Streamlined Life Cycle Assessment of Transparent Silica Aerogel Made by Supercritical Drying

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Abstract

When developing sustainable building fabric technologies, it is essential that the energy use and CO_2 burden arising from manufacture does not outweigh the respective in-use savings. This study investigates this paradigm by carrying out a streamlined life cycle assessment (LCA) of silica aerogel. This unique, nanoporous translucent insulation material has the lowest thermal conductivity of any solid, retaining up to four times as much heat as conventional insulation, whilst being highly transparent to light and solar radiation. Monolithic silica aerogel has been cited as the 'holy grail' of future glazing technology. Alternatively, translucent granular aerogel is now being produced on a commercial scale. In each case, many solvents are used in production, often accompanied by intensive drying processes, which may consume large amounts of energy and CO_2. To date, there has been no peer-reviewed LCA of this material conducted to the ISO 14000 standard.

Primary data for this 'cradle-to-factory gate' LCA is collected for silica aerogel made by low and high temperature supercritical drying. In both cases, the mass of raw materials and electricity usage for each process is monitored to determine the total energy use and CO_2 burden. Findings are compared against the predicted operational savings arising from retrofitting translucent silica aerogel to a single glazed window to upgrade its thermal performance. Results should be treated as a conservative estimate as the aerogel is produced in a laboratory, which has not been developed for mass manufacture or refined to reduce its environmental impact. Furthermore, the samples are small and assumptions to upscale the manufacturing volume occur without major changes to production steps or equipment used. Despite this, parity between the CO_2 burden and CO_2 savings is achieved in less than 2 years, indicating that silica aerogel can provide a measurable environmental benefit.

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1.0 Introduction

This study forms part of a systematic approach to improving UK building fabrics while considering both embodied impacts and operational savings - a critical balance that is typically ignored. A number of innovative insulation technologies have been developed to satisfy the growing demand for energy efficient buildings. Finding a balance between thickness, cost and in-situ performance is essential, particularly for measures that rely on sustaining a vacuum. Above all, however, technologies must provide a measurable benefit over their lifecycle i.e. the in-use savings must not be outweighed by the respective energy and CO_2 burden arising from manufacture, transport and end-of-life processing. This study investigates this paradigm by carrying out a streamlined 'cradle-to-factory gate' life cycle assessment (LCA) of transparent silica aerogel following the environmental standard BS EN ISO 14044:2006 [1]. Silica aerogel is an emerging super insulation material, rapidly gaining interest within the new build and refurbishment markets. Despite many solvents being used in silica aerogel production, often accompanied by intensive drying processes, which may consume large amounts of energy and CO_2, there has been no peer-reviewed LCA of this material conducted to date.

1.1 What is Aerogel?

Aerogels are synthetic low-density materials with unique physical properties [2]. They are formed by removing the liquid from a gel under special drying conditions, bypassing the shrinkage and cracking experienced during ambient evaporation [3]. This creates a solid three-dimensional nanoporous structure, containing 80-99.8% air [4,5]. Due to their high porosity, aerogels exhibit the lowest thermal conductivity of any solid, whilst being transparent to light and solar radiation [6,7]. Aerogels are often cited as a promising material for translucent insulation applications [2-7]. The material can be produced in monolithic or granular form. Commercial products for the building sector include cavity insulation, glazing units and cladding systems containing granular aerogel, along with translucent and opaque insulation boards, blankets and tensile roof membranes embedded with aerogel particles. Alternatively, transparent monolithic silica aerogel has been cited as the 'holy grail' of future glazing technology, with potential to achieve U-values as low as 0.1 $W/m^2.K$ [8]. Current research and development into monolithic glazing is limited by the high cost of production, long processing time and difficulty creating large uniform samples with complete transparency [9].

1.2 How is it Made?

Aerogels were first reported by Samuel Stephens Kistler in the early 1930's [10]. Kistler aimed to test the hypothesis that "liquid inside a jelly can be replaced by a gas with little or no shrinkage".

Kistler's three-step experiment began by preparing a porous 'sol-gel' (a rigid body containing continuous solid and liquid networks) using sodium silicate and hydrochloric acid. This 'hydrogel' (where the liquid in the pores is water) was then soaked in alcohol several times over a 1-2 week period to strengthen the gel, causing the water inside the pores to be displaced. The resultant 'alcogel' (pores containing alcohol) was dried inside an autoclave using supercritical drying. The temperature and pressure of the autoclave were simultaneously raised to 270 ^{o}C and 100 bar, causing the alcohol to become supercritical (i.e. it begins to vaporise without completely changing phase due to the high pressure). As a result, the alcohol gains properties of both a liquid and a gas, eliminating surface tension inside the gel and enabling the fluid inside the pores to drain out without collapsing the solid structure. The newly formed 'aerogel' (pores containing air), could be safely handled when cool. The material possessed a low density and was opalescent. Kistler stated that numerous other materials had been successfully prepared, and that aerogels could be made from practically any material. Nowadays, silica aerogel is still the best-known and most widely prepared aerogel [11].

Up until mid 1980, risks associated with supercritical drying of alcohol were major obstacles to high volume aerogel production [12]. However, improvements in the manufacturing processes have yielded more cost effective aerogels that are economic to produce on a commercial scale [2,6,13,14]. The process has three steps: gel preparation, ageing and drying. Drying takes place through either high temperature supercritical drying (HTSCD), low temperature supercritical drying (LTSCD) or ambient pressure drying.

The most common technique for gel preparation involves reacting a silicon precursor, such as sodium silicate ("water-glass"), tetramethoxysilane (TMOS) or tetraethoxysilane (TEOS) with water in a solvent such as ethanol or methanol at ambient temperatures and pressures [11], forming silica nanoparticles. Gelation occurs when enough silica nanoparticles agglomerate to form a continuous network spanning throughout the entire volume of sol. Once the gel is prepared, it must be aged through a solvent exchange process to strengthen the gel network and prevent cracking during drying. This is achieved by soaking the gel within a pure organic solvent, usually methanol, ethanol or acetone for at least 24 hours. The solvent is replaced each time equilibrium in concentration is reached – typically, this step is repeated 3-4 times.

High temperature supercritical drying was originally used by Kistler to dry aerogel. It relies upon heating and pressurising the wet gel to ~240 ^{o}C and ~100 bar, i.e. the conditions that transform the alcohol within the gel into a supercritical fluid. The process can be dangerous if proper safety precautions are not taken. In 1984, the Airglass laboratory in Sweden was destroyed due to an autoclave leaking out 1000 litres of explosive methanol [11].

Low temperature supercritical drying was developed in the mid 1980s [15]. Here, the solvent inside the wet gel is replaced with liquid CO_2 prior to drying, as it possesses a critical point closer to ambient temperature. Drying therefore takes place at ~40 ^{o}C and ~100 bar, making the process more viable for commercial production. To increase the efficiency of production, supercritical CO_2 can be substituted instead of liquid CO_2. CO_2 recycling can also occur [14].

Ambient pressure drying, also called 'subcritical drying' emerged in the mid 1990s [6,13]. This process involves chemically modifying the surface of a wet-gel so that it becomes hydrophobic prior to drying. When dried ambiently, the gel partially collapses but re-expand to 85% of its original volume, since the internal network does not stick together. Gels dried by ambient pressure drying typically have 50-80% denser porosities than supercritically dried aerogels, thus are less transparent but mechanically stronger [11].

1.3   What is its Environmental Impact?

According to Lawrence Berkeley Laboratory, the production and use of silica aerogels is environmentally benign, the product is non-toxic, non-flammable, and it can be easily recycled [16]. Conversely, according to manufacturing studies, silica aerogel requires reasonably toxic chemicals, diffusion-controlled processes that consume a lot of solvent, and depending on the drying process, high-pressure vessels running for a long time [4,11].

Two major manufactures of silica aerogel are Cabot Corporation and Aspen Aerogels. Cabot produces translucent granules and insulation blankets via ambient pressure drying. Aspen produces opaque insulation boards and blankets embedded with silica aerogel particles via LTSCD. In 2008, both companies received a 'Silver' Cradle-to-Cradle environmental award from McDonough Braungart Design Chemistry (MBDC) for their aerogel production. MBDC claim to evaluate a products complete formulation, energy use, water use and recycling potential when assessing environmental impacts [17]. Unfortunately, data from these studies is confidential, making it difficult to assess the rigour and validity of the results. Moreover, as the

MBDC Cradle-to-Cradle programme does not undergo third party certification, it does not comply with the ISO standards for life cycle assessment [18].

At present, the only data on embodied energy and CO_2 of silica aerogel comes from Aspen Aerogels opaque insulation blankets. According to the manufacturers, its production energy is 53.9 MJ/kg and its CO_2 burden is 4.3 $kgCO_2$/kg, excluding CO_2 used for supercritical extraction as it is recovered from external industrial processes. Compared to conventional insulation, these values are reasonably high. According to the University of Bath's Inventory of Carbon and Energy [19], the production energy and CO_2 burden in organic insulation ranges from 3.5-26.8 MJ/kg and 0.2-1.7 $kgCO_2$/kg respectively. Contrarily, mineral insulation ranges from 16.6-38.8 MJ/kg and 1.1-1.4 $kgCO_2$/kg respectively. Oil derived insulation ranges from 70-98.3 MJ/kg and 2.5-3 $kgCO_2$/kg. Note that available data for double-glazing indicates that production energy and CO_2 burden can be much higher at 360-5470 MJ/m^2 and 18-279 $kgCO_2/m^2$, depending on the frame type and gas fill. Nonetheless, it is interesting that Aspen do not disclose the amount of CO_2 required during supercritical extraction of their aerogel. According to the manufacturers, this CO_2 is a recycled waste product recovered from ethanol and ammonia production plants. Evidently, the actual amount of CO_2 used is unclear, thus highlighting a need for further investigation.

2.0 Streamlined LCA

Life cycle assessment (LCA) is a process by which the environmental impacts associated with a product can be quantified over its lifecycle from 'cradle-to-grave'. To date there have been no peer-reviewed life cycle assessments of silica aerogel meeting the ISO 14000 standards. This study addresses this issue by conducting a streamlined LCA following BS EN ISO 14044:2006.

2.1 Goal

The aim of this study is to establish the CO_2 and energy costs associated with two different methods for manufacturing silica aerogel. Data will be compared against potential CO_2 and energy savings when retrofitting aerogel to building fabrics in-situ. The purpose of this investigation is to identify whether the production costs of silica aerogel can be recovered by its operational savings within a realistic product lifespan. The study will also serve to provide a unique comparison between two (of the three) methods of aerogel production.

The intended audience for this study includes environmental engineers, architects, materials scientists and product designers. Results are intended to be publicly available. It is anticipated that results may be compared against the life cycle impacts of conventional and emerging building fabric technologies. All comparisons must recognise that the results of this study are based on a laboratory experiment, scaled-up to produce 1 m^3 volumes. Scaling assumptions must be treated as conservative estimates for commercial production due to the lack of information from industry concerning the actual economies and efficiencies of scale associated with mass production.

2.2 Scope

This study is a 'cradle-to-factory gate' assessment. Primary data is collected for two methods of aerogel production. Secondary data is used to account for the energy use and CO_2 burden from extracting raw materials, as well as the grid intensity of electricity. The impact of transport (of raw materials / finished products) and end of life processing (e.g. product re-use, recycling, landfill etc) are omitted from this study. However, their significance is discussed.

At the University of Bath, aerogels are produced using both low and high temperature supercritical drying for research into optical applications. We made use of their experience in aerogel production to conduct two studies, monitoring the CO_2 and energy usage associated

with manufacturing small samples of silica aerogel using both drying techniques. It should be noted that there is a third method for aerogel production, via ambient pressure drying. Currently the group does not produce aerogels in this way.

The results of this study should be treated as a conservative estimate for the production cost of aerogel. The processes developed at the University of Bath have not been developed or refined for mass manufacture. As such, no recycling of solvents or CO_2 occurs. Furthermore, just 40ml of aerogel is produced during each production run. The aerogels are solid (not granular) and have a high optical quality. Scaling assumptions to upscale the manufacturing volume, without major changes to production steps or equipment must be treated with caution.

The functional unit for this investigation is the energy use (kWh) and CO_2 burden (kgCO$_2$) required to produce 1 m^3 of aerogel. Results are compared against the operational energy and CO_2 savings arising from retrofitting a 1cm thick, 1 m^2 twin-wall polycarbonate panel filled with aerogel granules to a single glazed window. The predicted performance of this product over a 15 year lifespan is estimated based upon the results of in-situ testing, carried out by the corresponding author prior to this streamlined LCA [20].

3.0   Data Collection

Data collection is split across three stages: Gel preparation, ageing and drying. Gel preparation took place during April 2010. Following this, gels were aged in solvent for 3 weeks, and then supercritically dried. Figure 1 displays the system boundary for both methods of aerogel manufacture studied. Monitoring procedures and omitted factors are outlined.

System Boundary for Manufacturing Study:

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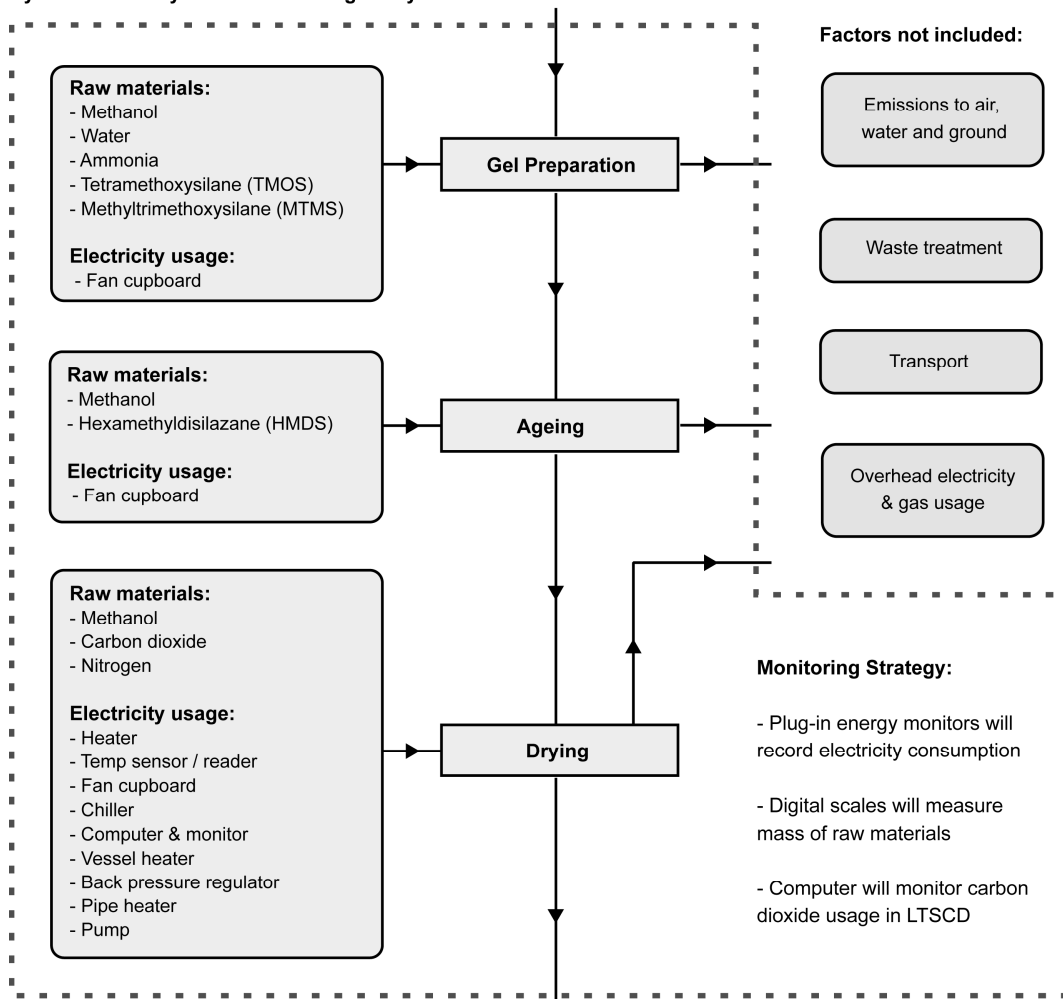
[Figure 1: System boundary and monitoring strategy for manufacturing study]

3.1   Gel preparation

The first stage of aerogel production involved mixing the chemicals together at the correct proportions, inside a ventilated fume cupboard. Approximately 40 ml of solution was prepared for both drying methods. The HTSCD samples were prepared in 4 glass test tubes, and the LTSCD samples were prepared in 18 smaller plastic cuvettes. Approximately 10-12 minutes after the raw ingredients were mixed the samples became rigid alcogels. The mass of all raw ingredients was measured using digital scales and the electricity use of the fume cupboard was logged using an 'Eco-Eye Plug-in Energy Monitor', displaying the rate of energy use (W) and the total energy use (kWh). Table 1 and Table 2 show the respective data collection inventories for the HTSCD and LTSCD samples. Note that gel preparation time, and consequent electricity usage was higher than normal, due to time spent weighing each ingredient on the digital scales.

Raw materials used in gel preparation (HTSCD)	Volume (ml)	Mass (g)	Material Supplier
Tetramethoxysilane	14.4	14.64	Fisher
Methyltrimethoxysilane	1.6	1.36	Alfa Aesar
Methanol	16	12.52	Fisher
Analytical reagent grade water	8	8.00	Fisher
Ammonia 2M	0.016	0.014	Sigma Aldrich
Electrical equipment:	**Running time (h)**	**Total kWh**	**Equipment supplier**
Fan cupboard	00:43	0.063	Astec

[Table 1: Data collection inventory for HTSCD gel preparation]

Raw materials used in gel preparation (LTSCD)	Volume (ml)	Mass (g)	Material Supplier
Tetramethoxysilane	16.2	16.56	Fisher
Methanol	16.2	12.96	Fisher
Analytical reagent grade water	8.1	8.10	Fisher
Ammonia 2M	0.072	0.062	Sigma Aldrich
Electrical equipment:	**Running time (h)**	**Total kWh**	**Equipment supplier**
Fan cupboard	01:20	0.112	Astec

[Table 2: Data collection inventory for LTSCD gel preparation]

3.2   Ageing

After gel preparation, 2 ml of methanol was added to each sample to prevent ambient drying and they were covered with Parafilm. Over the next 3 weeks, the gels were fully immersed in several solvent baths within the sealed plastic containers. During this step, all unreacted water diffused out from the gel, and the network had time to strengthen. In total, the HTSCD samples went through two solvent exchanges during the ageing process. The LTSCD samples went through five, where the fourth included a surface modification to make the gel hydrophobic. All saturated aging solvents were disposed into waste containers sent to the universities waste management facility. Table 3 and Table 4 display the respective data collection inventories.

| Raw materials used in ageing (HTSCD)	Volume (ml)	Mass (g)	Material Supplier
Methanol - *covering samples*	8	6.33	Fisher
Methanol - *1st exchange*	125	98.88	Fisher
Methanol - *2nd exchange*	125	98.88	Fisher
Electrical equipment:	**Running time (h)**	**Total kWh**	**Equipment supplier**
Fan cupboard	00:10	0.014	Astec

[Table 3: Data collection inventory for HTSCD ageing process]

Raw materials used in ageing (LTSCD)	Volume (ml)	Mass (g)	Material Supplier
Methanol - *covering samples*	36	28.48	Fisher
Methanol - *1st exchange*	250	197.75	Fisher
Methanol - *2nd exchange*	250	197.75	Fisher
Methanol - *3rd exchange*	200	158.20	Fisher
Hexamethyldisilazane - *4th ex*	40	30.96	Sigma Aldrich
Methanol - *4th exchange*	160	126.56	Fisher
Methanol - *5th exchange*	200	158.20	Fisher
Electrical equipment:	**Running time (h)**	**Total kWh**	**Equipment supplier**
Fan cupboard	00:35	0.049	Astec

[Table 4: Data collection inventory for LTSCD ageing process]

3.3 High Temperature Supercritical Drying

Figure 2 displays the equipment used and monitored during HTSCD. The process utilises an autoclave with a 1-litre capacity, connected to an electric heater and temperature sensor. A nitrogen bottle is connected prior to drying to create an inert atmosphere within the autoclave and check that the seals are capable of withstanding supercritical pressures. During supercritical drying, the temperature is controlled by manually entering set-points on the heater controller and the pressure is controlled using a needle valve. Excess solvent is drained away into a container as pressure is released.
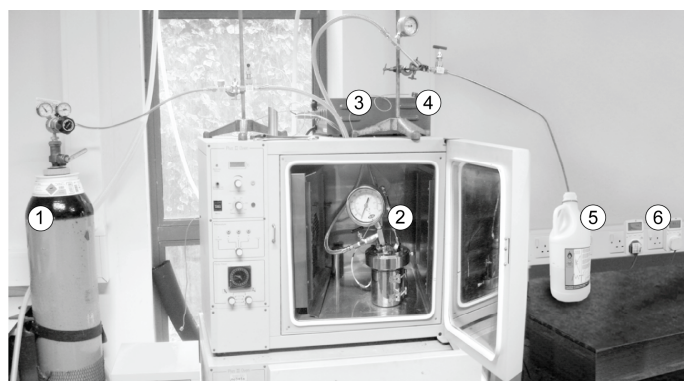
HTSCD: (1) Nitrogen canister   (3) Heater controller   (5) Solvent capture
        (2) Autoclave   (4) Temperature reader   (6) Plug monitors

[Figure 2: Equipment for HTSCD]

To begin, the 4 gel samples (still inside the test tubes) were placed inside the autoclave with 400 ml of methanol. Two steel bars were then inserted to displace some of the unused volume. The autoclave was then sealed and filled with regulated nitrogen to 100 bar to check the integrity of the chamber. After approximately 5 minutes, the nitrogen flow was disconnected and the pressure inside the autoclave was dropped to 10 bar.

The heater was programmed to a set point of 75 ^{o}C. Temperatures were raised 25 ^{o}C every 10 minutes until reaching 250 ^{o}C. Between 95-110 minutes, the set point was gradually increased to 280 ^{o}C. During this time, the pressure was allowed to rise to 100 bar, and then carefully controlled to stay at this level. At 120 minutes, solvent was manually drained out of the autoclave causing the pressure to fall. 15 minutes later, the heater set point was reduced to 100 ^{o}C and the unit was switched off. The autoclave was then left to cool for 100 minutes, after which the aerogel could be removed. 95% of waste solvent was recovered and disposed via the universities waste management facility.

Although the fluid in this process exceeded the critical point of methanol by ~40 °C and ~25 bar, we have noticed that if the pressure and temperature do not reach at least these values, the aerogel will be cracked and more shrunken. We attribute this to excess water causing a change in the critical point of the pore fluid, requiring higher temperature and pressure to reach supercritical conditions.

During the supercritical drying process, two plug-in electricity monitors were used to record the total kWh of the heater and temperature sensor. The total energy use of the process was 0.895kWh. Table 5 displays the data inventory for the entire HTSCD process. The mass of nitrogen used was calculated using the formula n=PV/RT. Here n= quantity of nitrogen consumed (moles), P = Pressure (bar), V = volume (litres), R = universal gas constant (0.0832), and T = temperature (Kelvin).

Raw materials used in drying (HTSCD)	Volume (ml)	Mass (g)	Material Supplier
Methanol	400	316.4	Fisher
Nitrogen (N_2 at 100 bar, 296K)	~500	0.057	BOC gases
Electrical equipment:	**Running time (h)**	**Total kWh**	**Equipment supplier**
Heater	02:15	0.882	SciMed
Temperature Sensor	04:00	0.013	RS

[Table 5: Data collection inventory for HTSCD]

3.4   Low Temperature Supercritical Drying

Figure 3 shows the equipment used and monitored during LTSCD. The process utilises a 1-litre capacity autoclave with a window for viewing supercritical extraction. The autoclave is connected to a liquid CO_2 canister, chiller, pipe heater, pump and vessel heater. A backpressure regulator controls the outflow of CO_2 and depressurisation rate of the autoclave. The entire process, including the flow rate of liquid CO_2, is controlled by a computer.

LTSCD:
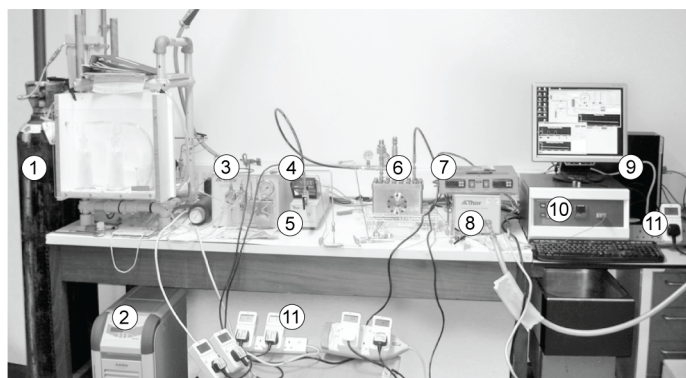
1. CO_2 canister
2. Chiller
3. Pump
4. Temperature sensor
5. Pipe heater
6. Autoclave
7. Pipe heater controller
8. Back pressure regulator
9. Computer & monitor
10. Autoclave heater controller
11. Plug monitors

[Figure 3: Equipment for LTSCD]

To begin, the 18 gel samples (removed from the cuvettes) were placed inside the autoclave filling approximately 5-10% of the usable space. 200ml of methanol was added to prevent the samples from cracking during the drying process. Next, the autoclave was sealed and liquid CO_2 flowed in until in equilibrium with the bottle pressure (~55 bar). Prior to entering the autoclave, the liquid CO_2 was chilled to 0 °C. A dual-piston pump increases the CO_2 pressure to 100 bar, flowing through a pipe heater at 45 °C into the autoclave.
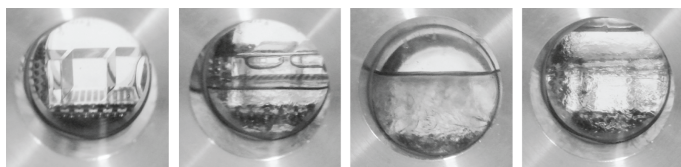
[Figure 4: Liquid CO_2 entering autoclave (photos taken at 0-30 minutes into drying)]

Figure 4 shows photographs taken through the window of the autoclave as liquid CO_2 enters and submerges the gel. Once supercritical conditions were reached, a vessel heater maintains the supercritical temperature at 45 ^{o}C, for approximately 4 hours. When depressurisation occurred, the chiller was switched off. As pressure dropped below 50 bar, the pipe heater and vessel heater were also switched off. Once cooled, the autoclave was opened and the aerogel could be removed.

Throughout LTSCD, no recycling of CO_2 occurred. Instead, all excess CO_2 was trailed through a pipe out of a nearby window. The total duration of CO_2 flow was 4 hours, 20 minutes. The total amount of CO_2 used, monitored by a computer during the drying process, was 4.538 kg, split across four main cycles. This value was verified by weighing the bottles using mechanical scales before and after supercritical drying.

During LTSCD, seven plug-in electricity monitor monitors were used to study the energy use of each piece of equipment. The total energy use was 3.063 kWh. The chiller (which cooled the CO_2 before entering the autoclave) accounted for over half of the total energy use, using 1.629 kWh. The computer with monitor had the second largest energy use accounting for 0.641 kWh. Table 6 displays the data collection inventory for the entire LTSCD process.

Raw materials used in drying (LTSCD)	Volume (ml)	Mass (g)	Material Supplier
Methanol	200	158.2	Fisher
Carbon dioxide	-	4538	BOC gases
Electrical equipment:	**Running time (h)**	**Total kWh**	**Equipment supplier**
Chiller	04:30	1.629	Thar
Computer & Monitor	07:00	0.641	Dell
Vessel Heater	05:45	0.206	Syrris & Lenton
Back Pressure Regulator	07:00	0.200	Thar
Pipe Heater	05:45	0.185	Thar & SciMed
Pump	07:00	0.180	Thar
Temperature sensor	07:00	0.022	RS

[Table 6: Data collection inventory for LTSCD]

4.0   The Aerogel

Figure 5 show photographs of the aerogel produced from HTSCD and LTSCD respectively. Samples have good optical quality and no internal cracks. They can be handled with care, but are fragile at the edges. Samples appear blue against a dark background and yellow against a light background. This is due to different wavelengths of light being transmitted, absorbed and reflected by the nanosized pores due to Rayleigh scattering [3].
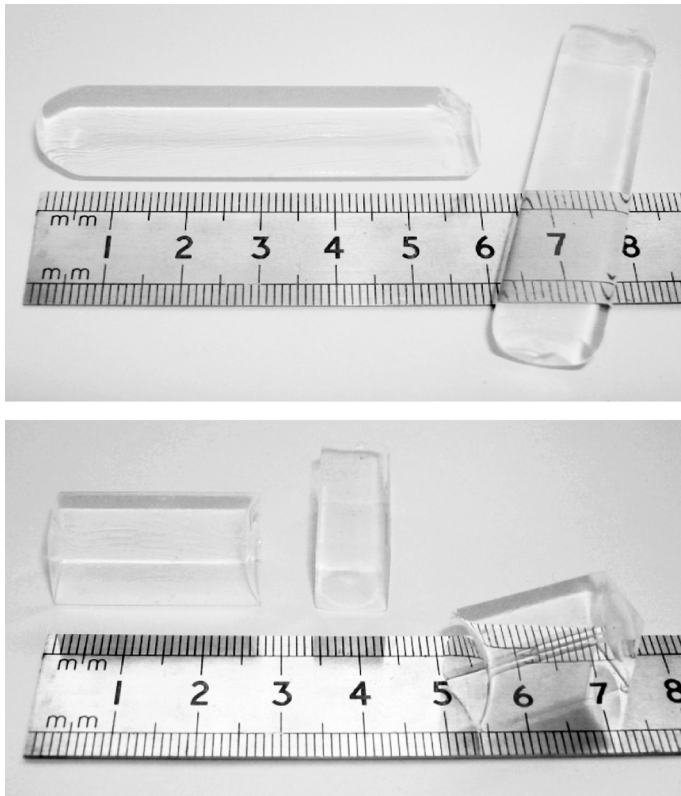
[Figure 5: Aerogel samples produced in the experiment. Top image shows the aerogel produced by HTSCD. Bottom image shows the aerogel produced by LTSCD.]

To assess the properties of the aerogel samples made in a lab compared to industrially produced aerogel, scanning electron microscopy (SEM) was used to investigate the general topography of the LTSCD and HTSCD samples alongside ambiently dried translucent granular aerogel produced industrially by Cabot Corporation. All samples were fractured prior to investigation. The industrial granules were fractured by crushing them against the viewing plate. The HTSCD and LTSCD aerogel samples were fractured by cutting them with a scalpel. All samples were brittle.

Figure 6 displays low magnification SEM images of the three aerogels showing their fracture conditions. As shown, the surface of all three samples appeared smooth and it was not possible to see individual pores or particles, indicating that these features are on a nanoscale. Micro-cracks and cleavage marks were clearly visible across the surface of each sample where fracturing occurred. This fracturing characteristic implied that the lab samples should have similar properties to industrially produced aerogel.

HTSCD aerogel   LTSCD aerogel   Aerogel granule produced by Cabot
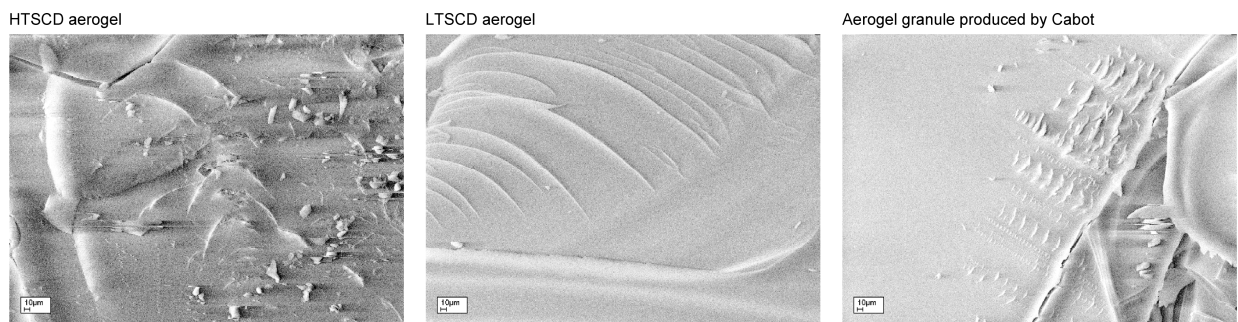
[Figure 6: Scanning electron microscopy showing the surface characteristics of three different aerogel samples at 500x magnification. The left image shows the HTSCD aerogel. The middle image shows the LTSCD aerogel. The right image shows ambiently dried translucent granular aerogel produced industrially by Cabot Corporation].

5.0 Inventory Analysis

Figure 7 displays the production energy and CO_2 burden associated with making 40 ml of aerogel via LTSCD and HTSCD. For each raw material and electrical usage, the production energy and CO_2 burden was calculated, based upon the following assumptions. The energy and CO_2 spent to produce methanol was used to represent the impact of all chemicals. Methanol accounted for ~96% of all chemicals used in both processes. An energy cost of 47 MJ/kg and CO_2 burden of 0.4 $kgCO_2$/kg was used for pure methanol manufacture [21,22]. Note that this reference for methanol also contains a combustion value of 30 MJ/kg, which was not included at this stage [22]. The impacts of nitrogen and water use were also disregarded. A carbon factor of 0.517 $kgCO_2$/kWh was assumed for grid electricity in the UK [23].
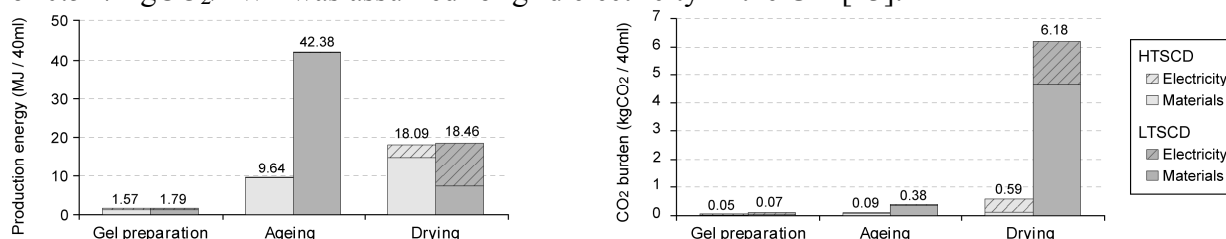
[Figure 7: Production impact associated with making 40ml aerogel via HTSCD and LTSCD. Left graph compares production energy. Right graph compares CO_2 burden]

The total production energy associated with HTSCD was 29.3 MJ/40ml. The total production energy associated with LTSCD was higher at 62.6 MJ/40ml. Regarding the total CO_2 burden, HTSCD was accountable for 0.73 $kgCO_2$/40 ml. LTSCD was higher at 6.64 $kgCO_2$/40 ml. The methanol used during ageing had the most significant impact on the total production energy. The CO_2 consumed during LTSCD had the most significant impact on total CO_2 burden.

6.0 Impact Assessment

To attain the functional unit, we have considered several ways both laboratory scale processes could be optimised to create 1 m^3 (1000 litres) of aerogel without major changes to equipment or manufacturing steps. These changes are:

Firstly, the maximum batch size during gel preparation could be expanded to 1-litre without different stirring mechanisms. Secondly, gel preparation time could be reduced to 20 minutes (as weighing ingredients during data collection prolonged the process). Thirdly, the amount of solvent used during ageing could be reduced, as the least amount of solvent required for aging is an identical volume to that of the gel. On this basis, 4 soaks are required for HTSCD and 7 soaks are required for LTSCD to completely remove the water before supercritical drying. This was calculated from tolerances of 0.16% water for HTSCD and 0.00128% water for LTSCD, the same water % used to successfully make gels in this study. Finally, we estimate that both 1-litre autoclaves could be filled with up to 500 ml of gel without changing the equipment, making drying 12.5 times more efficient.

Applying each change still means 1000 batches of gel must be prepared and aged, then supercritically dried over 2000 cycles, to produce 1 m^3 of aerogel. Nonetheless, the production energy and CO_2 burden arising from these scaled batches is shown in Figure 8.
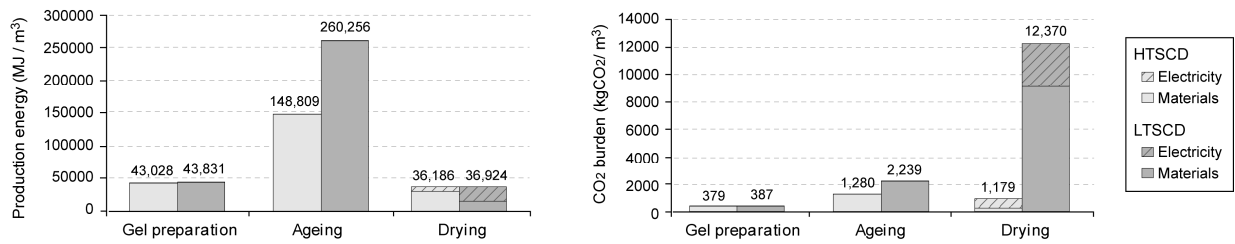
[Figure 8: Scaled production impact for making 1m^3 aerogel via HTSCD and LTSCD. Left graph compares production energy. Right graph compares CO_2 burden]

7.0   Interpretation

Prior to this investigation, in-situ testing found that retrofitting a 1 cm thick, 1 m^2 twin-wall polycarbonate panel filled with aerogel granules to single glazing could reduce heat loss by 80% [20]. If adapted into removable secondary glazing, for example, fitted permanently from 1st October – 31st May in a gas heated home in London, UK, then annual energy savings of approximately 400 kWh/m^2/year are predicted, equivalent to 1440 MJ/m^2/year and 80 kgCO$_2$/m^2/year (assuming a boiler efficiency of 84%, a gas carbon factor of 0.198 kgCO$_2$/kWh [23], and that the house is heated to 21 $^\circ$C all year round with an 18 $^\circ$C night-time set back). Taking a wall thickness of 0.5 mm, approximately 0.008 m^3/m^2 of granular aerogel was required to fill the twin-wall polycarbonate panel. Figure 9 displays the predicted energy and CO_2 production cost for these production runs. Values are compared against the material's estimated operational savings over a 15-year product lifespan.
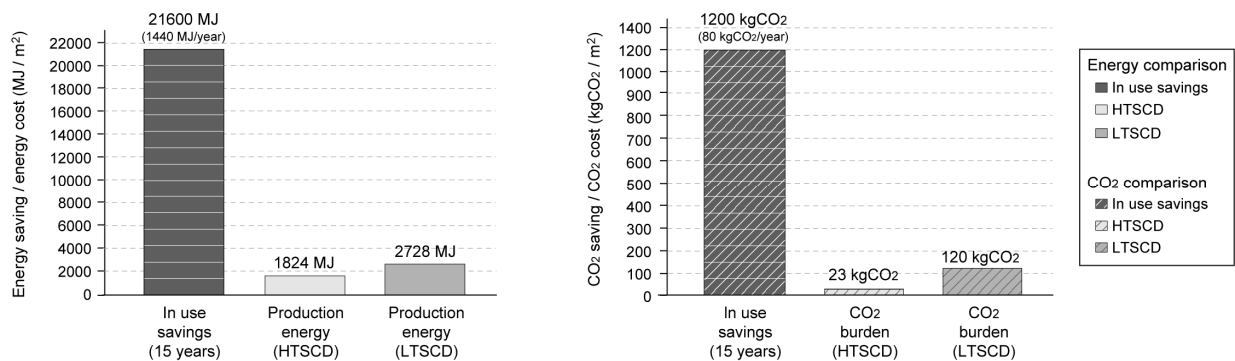
[Figure 9: Production costs of aerogel vs. in-use savings over product lifespan. Left graph compares production energy. Right graph compares CO_2 burden]

Results show that aerogel can provide a positive energy and CO_2 contribution within 0.3-1.9 years. Aerogel produced by HTSCD can recover its production energy within 1.3 years and its CO_2 burden within 0.3 years. Contrarily, aerogel produced by LTSCD can recover its production energy within 1.9 years and its CO_2 burden within 1.5 years.

Two factors omitted in this comparison were transport (of raw materials / finished products) and the impact of end of life processing (e.g. product re-use, recycling, landfill etc). Transport can be complicated to assess since it is unclear where a system boundary should be drawn in a global economy. A full sensitivity analysis should consider the type of vehicle, transport distance and loading etc. Contrarily, end of life processing can be complicated to assess since it is uncertain what might happen to products at the end of their usable lifespan. Presumably, aerogel would just be crushed and disposed of in the same way as sand or rocks, since the material consists of amorphous silica, which is not carcinogenic. Conversely, provided the aerogel has not been contaminated during its incorporation into a building, the thermal and optical properties are not expected to degrade and the material can be re-used again, resulting

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in further operational savings. According to the Cambridge Eco Selector (a comprehensive materials selection tool developed by Cambridge University), transporting 1 kg of insulation 100 km by ship and 300 km using a 32 ton truck accounts for just 0.15 MJ and landfill accounts for 0.2 MJ. As such, these factors are not expected to have a significant impact on the interpretation of results.

8.0   Limitations

A significant factor affecting the accuracy of this study is the differences between laboratory and industrial scale aerogel manufacture. Currently, scaling assumptions used to produce a 1 m^3 volume of aerogel, do not accurately represent the energy use and CO_2 burden that would result from producing this volume industrially. This issue is difficult to resolve, due to the lack of information from industry concerning the actual economies and efficiencies of scale associated with mass production of aerogel. As such, the interpretation of these results should be treated as conservative estimates, used to provide judgement as to whether silica aerogel is a good environmental technology or not. Primary sources of discrepancy are given below:

The laboratory process was scaled with no major changes to equipment or production steps
The maximum batch size for gel preparation and drying was restricted to 1 litre and 0.5 litres respectively. This meant 1000 batches of gel would have to be separately prepared and dried over 2000 cycles to produce 1 m^3 of aerogel. This is unrealistic in the context of commercial production. Larger batch sizes or continuous production would result in far greater efficiencies.

No recycling of CO_2 occurred
In the laboratory study, 4.5 kg of CO_2 was used to dry 40 ml of aerogel using LTSCD. This mass was directly scaled by 2000 times to produce 1 m^3 of aerogel. This scaling factor could be eliminated if CO_2 recycling had occurred. According to Aspen Aerogels, all CO_2 is recycled at their production facility.

No recycling or energy recovery from solvents occurred
The energy used to produce methanol was taken as 47 MJ/kg. The material has a combustion value of 30 MJ/kg, which was not included in the impact assessment. When producing aerogel on a mass scale, it can be assumed that solvents would be recycled/re-used or burnt for energy recovery. If recycled, then less methanol would need to be used. If the energy were recovered, this would result in the life cycle energy use in producing methanol being reduced to 17 MJ/kg.

The aerogels were solid, crack free and possessed high optical quality
Scaling assumptions predicted that 4-7 solvent exchanges were required to reproduce the high quality aerogel from the laboratory study. These exchanges aimed to purify the gels and completely remove the water to prevent cracking. Manufacturing granules with lower optical quality could mean that fewer solvent exchanges are required, and less control is needed to prevent cracking. Additionally, thinner granules require less time in the supercritical drying equipment as the time taken to remove the solvent scales with the square of the thickness.

Electrical equipment could be more efficient
The chiller used the largest amount of electricity during LTSCD. This unit was large and not been appropriately sized for its function. In industry, issues such as this would be corrected for cost savings, resulting in reductions in overall energy and CO_2 usage.

9.0   Industrial Economies of Scale

If we compare the results generated in this experiment to the corresponding benchmarks for Aspen Aerogel's Spaceloft® insulation, we find that our production energy and CO_2 burden, per m^3, is 28-42x and 4.4-23x larger, respectively, than industrial benchmarks. Note that the

lower value in each of these ranges represents the magnitude of difference for HTSCD aerogel and the higher value represents LTSCD aerogel. In addition, note that the production energy and CO_2 burden, per m^3, for Spaceloft® is 8139 MJ/m^3 and 648 $kgCO_2/m^3$ respectively, generated by multiplying the products impacts, per kg, by its nominal density of 151 kg/m^3 [24].

In an effort to understand (and bridge the gap) between laboratory scale and industrial scale manufacture, Figure 10 demonstrates how altering the scaling assumptions can significantly reduce this discrepancy.
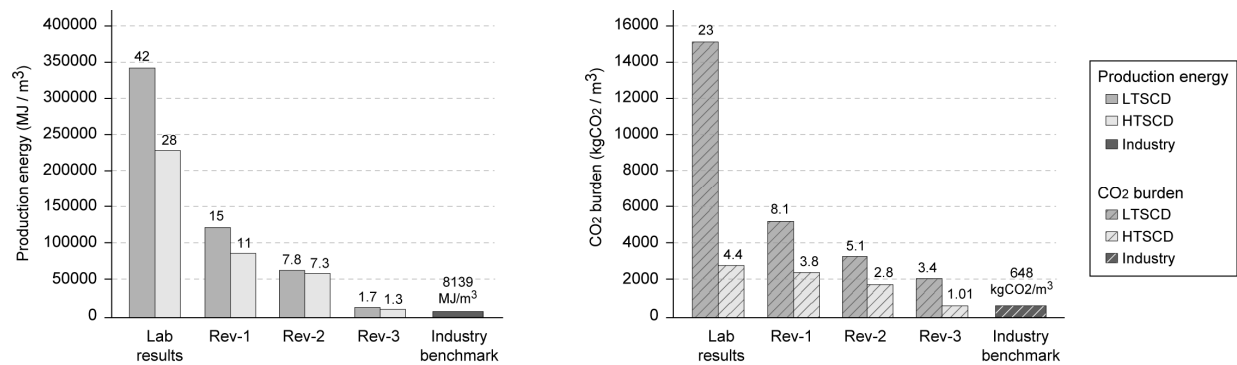
[Figure 10: Scaling revisions to bridge discrepancies between laboratory and industrial production. Left graph compares production energy. Right graph compares CO_2 burden. Bars are labelled showing the magnitude of difference compared to industry benchmarks]

Firstly, in revision 1, the batch size for gel preparation and drying is increased to 1000 litres enabling $1m^3$ of aerogel to be manufactured over one production run (as opposed to preparing 0.5 litres of gel 2000 times, followed by drying 1 litre of gel 1000 times). Additional changes include scaling up electricity use by 1000, assuming 100kg of CO_2 is used during drying for LTSCD and that all methanol usage in both manufacturing methods is combusted for energy recovery. The culmination of this revision causes the magnitude of difference to reduce to 11-15x and 3.8-8.1x respectively for production energy and CO_2 burden.

Going further, revision 2, assumes that two solvent exchanges for HTSCD and three for LTSCD are carried out and the chiller efficiency in LTSCD is increased by 80%. This causes the production energy and CO_2 burden discrepancy to be reduced further to 7.3-7.8x and 2.8-5.1x respectively.

Finally, revision 3, assumes that all CO_2 used during drying for LTSCD and all methanol used in both processes is recovered/recycled (thus eliminating the impact). This causes the difference to reduce to 1.3-1.7x for the embodied energy and 1.01-3.4x for the embodied CO_2.

10.0   Conclusion
The aim of this study was to investigate whether the use of aerogel as an insulation technology for the building sector provides a measurable environmental benefit over its life cycle. Two methods of aerogel production have been studied to compile a data inventory for this assessment. For each, the production energy and CO_2 burden was quantified, and scaled up to produce a 1 m^3 volume of aerogel. The impact was then compared against the operational savings over 15 years, arising from retrofitting translucent aerogel to single glazing.

Preliminary results indicate that aerogel produced by LTSCD and HTSCD could recover its production cost within 0.3-1.9 years. These results are well within the predicted lifespan of building products containing aerogel. The LTSCD method of aerogel manufacture had the

longest environmental payback. This was largely due to LTSCD having a higher amount of solvent use during the ageing process and because supercritical drying required more energy intensive equipment, whilst directly consumed CO_2.

The environmental impact of both manufacturing techniques could be reduced if larger batches were produced, more energy efficient equipment were used and/or if recycling or energy recovery of solvents took place. The greatest improvements are expected from LTSCD, since there is an opportunity to recycle the CO_2 used during drying. If the desire is to produce granular aerogel, there may also be opportunities to reduce the amount of solvents used, which account for a significant proportion of the total production energy. It must be emphasised that the true economies and efficiencies of scale associated with mass production are unclear due to a lack of information regarding commercial manufacturing of aerogel. Despite these factors, results have demonstrated that aerogel can provide a measurable benefit over its life cycle. Furthermore, an analysis of the scaling assumptions has shown that the discrepancies between laboratory and industrial scale manufacture can be significantly reduced.

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