closer to the mandrel) where winding of the fiber was tighter.

Figure 62. Mapping of the polyester S-40 reinforced aerogel roll

Coupons were cut from the roll and tested for TC and light transmittance (Table 20).

Table 20. Evaluation of coupons from aerogel roll 51308

Coupon	TC (mW/mK)	Thickness (mm)	R-value	Transmittance (%)
1	11.9	2.1	1.03	64
2	11.8	3.7	1.78	61
3	12.3	7.2	3.31	58

A comparison of the transmittance of this roll with medium winding and the one with tight winding (PE 2-20-08) is given in Figure 63. The thickness and transmittance were measured for single layer coupons of 4"x 4" size, except for one data point. Overall, the transmittance was higher for the roll with medium winding; however, the data point for overlaid coupons (two layer of fiber) from roll PE 2-20-08 should be disregarded since the rest of the measurements were reported for a single layer.

Figure 63. Transmittance of polyester reinforced rolls function of thickness

The transmittance of the polyester and glass fiber reinforced aerogels appears to be similar considering their transmittance versus R-value (Figure 64). The glass fiber sample with 37% transmittance is for two layers of aerogel. The glass fiber reinforced sample that was 11 mm thick had excellent transmittance and an R-value of 3.82.

Figure 64. Transmittance as a function of R-value for glass fiber and polyester reinforced aerogels.

Samples listed in Table 21 were submitted to Major Industries for SHGC testing. Square foot coupons were cut from polyester and glass fiber rolls and submitted for ASTM C-518 testing (Table 22).

Table 21. Aerogel sheets of 2'x3' for testing at Major Industries.

Sample 2'x3'	TC (mW/m ² •K)	Thickness average (mm)	R-value	Density (g/cc)	Areal density (g/sq ft)	Transmittance* (%)
S51308	12.3	6.8±0.5	3.13	0.110	72.3	58
FG41408_1	13.2	5.7±1.0	2.45	0.130	65.2	64
FG41408_2	13.2	5.6±0.7	2.40	0.131	63.5	64

*for coupons of similar thickness; S-polyester; FG-glass fiber

Table 22. Evaluation of coupons submitted for ASTM-C518 testing.

Sample 12"x12"	TC (mW/m ² •K)	Thickness average (mm)	R-value	Density (g/cc)	Areal density (g/sq ft)	Transmittance* (%)
S22008_1	12.1	3.7±0.6	1.73	0.120	40.8	57
S22008_2	10.7	3.1±0.7	1.64	0.126	36.5	59
S22008_3	11.8	3.3 ±0.3	1.58	0.125	38.3	58
FG414_1	13.4	4.4±0.2	1.86	0.131	53.1	67
FG414_2	13.0	3.5±0.2	1.53	0.135	43.5	70
FG414_3	13.2	4.1±0.5	1.76	0.123	47.1	66

*for coupons of similar thickness; S-polyester; FG-glass fiber

2.7. Testing of Aerogels for Transmittance and UV Stability

Discussions were held with DOE, NETL, LBNL, and NREL to determine the number and size of aerogel samples required for advanced testing of aerogel fenestration systems. It was determined that four 5" x 5" samples were needed for durability testing at NREL. These four samples included one glass fiber sample and 3 polyester samples. LBNL required two 12" x 12" samples and one 2" x 2" sample for testing in a large integrating optical sphere and spectrometer, respectively. The aerogel samples required for these tests were cut from the large scale "blankets" showing that the reinforced aerogels can be cut using scissors as shown in Figure 65 and Figure 66.

Figure 65. Aerogel reinforced with polyester fibers.

Figure 66. Aerogel reinforced with glass fiber.

2.7.1. Properties of Aerogels used for Fabrication of IGUs for Testing

The properties of the aerogel samples prepared for the fenestration tests are listed in Table 23 and illustrated in Figure 67.

Table 23. Fiber reinforced aerogels sent for testing at NREL and LBNL.

Coupon	Dimensions cm x cm (in x in)	Thickness (mm)	Transmittance (%)	TC (mW/mK)
Glass fiber (hand rolled)	29.8 x 30.8 (11.7 x 12.1)	16.5±0.9	59-63	16.6
Polyester (hand rolled)	28.0 x 29.8 (11.0 x 11.7)	14.1±1.0	63-65	14.2
1. Polyester (pre-formed roll)	12.1 x 12.7 (4.8 x 5.0)	9.1 ±0.5	62	13.4
2 Polyester (pre-formed roll)	12.3 x 12.7 (4.8 x 5.0)	7.4 ±0.7	63	12.8
3. Polyester (pre-formed roll)	12.2 x 12.7 (4.8 x 5.0)	7.4 ±0.7	64	13.4
4. Polyester (pre-formed roll)	5.1 x 5.2 (2.0 x 2.0)	8.9±0.2		
Glass fiber coupon	12.7 x 12.7 (5.0 x 5.0)	13.5±0.6	63	16.1

Figure 67. The two 12''x 12'' panels prepared: panel one on the left is reinforced with polyester fibers and the one on the right is reinforced with glass fibers.

2.7.2. Fabrication and Testing of Insulated Glazing Units (IGUs) with Low-e Coatings at NREL and LBNL.

NREL tested the UV stability and environmental durability of the aerogel samples. The effect of low-e glass on the aerogels stability was also determined at NREL. Four 5'' x 5'' samples (one glass fiber sample, 3 polyester samples) were shipped to NREL in March 2007. The glass fiber sample and one of the polyester samples were tested in a clear glass IGU with aluminum tape around the edges. One polyester sample was tested with one clear glass glazing and one low-e glass glazing to determine the effects of the low-e glass on the aerogel. This IGU will also be

made with aluminum tape around the edges. The final polyester sample will be tested for prolonged durability in an aging chamber (6 – 12 months). LBNL sent the required glass and aluminum tape for preparing the IGU's directly to NREL.

LBNL sent Aspen an IGU kit to fabricate two IGUs using the aerogel panels illustrated in Figure 67. The IGU kit consisted of foam spacers, aluminum tape and four sheets of glass. Aspen Aerogels used the IGU kit sent by LBNL to fabricate two aerogel insulated IGUs, and photographs of the finished insulated glazing units prepared are shown in Figure 68. These two IGUs were sent to LBNL in May 2007 for further testing of their optical properties. The smaller 2" x 2" aerogel sample will be tested in LBNL's spectrometer.

Figure 68. 1' x 1' IGU insulated with aerogel reinforced with A) glass fibers and B) with polyester fibers.

2.7.2.1. Optical Testing at LBNL

LBNL received the two 12" square IGUs and the 2" x 2" translucent aerogel sample. LBNL determined that the polyester fiber reinforced sample had a thickness of approximately 0.5" and the glass fiber reinforced sample had a thickness of approximately 0.75". The aerogel sample surfaces were not perfectly flat so these are approximate thicknesses. Based on reported thermal properties from Aspen Aerogels, neither of these samples alone was likely to reach the target thermal performance of 0.10 BTU/h- ft²-F. Since the objective was to fabricate a window with 0.10 BTU/h- ft²-F, LBNL created an Insulated Glazing Unit (IGU) by stacking the two aerogel layers between glazings (Figure 69). The total thickness of the glazing cavity with the combined aerogel layers was 1.34", and the glass layers on either side were 0.12" thick. The thermal and optical tests described below were performed on this sample with a 1.34" thick layer of aerogel.

LBNL initially tested the individual aerogel samples using the Perkin Elmer 950 in their optics lab. Since the samples are "optically thick" and diffusing, the integrating sphere on the device did not receive a sufficiently large signal; therefore, LBNL determined that this approach would not provide useful results. The overall IGU sample (Figure 69) was then tested on the LBNL large integrating sphere.¹⁴ This 7.0 ft. diameter sphere can accommodate large samples up to about 12" square. A 4" square aperture port was used for these measurements on the 12" x 12"

aerogel samples supplied to LBNL. The measurements were performed outdoors in direct sunlight in accordance with ASTM E1175. LBNL measured visible transmittance using Licor (LI-210) detectors which are spectrally corrected for photopic illuminance.

Figure 69. Section of insulating glass unit containing two aerogel samples.

A schematic of the integrating sphere is illustrated in Figure 70. The integrating sphere (A) is positioned so that the sample (B) is perpendicular to the sun. Sensor (C) measures the incident illuminance, and sensor (D) measures the interior illuminance level. An ‘open hole’ calibration measurement was first performed without sample (B) in place, and the ratio of signals from sensors C and D was calculated:

$$T_{\text{open hole}} = I_c/I_d \quad (4)$$

Then the sample was put in place, and another measurement was taken:

$$T_b = I_c/I_d \quad (5)$$

The transmittance of the sample (B) is the ratio of the two measurements:

$$T_{\text{sample}} = T_b/T_{\text{open hole}} \quad (6)$$

All measurements were taken outdoors on a clear day at normal incidence angle to the sun and with combined direct and diffuse illumination. Since it was a clear day with bright sunshine LBNL did not make additional measurements to compensate for the diffuse illuminance component. The visible light transmittance for the total IGU sample (2 pieces of clear glass, and 2 stacked aerogel layers) was determined to be 0.30. Using nominal transmittance values for 0.12" thick clear glass ($T_{\text{vis}} = 0.90$) LBNL estimated the net aerogel transmittance of this composite sample to be 0.37, which is an acceptable value for daylighting applications.

Figure 70. Schematic (not to scale) of 7 foot diameter integrating sphere.

2.7.2.2. Thermal Testing at LBNL

The thermal testing was performed using LBNL's Infrared (IR) thermography equipment with a freezer serving as the coldbox as shown in Figure 71. (Note- the larger scale controlled thermal chamber normally used in IR imaging tests was not available for these tests). This method does not directly determine the conductivity or R-value, but rather measures surface temperatures under controlled environmental conditions and can be used for comparative thermal tests relative to a standard. In these tests, LBNL measured the thermal performance of the aerogel IGU sample in comparison to the thermal performance of a reference insulating material. LBNL compared the stacked aerogel IGU specimen (overall 1.58" thick) to a 2.07" thick foam board with a nominal conductance of 0.1 BTU/h- ft²-F. The temperature difference across the samples (cold chamber to room side) was 54 °F. As can be seen in the IR image in Figure 72, the two materials had very similar surface temperatures and therefore would have a similar overall thermal conductance. Since the 1.34" thick stacked aerogel assembly (we ignore the contribution of glass) has a conductance of approximately 0.1 BTU/h- ft²-F, a nominal 1" thick sample would have a conductance of approximately 0.13 BTU/h- ft²-F or about R7.7 per inch.

Figure 71. IR thermography schematic setup.

Figure 72. IR image of aerogel mounted in R-10 foam; temperature difference was 30 °C.

2.7.2.3. Testing of UV Stability at NREL

NREL fabricated IGUs and loaded the translucent aerogel samples (both polyester and glass fiber samples) into a XR260 chamber to measure UV stability. The samples were tested at 85 °C under 1 sun illumination for approximately 2000 hrs to coincide with typical EC window testing circumstances.

Photographs taken during the fabrication of the IGU test samples are shown in Figure 73. The aerogel samples were placed between pieces of glass and taped around the edges to make the IGUs. Sample D3 was fabricated with one piece of regular glass and one of a low-e glass. Photographs of the completed IGUs are shown in Figure 74. Each of the insulating devices was outfitted with a 0.005"-diameter Type K thermocouple pasted to the top center surface with an 8 mm-square patch of aluminum tape (see photographs in Figure 75).

Figure 73. Fabrication of the test samples.

Figure 74. Photographs of the IGUs before start of testing.

Figure 75. Photographs of the IGUs and the attached Type K thermocouples.

Photographs of the Lambda 900 Spectrophotometer configuration and the initial optical data are presented in Figure 76 and Figure 77, respectively and summarized in Table 24.

Figure 76. Spectrometer used to determine spectral transmittance of aerogels.

Figure 77. Spectral transmission of samples D1-D4 before the test.

Table 24. Summary of transmittance results from NREL.

NREL #	Aerogel Coupon I.D. (material)	Thickness (mm)	Aspen Trans (%)	Trans measured at 550 nm (%)	IGU Structure
D1	S 1 Polyester	9.1 ± 0.5	62	48.9	No glazings
D2	S 2 Polyester	7.4 ± 0.7	63	45.9	G/G
D3	S 3 Polyester	7.4 ± 0.7	64	43.2	G/LEG
D4	GF Glass fiber	13.5 ± 0.6	63	33.7	G/G

All of the samples were less transparent than estimated by our internal measurements. The glazings did not appear to decrease the transmittance in the visible region but the transmission in the NIR was lower for the aerogels inserted into simulated IGUs. This result was anticipated since the glass glazings are expected to have a higher reflectance (~10%) than the aerogels. Therefore, the IGUs would be expected to have 10-20% lower transmissions than determined for the original aerogels. However, the % transmission for sample D1, which is the spectrum for the original aerogel without glazings, was expected to be higher than measured. Perhaps the most interesting and encouraging result was for sample D3. The simulated IGU for sample 3 was fabricated with low-e glass for glazing 2. The spectrum obtained is that expected for the low-e glass and indicates that the aerogel insulation insert is compatible with low-e coated glazings. The low-e coating decreased the transmission at 550nm by only 2.7% and this IGU would have a lower SHGC than those made with clear glass.

NREL's Atlas XR-260 large-component environmental test chamber, as detailed in Figure 78, provides simulated solar irradiance using four 6500-watt, water-cooled, long-arc xenon lamps mounted in the ceiling above a height-adjustable 4'x 6' sample plane. The xenon lamps are spectrally filtered (CIRA inner filter, soda lime outer filter) to provide precise solar cut-on and an excellent spectral match to an AM 1.5 terrestrial global solar spectrum from 300 to 800 nm. At longer wavelengths, xenon emission lines, which are unimportant for our test purposes compromise the fit to the solar spectrum, because they generally do not contribute to photo-induced degradation. The aerogel sample locations are indicated by EC in Figure 78.

Figure 79 shows the typical xenon lamp spectra as compared to an AM 1.5 global solar spectrum. Figure 80 shows the spectral irradiance measured inside the test chamber, and Figure 81 shows a map of the sample plane irradiance. Photographs of the IGUs within the XR-260 chamber are shown in Figure 82.

Figure 78. Schematic of Atlas XR-260 large-component environmental test chamber.

Figure 79. Xenon lamp spectra as compared to an AM 1.5 global solar spectrum.

Figure 80. Xenon lamp spectra as compared to an AM 1.5 global solar spectrum measured inside the XR-260 between 300 and 500 nm.

Figure 81. Map of spectral irradiance on test chamber floor.

Figure 82. Photographs of samples in test chamber before UV exposure test.

Final room temperature photographs of the aerogel samples after 1934 hours of testing are presented in Figure 83, along with the corresponding final optical spectra in Figure 85 and Figure 86 and a tabulation of all initial and final optical data in Table 25.

Visual observations of the devices after testing are as follows:

- All samples showed very little visual degradation (to the naked eye) after 1934 hours in the XR260.
- On sample D1, the area beneath the band of aluminum foil exhibited slightly less darkening than the surrounding areas. This effect can be seen ever so faintly in Figure 84.

Figure 83. Photographs of the samples after the UV exposure test.

Figure 84. Polyester sample after test.

Figure 85. Transmission spectra of the samples after UV exposure for 1934 hours.

Figure 86. Comparison of spectra before and after the UV exposure test.

After 1934 hours of testing the samples were placed back in the XR-260 and tested for another 1940 hours. The spectra after 3874 hours of testing are shown in Figure 87. Figure 88, Figure 89, Figure 90, and Figure 91 show the spectra for D1, D2, D3, and D4 before and after UV exposure. A tabulation of all initial and final optical data appears in Table 25.

Figure 87. Transmission spectra of the samples after UV exposure for 3874 hours.

Figure 88. Transmission spectra of sample D1 before and after the UV exposure test.

Figure 89. Transmission spectra of sample D2 before and after the UV exposure test.

Figure 90. Transmission spectra of sample D3 before and after the UV exposure test.

Aspen Aerogel Samples - Hemispherical Transmittance (%)
Sample D4, MFG ID No. FG Fiberglass

Figure 91. Transmission spectra of sample D4 before and after the UV exposure test.

Table 25. Aspen Aerogel Hemispherical Optical Data (Perkin Elmer 900 spectrophotometer).

Sample	Hemispherical Transmittance (%) at Room Temperature								
	INITIAL			1934 Hours			3874 Hours		
	550 nm	SWV	PWV	550 nm	SWV	PWV	550 nm	SWV	PWV
D1	48.93	50.49	49.33	32.43	33.79	32.53	33.18	35.21	33.33
D2	45.95	39.60	45.70	29.52	27.00	29.35	13.83	12.29	13.65
D3	43.19	21.76	42.80	24.14	12.83	23.86	17.37	8.67	17.22
D4	33.74	30.17	33.72	16.11	15.20	16.08	24.47	23.9	24.45

SWV = solar-weighted value

PWV = photopic-weighted value

All of the samples had lower transmission after the UV exposure test. One explanation for the decrease in transmittance is that the samples are no longer hydrophobic. NREL was able to do a quick hydrophobicity test on sample D1. Three photographs of the results are shown in Figure 92. The photo on the left shows the entire sample and the photo on the right shows a close-up view of the region of interest. In all the photographs, there are two “wetted” spots. The spot on the right was wetted in a region of a crack (the sample is quite brittle), and it appears that the water was able to seep into the sample via the crack, and created an opaque stain. The spot on the left was wetted over a noncracked region, and the hydrophobicity was maintained. After the water drop dried, the aerogel was undamaged.

Figure 92. Photographs of hydrophobicity test after 1934 hours of UV exposure.

The hydrophobicity test implies that only the surface of the samples are hydrophobic and that either the interior of the sample lost its hydrophobicity during the test or the hydrophobicity treatment during preparation of the aerogels did not completely penetrate the sample and the interior of the samples were never hydrophobic.

To answer some questions concerning the hydrophobicity of the aerogels, Aspen sent several additional samples to NREL for testing. The hydrophobicity of the samples was tested before they were sent to NREL to be sure that they were hydrophobic. The spectroscopy results for these samples are shown in Figure 93 through Figure 96. The data are summarized in Table 26.

Figure 93. Transmission spectra of P22008, P41408, and P51308 before and after the UV exposure test.

Figure 94. Transmission spectra of the P51308 before and after the UV exposure test.

Figure 95. Transmission spectra of P22008 before and after the UV exposure test.

Figure 96. Transmission spectra of P41408 before and after the UV exposure test.

Table 26. Aspen Aerogel Hemispherical Optical Data of P51308, P22008, and P41408 after 1940 hours (Perkin Elmer 900 spectrophotometer).

	Hemispherical Transmittance (%) at Room Temperature					
	INITIAL			1940 HOURS		
Sample	550 nm	SWV	PWV	550 nm	SWV	PWV
P51308	37.51	52.31	37.27	36.51	38.18	36.05
P22008	40.24	54.07	40.19	44.22	46.85	43.90
P41408	43.67	53.03	43.63	46.88	47.33	46.48

SWV = solar-weighted value

PWV = photopic-weighted value

The results for the UV exposure test for these samples are very confusing since the transmittance increased after the test. Most likely the increase in transmittance is due to instrumental error and there was no major change in the transmittance for these samples. The origin of the error is not known but may be related to the variation in thickness of the aerogels.

2.7.2.4. Summary of UV Stability Test Results

After exposure of the aerogel insulated IGU prototypes (samples D1-D4) to accelerated environmental testing for 3874 hours at 71-88 °C under simulated 1.1 sun illumination of an Air mass 1.5 solar spectrum, all devices exhibited very little visual degradation to the naked eye. However, tabulated hemispherical transmittance measurements showed a significant decrease in transmittance for all samples after UV exposure for 1934 hours, and the transmittance decreased further after another 1940 hours of exposure.

For the samples listed in Table 26, the hydrophobicity of the samples was measured after the UV exposure tests and the results are listed in Table 27. After UV exposure, all of the samples were less hydrophobic than before the test and exhibited some degree of yellowing in the middle. This yellowing was the highest for sample D1 and the lowest for the sample (P41408) reinforced with glass fibers. Previous experience indicates that hydrophilic aerogels lose transparency when exposed to the ambient conditions for longer than 3 months and could explain the decrease in transparency upon UV exposure.

Table 27. Hydrophobicity of samples tested at NREL after 1934 hours.

Sample	Reinforcement	Observation	Hydrophobicity
P51308	polyester	Water beads up, but leaves a trace	Fair
P22008	polyester	Bottom Face	Good
		Top Face	Fair
P41408	glass fiber	Top of coupon	Fair
		Bottom of coupon	Good
D1 (after 3874 hours UV exposure)	polyester	Bottom Face	Poor (crunchy, broken)
		Top Face	Hydrophilic (crunchy, broken)

2.8. Fabrication of Prototypes

We sent two aerogel blankets to Major Industries to be fabricated into prototype translucent windows. The properties of the aerogels are listed in Table 28. Major Industries fabricated a 2' x 3' translucent window. Two cells of the window were fabricated with 1 layer of S51308 polyester reinforced aerogel on each side of the window with standard 28 gram daylighting insulation in between the aerogel layers. The R value was 10.5 hr·ft²·°F/Btu and the transmittance was 20%. The other two cells were fabricated with 1 layer of FG41408 glass fiber reinforced aerogel on each side of the window with standard 28 gram daylighting insulation in between the aerogel layers. These two cells had an R value of 9 hr·ft²·°F/Btu and the transmittance was 20%. A schematic diagram of the insulated window is shown in Figure 97 and a photograph is shown in Figure 98.

Table 28. Properties of aerogel blankets sent to Major Industries.

Sample 2'x3'	TC (mW/mK)	Average thickness (mm)	R-value	Density (g/cc)	Areal Density (g/sq ft)	Transmittance (%)
S51308	12.3	6.8	3.13	0.110	72.3	58
FG41408	13.2	5.7	2.45	0.130	65.2	64

Figure 97. Schematic of the aerogel insulated translucent window.

Figure 98. Photograph of the aerogel insulated window fabricated by Major Industries.

The two different types of aerogel (polyester = P220 and glass fiber = FG414, 3 specimens of each) used in the daylighting window presented in Figure 98 were sent to Architectural Testing, Inc. (ATI) to have the thermal conductivity determined by ASTM C 518-02 per NFRC 101-2004. It was necessary to obtain certified data for U-value simulations that were conducted at Major Industries. The information in Table 29 was collected internally at Aspen Aerogels and is shown as a comparison to the certified data obtained at ATI that is presented in Table 30. The test conditions used to collect the data were as follows:

- Cold plate temperature 0 °F
- Warm plate temperature 70 °C
- Mean specimen temperature 35 °F
- Vertical heat flow (Down) – Horizontal specimen
- Heat flux transducer on warm side
- Specimen test size 12" x 12", metering area 4" x 4"

Table 29. Properties determined at Aspen for the aerogels sent to Architectural Testing.

Sample 12"x12"	TC (mW/m·K)	Thickness average (mm)	R-value	Density (g/cc)	Areal density (g/sq ft)	Transmittance * (%)
P220-1	12.1	3.7±0.6	1.73	0.120	40.8	57
P220-2	10.7	3.1±0.7	1.64	0.126	36.5	59
P220-3	11.8	3.3 ±0.3	1.58	0.125	38.3	58
F414-1	13.4	4.4±0.2	1.86	0.131	53.1	67
F414-2	13.0	3.5±0.2	1.53	0.135	43.5	70
F414-3	13.2	4.1±0.5	1.76	0.123	47.1	66

P = polyester and F = glass fiber

Table 30. NFRC 101-2004 certified ASTM C 518-02 thermal conductivity results determine by Architectural Testing.

Sample 12"x12"	Thickness (inches)	Density (lbs/ft ³)	Thermal conductance (Btu/hr·ft ² ·°F)	Thermal Resistance (hr·ft ² ·°F/Btu)	Thermal Conductivity (Btu-in/hr·ft ² ·°F)	Thermal Conductivity (mW/m·K)
P220-1	0.146	7.4	0.509	1.96	0.074	10.7
P220-2	0.122	7.7	0.631	1.59	0.077	11.1
P220-3	0.130	8.1	0.542	1.84	0.071	12.7
P220 Avg.					0.074	10.7
F414-1	0.173	7.8	0.471	2.12	0.081	11.7
F414-2	0.138	8.2	0.634	1.60	0.086	12.4
F414-3	0.161	7.6	0.544	1.84	0.088	12.7
F220 Avg.					0.084	12.2

Another one of the blankets sent to Major Industries was used to fabricate a prototype translucent window without grids. The properties of the aerogel are listed in Table 31. The window was fabricated with 1 layer of FG41408 glass fiber reinforced aerogel on one side of the window with standard 28 gram daylighting insulation filling the remainder of the cavity. The R value was

7.17 hr·ft²·°F/Btu and the transmittance was 24%. A schematic diagram of the insulated window is shown in Figure 99 and a photograph of the window is shown in Figure 100.

Table 31. Properties of aerogels for second prototype fabricated by Major Industries.

Sample 2'x3'	TC (mW/m-K)	Thickness average (mm)	R- value	Density (g/cc)	Areal density (g/sq ft)	Transmittance (%)
FG41408 2	13.2	5.6±0.7	2.40	0.131	63.5	64

Figure 99. Schematic of the aerogel insulated translucent window.

Figure 100. Photograph of a 2' x 3' aerogel insulated translucent daylighting window fabricated by Major Industries.

Aerogel blankets were also sent to Traco who fabricated IGUs and a skylight. The IGUs and the finished skylight and shown in Figure 101 and Figure 102. Based on feedback from Traco, they would prefer thicker battings (~ 0.5 inches) with a smooth, “non-wavy” surface.

Figure 101. IGUs fabricated by Traco.

Figure 102. Finished skylight fabricated from aerogel insulated IGUs.

3. Conclusions

During BP1, Aspen prepared transparent aerogels from TMOS by adding drying additives to help control the drying process. These additives helped to prepare monolithic transparent aerogels that were 1 inch thick. We improved the transparency of aerogels by using a modified silica precursor for silica aerogels and were able to prepare aerogels that were >90% transparent. Although, we improved the transparency of the aerogels, all attempts to make large panels failed. These results suggested that it would be difficult and expensive to manufacture monolith panels of transparent aerogels to make transparent windows. In addition, feedback from windows manufactures indicated that even 1% haze in the aerogel would be unacceptable to consumers.

Therefore, a market study was undertaken to determine the size of the market for translucent fenestration products. The market study concluded that the size of the daylighting market could range between 123 and 371 million square feet of glazing in North America. Capturing a significant portion of the available market for an aerogel insulated product would depend on many factors including energy economics (which should always be favorable for aerogel) and the transmittance of an aerogel insulated product which was identified at or above 0.35 VT. The study also concluded that there is a sizable market for translucent daylighting products, a decent sized early entry niche market, and a short, responsive path to this market. Therefore, translucent aerogels would be a viable product to develop.

Based on the market study, daylighting applications appeared to be the most appropriate market for translucent aerogel-based windows. Therefore, Budget Period 2 focused on preparing translucent aerogels for the skylight and daylighting markets, where energy savings potential is significant, and transparency criteria are not as stringent. During Budget Period 2, Aspen developed formulations using TEOS that were almost as transparent as the aerogels prepared using TMOS and used these formulations to prepare fiber reinforced aerogels that were translucent. Two 1' x 1' aerogel panels were prepared and used to make insulated glass units (IGUs) that were sent to LBNL and the NREL for testing.

Simulations using WINDOW 5 and THERM 5 showed that a double glazed window insulated with a 0.5 inch thick aerogel between clear glazings would be expected to have a U value of 0.77 W/m²K (0.136 BTU/ft² h F) and a SHGC of 0.618 and with a low-e glazing the U value would be 0.77, but the SHGC decreased to 0.365. The spectroscopy results conducted by NREL indicated that an aerogel insulated glazing unit fabricated with low-e glass is a viable option.

Budget Period 3 focused on scale-up of the fiber reinforced translucent aerogel panels identified during Budget Period 2. Aspen Aerogels successfully prepared aerogels with excellent transmittance from TEOS and other ethylsilicate precursors using ethanol as the solvent and used these aerogels to develop a low cost method for producing fiber reinforced translucent aerogels with a VT of 60-70% that are 7-12 mm thick and have a R factor of 10/inch. With further development, the process developed could be scaled-up to make production quantities of translucent aerogel blankets to manufacture translucent aerogels for use in fabricating superinsulated windows for daylighting applications.

Prototype IGUs were fabricated, and after exposure of the aerogel IGU prototypes to accelerated environmental testing for 3874 hours at 71 - 88 °C under simulated 1.1 sun illumination of an air mass 1.5 solar spectrum, all the IGUs exhibited very little visual degradation to the naked eye. However, tabulated hemispherical transmittance measurements showed a significant decrease in transmittance for all samples after exposure for 1934 hours, and the transmittance decreased further after another 1940 hours of exposure. After exposure to the simulated sun illumination, all of the samples were less hydrophobic than before the test and exhibited some degree of yellowing in the middle. Previous experience indicates that hydrophilic aerogels lose transparency when exposed to the ambient conditions for longer than 3 months and could explain the decrease in transparency upon UV exposure. These results indicate that a more stable hydrophobe will be required to meet the lifetime requirements for an aerogel insulated fenestration product.

Aspen Aerogels successfully met the overall objective for this project and developed aerogel transparent insulation materials for fenestration systems that could be produced economically. The developed fenestration products increased the thermal performance compared to existing solutions, while satisfying constraints such as cost, user acceptance, size limits, and environmental, safety, and health concerns. As indicated above, the durability needs to be improved to meet the lifetime requirements.

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