

# ENERGY EFFICIENT AND DURABLE SKYLIGHTS AND ROOF WINDOWS



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Energy and Transportation Science Division

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## **1. Abstract**

Fenestration systems play essential roles in building aesthetics, the most important roles being visual and thermal comfort. Fenestration products are not as thermally efficient as fully insulated building walls and roofs, therefore, they are the main source of heat loss/gain from the building envelope. Various technologies are used to increase the insulation performance of fenestration glazing, including the introduction of a vacuum or an inert gas between the glass panes and the use of triple or quadruple panes. Each of these solutions has its own disadvantages—for example, leakage, thermal stress, the high cost for vacuum between panes, and leakage and higher convection for gas between panes. Plastic or glass capillaries or honeycomb structures are good candidates for enhanced transparency and thermal insulation. Plastic becomes unstable at higher temperatures and glass increases the glazing unit weight. Moreover, these structures need wide ( $\geq 5$  cm) interspace, which requires special sealing and spacers, and thus becomes incompatible with the current infrastructure. Aerogel is considered to be a state-of-the-art transparent insulating material, but haziness in aerogel-filled panes due to light scattering and the high cost of manufacturing aerogels hinders its commercialization. Therefore, a material that provides aerogel like insulation but with dramatically reduced haziness and potentially lower production cost is highly desirable. The work performed under this project addressed most of the above challenges by developing a low cost thermal insulation material with better visible transparency.

## **2. Statement of Objectives**

The goal of this project was to develop a transparent thermal insulation material with visible transmittance  $\approx 80\%$  and thermal conductivity  $\approx 0.032$  W/m.K. The main scientific principle that was exploited to achieve transparent thermal insulation material was to keep the size of individual units of material small enough to avoid the visible light scattering, and to minimize the heat transfer resulting from convection of air by reducing the pore size. The first year focused on exploring the different synthesis strategies and parameters to achieve the porous particles with sufficient air volume, low thermal conductivity, and minimal visible light scattering. The second year focused on developing a transparent thermal insulation material and optimization of the development process.

## **3. Benefits to the Funding DOE Office's Mission**

Buildings consume about 40% of the primary energy in the United States, and energy loss through windows comprises 10-15% of that 40%. Various DOE and privately funded scientific research, reports, reviews, and surveys has shown that there is a continuing need for low cost transparent thermal insulation technologies that can increase the insulation of fenestration systems such as windows, moon roofs, and skylights. In many states, new regulations and codes require the use of Energy Star Windows in new building construction. Several tax deductions or credits have been offered to the residents for using Energy Star windows in new houses. This combination of scientific research, local code and regulation requirements, incentive programs, and third-party ratings, have made energy efficient windows and skylights an important approach for improving the building energy efficiency in the United States and all over the world. The technology developed under this project will also help in achieving Building Technologies Office's (BTO's) overall goal of reducing energy consumption in buildings by 50% by 2030.

## **4. Technical Discussion of Work Performed by All Parties**

The technical objective of the project was to develop a low cost transparent thermal insulation material that can be scaled up at industrial scale. Oak Ridge National Laboratory (ORNL) and VELUX Design and Development Company USA, LLC developed a low cost and transparent thermal insulation material.

ORNL and VELUX synthesized, characterized, and scaled up porous silica particles that can be further employed in developing different types of thermal insulation materials. ORNL developed the materials and VELUX provided the inputs for material cost analysis, potential challenges in material characterization, and the prototype window units. ORNL addressed the challenges facing a successful commercialization of the technology by devising new strategies. The following section discusses the technical work in more detail:

## **FY Year 2014**

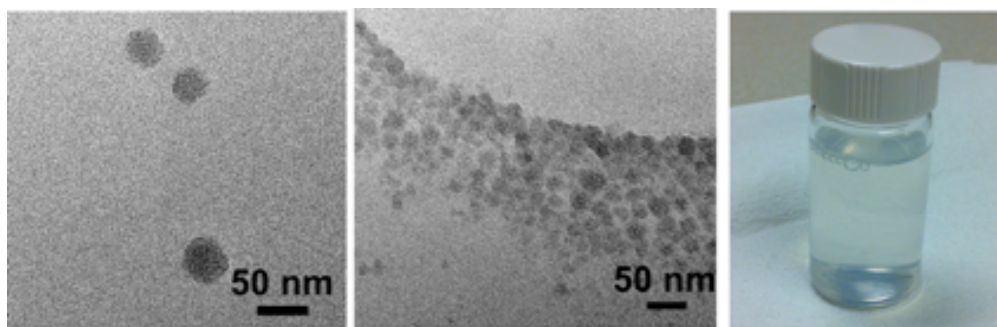
### *Synthesis of small sized silica particles:*

The objective of using porous silica particles to create the transparent insulation material is to reduce the heat transfer by thermal conduction through solid silica. Solid conduction decreases as the density of solid silica decreases or, in other words, as air volume increases. The actual conductivity of the solid does not change, but the area of the solid in a cross section of the material decreases, which lowers the solid conduction per square meter. However, it must be noted that pores should have a vacuum or be filled with a lower thermal conductivity material (e.g., air), not with a material that has higher conductivity than silica.

The basic principle of using a small pore size is that convection of air among the particles will be prevented because of the Knudsen effect; i.e., a pore size smaller than the mean free path of air results in any energy transfer being between the air molecules and the particle wall, rather than among air molecules. Thus, decreasing the pore sizes of the insulating material with air-or gas-filled cavities will decrease the thermal conductivity of air/gas. The elastic collisions between gas molecules and the solid particle will transfer less energy than collisions between gas molecules. Smaller pore sizes create a higher probability of collisions with pore walls than with other gas molecules.

When light interacts with a structure, a portion of it gets absorbed, reflected, transmitted or scattered. Rayleigh scattering is the main scattering related to the particles smaller than the wavelength of visible light (400–700 nm). It is proportional to the sixth power of particle diameter. Hence, by decreasing the particle size (interacting unit) below 100 nm, light scattering similar to the single glass pane can be achieved.

The porous particles of very small size and with high air volume were synthesized by dissolution of a polymer in a water and ethanol solution. This polymer made small micelles. Deposition of silica occurred around these micelles. Heating the particle powder at 550 °C for 4 hours produced the porous particles of 60 nm – 100 nm diameter. Figure 1. shows the transmission electron microscope (TEM) image of particles.



**Figure 1:** TEM images of very small porous silica particles (right image shows an as synthesized sample in ethanol).

In order to measure the pore size of the above porous particles, Barrett-Joyner-Halenda (BJH) analysis was used. Particles were small in size and contained very small pores in the range of 1.5 –3.0 nm.

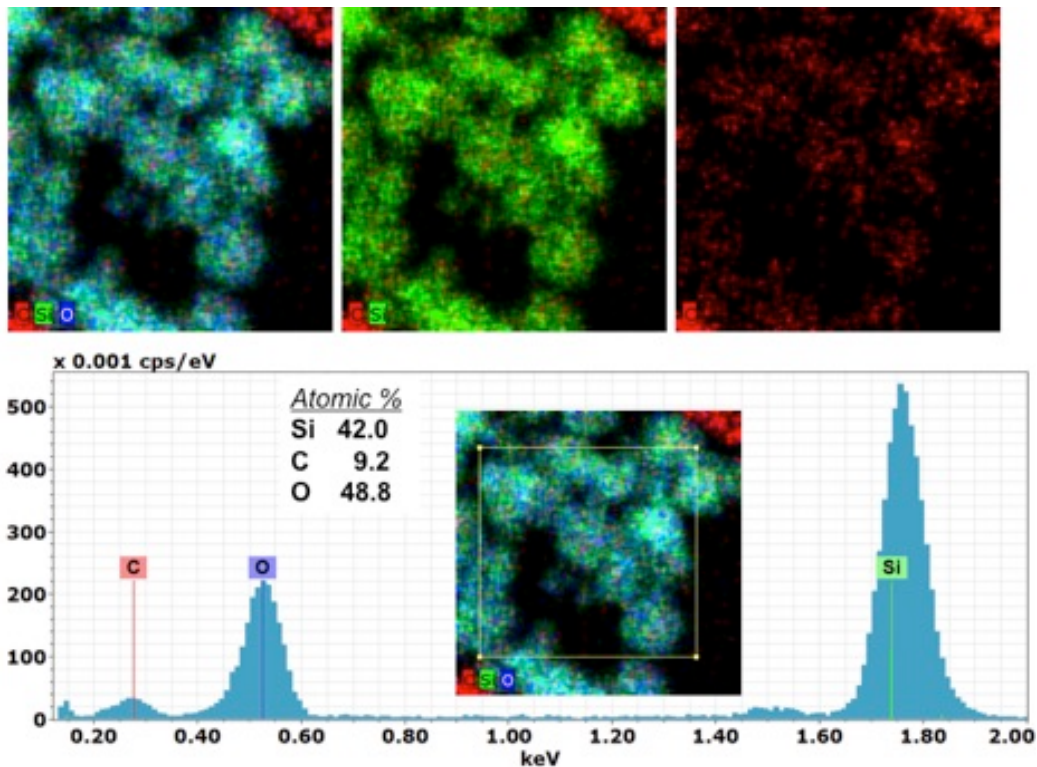
In order to measure the density of particles, we took a 1 cm<sup>3</sup> cube of particles and weighed the sample. We found that the density was = 0.3g/cm<sup>3</sup>. The density of solid amorphous silica is 2.2 g/cm<sup>3</sup>.

Therefore, the percentage of solid silica per cm<sup>3</sup> in the present sample was  $0.3\text{g}/2.2\text{g} \times 100 = 13.63\%$  and the % of air volume was  $100 - 13.63 = 86.36\%$

A heat flow meter apparatus was used to measure thermal conductivity of the sample. The thermal conductivity value was 0.037 W/m.K, which is representative of an *R*-rating of  $\approx 3.93/\text{inch}$ .

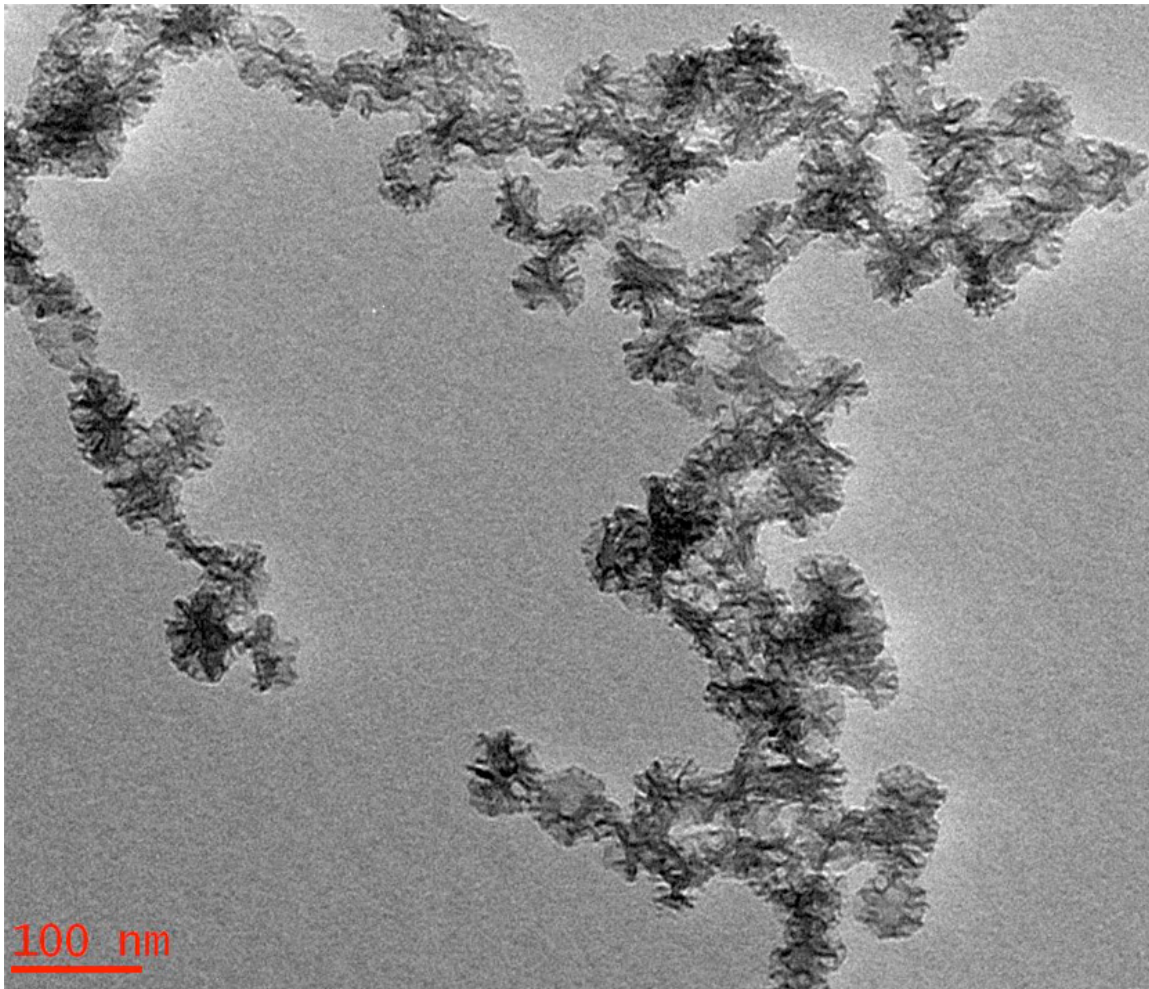
The possibility exists for a sizeable amount of carbon to remain in the particles following removal of the polymer by heating. This carbon may enhance the thermal conductivity of the overall material.

To investigate this hypothesis, we did energy dispersive X-ray spectroscopy (EDS) studies and found that there was some carbon still present but not enough to enhance the thermal conductivity significantly. Figure 2 shows the EDS images (top) of particles with distinct color for C, O, and Si; and EDS plot (bottom) shows the atomic % of C, O, and Si in the porous particles.



**Figure 2:** EDS images of the porous particles with elemental maps and atomic % of C, O, and Si in the porous particles.

To further increase the air volume in particles, we synthesized particles with large pore sizes (4-5 nm). Figure 3. shows TEM image of large pore size porous silica particles.



**Figure 3:** Porous silica particles with an average diameter  $\approx 60$  nm, and average micropore size of 3.5 nm.

*Scale up and thermal conductivity measurements:*

We scaled up the synthesis (just by using a large container for the reaction) to fill the 0.5" x 6" x 6" sample mold. The scaled up material proved that the syntehsis process can be adaped for industrial scale production of particles. The scaled-up material is shown in Figure 11.



**Figure 4:** Scaled up porous particles.

The thermal conductivity measurements were done by using heat flow meter.

Outcome: Thermal conductivity = 0.03293 W/m.K

This proved that by increasing the pore size (overall pore or air volume) inside the material, the thermal conductivity of bulk material can be further reduced to provide better thermal insulation.

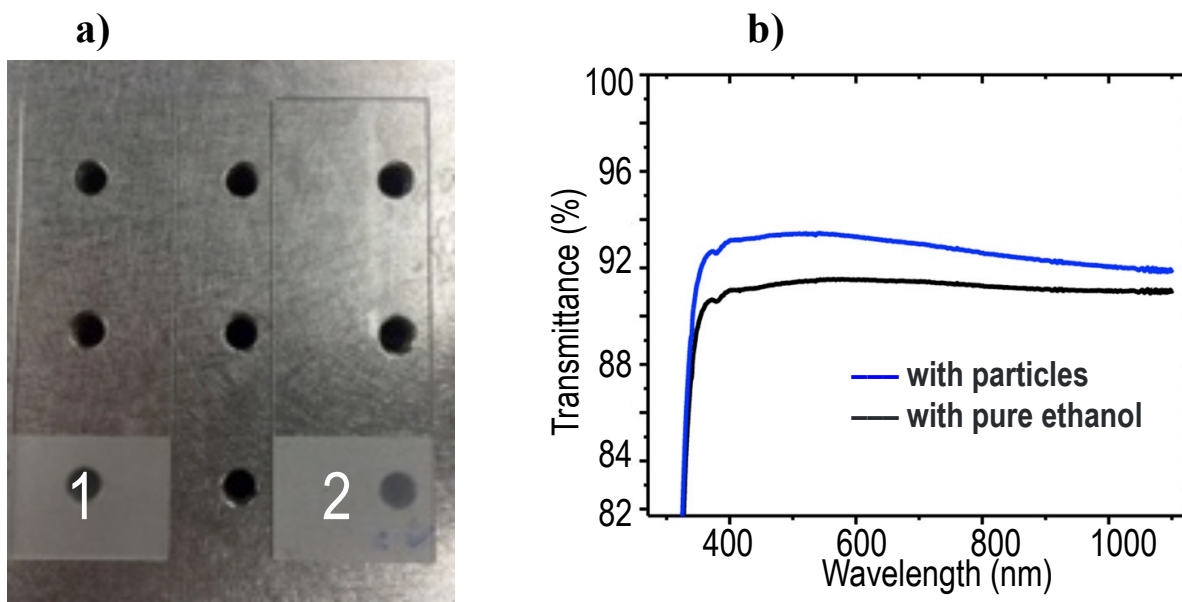
#### *Optical properties measurements*

Light scattering measurements of silica particles is quite complex. Several approaches were tried to measure the visible transmittance and scattering.

#### *Transmittance spectra of silica particle layers*

We dissolved the particles in ethanol, and then sonicated the solution in order to break the larger aggregates of particles. We then centrifuged the solution to separate and remove the largest aggregates. The solution with individual well-dispersed particles was then used for the transmittance measurements.

Two slides, one with the above particle solution (Figure 5a-1) and other with ethanol alone (Figure 5a-2), were coated using spin coating method. UV-vis spectra were obtained on a Shimadzu UV-1800 spectrometer. The measurement wavelength range was 190– 1100 nm and testing was performed in the transmittance mode. The wavelength-scanning rate was set to 0.5 s and the data were recorded at 1 nm intervals. Figure 5, (a) shows the slides coated with particle solution (1) and pure ethanol (2), and b) transmittance spectra of these slides.



**Figure 5:** a) Slides coated with particle solution (1) and pure ethanol (2), b) transmittance spectra of these slides.

A visual comparison between the slide coated with particles and the slide without particles fails to detect the appearance of any light scattering behavior that would be attributable to the presence of particles. Moreover, the transmittance spectra (Figure 5) indicate that light scattering by the particles is essentially non-existent. Scattering by sub-1000-nm particles should sharply attenuate transmission as the wavelength decreases, but no decrease was observed. In fact, the transmittance increased all wavelengths, a phenomenon that is attributable to the antireflective (refractive index matching) characteristics of the particles.

#### *Measurements of diffuse scattering*

Diffuse scattering in visible wavelength band (400-800 nm) from silica nanoparticles was measured using a Cary 5000 dual-beam spectrophotometer with a diffuse reflectance accessory.

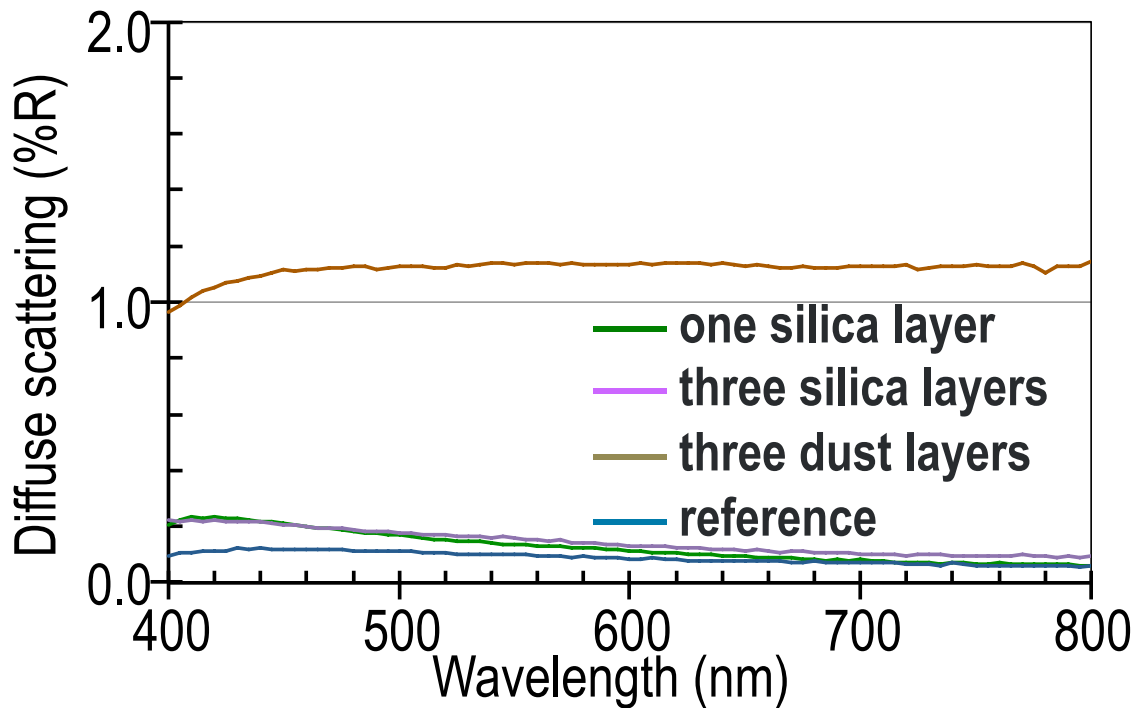
Scattering specimens were prepared by spin coating silica dispersed in ethanol on mirrored-glass substrates. The mirrors were silvered first surface optics, 30-mm x 30-mm square, Edmund Optics part number 84-442. Mirrors were cleaned prior to spin coating using Formula 409 cleaner followed by rinsing with distilled water and isopropyl alcohol. The surfaces were wiped dry using an optical tissue to eliminate spotting.

Spin coating was done at 1000 rpm for 60 seconds, which was fast enough and long enough to spread silica evenly without spotting. Prior to wetting, mirrors were spun once with 0.25 ml of ethanol to wet the mirror surface and reduce surface tension of the silica-dispersed solution. One mirror was coated with a single layer of the silica solution (0.25 mL), while another was coated with three layers (3 x 0.25 mL). The mirrors were allowed to air dry and were then stored in plastic specimen holders to protect the coating of deposited silica. The presence of a silica coating on each mirror was visible to the naked eye, but there was no discernible difference between the appearance of the single-layer mirror and the three-layer mirror. A third mirror was “coated” with a layer of a control liquid (100% ethanol). This mirror

served as the scattering reference. A fourth mirror was spin-coated with three layers of Arizona Test Dust (ISO 12103-1 A4, coarse) which has a particle size distribution in the range 1-300  $\mu\text{m}$ , with a peak in the distribution at 50-60  $\mu\text{m}$ . The dust is 68-76% silica and 10-15% alumina.

The diffuse scattering from each mirror was measured twice in the spectrophotometer. The incident beam was nearly normal to the surface, and the specular reflectance within a cone with a  $3^\circ 20'$  half angle from the normal was excluded from the detected scattered light.

The results of the diffuse scattering measurements are shown in Figure 6. The scattering from the single-layer and triple-layer silica is statistically indistinguishable from one another. The diffuse scattering at the longer wavelengths is identical to that of the reference mirror, and it increases monotonically with decreasing wavelength until it rises about 0.5% above the scattering of the reference mirror at 400 nm. The scattering is possibly due to agglomeration of the silica particles on the surface of the mirror that occurs as the solvent evaporates. The wavelength dependence is indicative of this phenomenon; the scattering increases as the wavelength approaches the size of the agglomerated particles. It is very difficult to prevent free nanoparticles from agglomerating due to electrostatic charges that gather on the particles. The diffuse scattering of the dust coated mirror is more than five times greater than that of the silica coated mirrors, and the amount of scattering is nearly constant across the spectrum, which is what we would expect for an even distribution of particles that are significantly larger than the wavelength of incident light. Results are shown in Figure 6.



**Figure 6:** Diffuse scattering measurement of silica particles.

From the above experiments, it becomes clear that the unaggregated particles do not cause significant scattering of light. An estimate of the total loss in transmission due to scattering is  $<0.5\%$ .

## FY Year 2015

### *Drying the silica gel without supercritical technique*

The second year mainly focused on finding ways to dry the silica gel in a cost effective way to make a silica aerogel. During the formation of a porous material, the main challenge is to remove the solvent from inside the pores while minimizing the collapse of the pores. A typical strategy for removing solvent is supercritical drying, but supercritical drying in a commercial application could be quite expensive. Therefore, the biggest challenge is to find an inexpensive strategy that can remove the solvent from inside the pores while keeping the pore network intact. Different strategies involving mixing the two solvents, use of low surface tension solvents, direct evaporation of solvent, and controlled evaporation of solvent were explored. Direct evaporation resulted in pieces of silica aerogel (Figure 7), while controlled evaporation of solvent that includes slow evaporation under constrained condition provided better results and kept the silica gel intact though with some cracks (Figure 8).



Figure 7: A silica gel after drying by direct evaporation. Gel breaks into small pieces.



Figure 8: A silica gel after drying using controlled evaporation method.

## **5. Subject Inventions (As defined in the CRADA)**

### *Patent applications:*

J. Sharma, M. Bhandari, P. Datskos, G. Polyzos, C. J. Rimsky, Hybrid silica gel made of porous silica particles, PCT Appl. # PCT/US17/34221, May 24, 2017

### *Invention Disclosures:*

1. Lightweight, mechanically strong, transparent, and highly thermally insulating barrier layer, ORNL invention disclosure # 201503607
2. Low cost, multilayer, highly transparent and thermally insulating hybrid silica-polymer film, ORNL invention disclosure# 201503608
3. Hybrid silica gel made of porous silica particles, ORNL Invention disclosure # 201603663
4. Semiconductor having thermal conductivity lower than that of aerogels, ORNL invention disclosure # 201603687
5. Hybrid hollow silica particles with unprecedentedly low thermal conductivity ORNL invention disclosure # 201603719

## **6. Commercialization Possibilities**

The development of transparent thermal insulation materials can benefit several areas in residential and commercial buildings, automotive industry, and aerospace industry. Our Industry partner VELUX is developing transparent skylights. The developed technology can be commercialized, however, further studies to understand its mechanical, optical, and thermal properties, are required for its commercialization.

## **7. Plans for Future Collaborations**

Further reduction in thermal conductivity of porous particles is required. Similarly, mechanical and thermal properties of newly developed transparent thermal insulation material are needed to be tested in more detail. Similarly, scale up the materials, enhance its transparency, and mechanical strength can be performed as a new collaboration.

## **8. Conclusions**

During this CRADA, several strategies to synthesize porous particles and control their size were explored. Porous particles that can provide a thermal conductivity  $\approx 0.032$  W/m.K were synthesized. Several strategies to dry the silica aerogels were investigated, and finally a room temperature and ambient pressure strategy was selected to dry the aerogels. A new type of transparent thermal insulation material that can provide better mechanical properties was developed. More optimization of the solvent removal from the pores of insulation material and detailed testing of its mechanical and thermal properties is still required.