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A FEASIBILITY ANALYSIS OF EVACUATED GLAZING WITH ADVANCED THERMAL PROPERTIES

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ZUSAMMENFASSUNG

After laying the technological foundation in 2007, the vacuum glazing project has entered the final prototyping and upscaling phase this year. Again all project goals could be met with exception of problems encountered on the prototyping side. The numerical analysis on heat transport and mechanical deflections was completed and the results published in scientific journals relevant to this field of research. Most importantly, the service life prediction model was completed and experimental model studies on cleaning efficacy of low-ε coated inner glazing surfaces carried out using state-of-the-art surface analytical tools. Those results suggest that the total pressure buildup over a typical 30 year service life can be kept at such low levels that the use of getter materials may become nonessential. In addition, a fast and economical edge sealing technology for vacuum glazing was developed based on anodic bonding of an activated tin-based soft solder alloy in the liquid state at a typical temperature of 300°C. This method is able to produce coherent, leak-tight seals over large glazing perimeters with relative ease. The adaptation of this technology to fabricate large glazing prototypes was demonstrated for a 50cm by 30cm sealing sample. Dispensing of the solder alloy in the form of prefabricated wires however has proven quite tricky. For this reason a follow up project is planned to separately investigate the solder dispensing problematic. To conclude, this project stands shortly before completion and all project milestones were complete success with the prototyping only counting as a partial success.

Projektziele

When compared with a conventional gas-filled glazing, the total heat transfer in an evacuated glazing rate is significantly reduced. Theoretically, an evacuated double glazing can achieve U-values of 0.2 - 0.5 $W \cdot m^{-2} \cdot K^{-1}$ [1]. For comparison, state-of-the-art double and triple glazings have total heat transfer coefficients of 1.0 and 0.7 $W \cdot m^{-2} \cdot K^{-1}$. This translates into energy savings of 40% to 80% of installed vacuum glazing area compared with commercial double glazing.

Goal and Milestones: The goal of this project is to develop the technology required for fabricating high-performance vacuum glazing which is superior to already existing concepts with a predicted service life of 30 years and by means of processes that are scalable to industrial dimensions and that can be realized cost-effectively. The following tasks (i) through (v) will need to be completed in order to give proof of principle of a working technology for commercial adaptation:

(i) development and demonstration of a technique for fabricating an edge seal with sufficient hermeticity and mechanical integrity *(completed)*

(ii) numerical analysis of heat transfer over the entire glazing assembly *(completed)*

(iii) numerical analysis of the mechanical behaviour of the glazing assembly *(completed)*

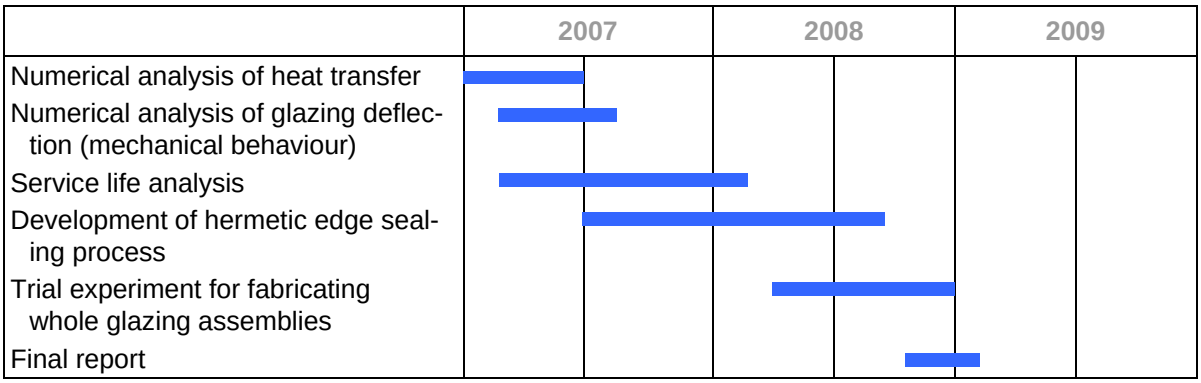
(iv) service life analysis based on leakage measurements of prototype assemblies and simulations *(completed)*

(v) fabrication of a roughly 0.5m x 0.5m prototype glazing assembly including support pillars and getter *(partially completed)*

Successful completion of the proposed research will lay the foundation for further upscaling and fabrication of larger prototype assemblies and construction of a pilot assembly line.

Durchgeführte Arbeiten und erreichte Ergebnisse

The following timeline for the work plan was established with the initial proposal for the program. The five key points will be addressed separately and current progress discussed in light of the defined project goals.



a), b) Numerical analysis of heat transfer and numerical analysis of glazing deflection (mechanical behavior)

These project milestones were completed in 2007, and the results published:

- H. Manz, On minimizing heat transport in architectural glazing, *Renewable Energy*, **2008**, 33(1), 119-128
- L. Wullschleger, H. Manz, K. Ghazi Wakili. Finite Element analysis of temperature-induced deflection of vacuum glazing, *Construction and Building materials*, Article online

c) Service life analysis

In the 2007 report, we described the three following sources of possible pressure buildup inside the cavity.

- **leakage of molecules through the edge seal**,
- **gas permeation through the glass panes**,
- **long-term outgassing and photofragmentation** behaviour: this involves release of volatile molecules by desorptive processes and fragmentation of larger hydrocarbon-based adsorbates to produce smaller more volatile fragments which populate the vacuum space.

In order to guarantee the superior thermal performance expected from a vacuum glazing unit, this pressure increase must not exceed 10^{-2} Pa (10^{-4}mbar) after a typical service life period of 30 years. Hence, a more detailed understanding of the leakage sources and mechanisms is crucial.

The leakage rate of the edge seal is given by the seal quality and integrity. The fabrication of such leaktight seals on an industrial scale has already been demonstrated and is in the realm of what modern process engineering is capable of. Glass soldering technology, first developed by the group of Collins [2], is used by manufacturers, mainly on the asian continent (China, Japan, Korea), to mass-produce vacuum glazing. Such units are however known for their limited thermal performance (typical U-values of ~1.2 – 1.5 $W \cdot m^{-2} \cdot K^{-1}$, two low-ε coatings) associated with the high processing temperatures.

For each new sealing technology the question of leaktightness is raised again and excessive testing is necessary before moving from laboratory to production scale. An additional uncertainty which requires special attention is the deterioration / aging of such a seal: due to alternating thermal and mechanical loading conditions induced by differential heating of interior and exterior panes, damage can occur. Such damages range from the formation of microcracks over partial delamination to complete failure or breakage. In order to better understand the probability of failure, cyclic loading tests need to be performed on sealing specimens and entire glazing prototypes. Thus far we have demonstrated the ability to manufacture leak tight samples which showed leak rates below the limit of detection of our leak testing device (< 10^{-10} $mbar \cdot l \cdot s^{-1}$). The fabrication of test specimens and the direct leak rate measurement is the easiest way to quantitatively assess the leak rate of a given edge sealing system.

Permeation of gases through the glazing surfaces can be estimated from gas permeation coefficients published in the literature for various glasses [3,4]. For each relevant constituent present in ambient air, the permeation coefficient is corrected by an Arrhenius correction to any given temperature and the corresponding pressure increase calculated from the volume of gas transported under standard conditions over a given period. The sum of all components yields the total estimated pressure increase. Due to its small size and huge permeation rate, helium accounts for the majority of the total pressure increase (> 90%). The permeation of water vapor through glass was also estimated using the method of Todd [5] and found to be negligibly small at close to ambient temperatures. Figure 1 shows the temporal pressure increase due to gas permeation through two 6mm thick glass sheets dimension with a 250μm vacuum gap at a constant temperature of 20°C.

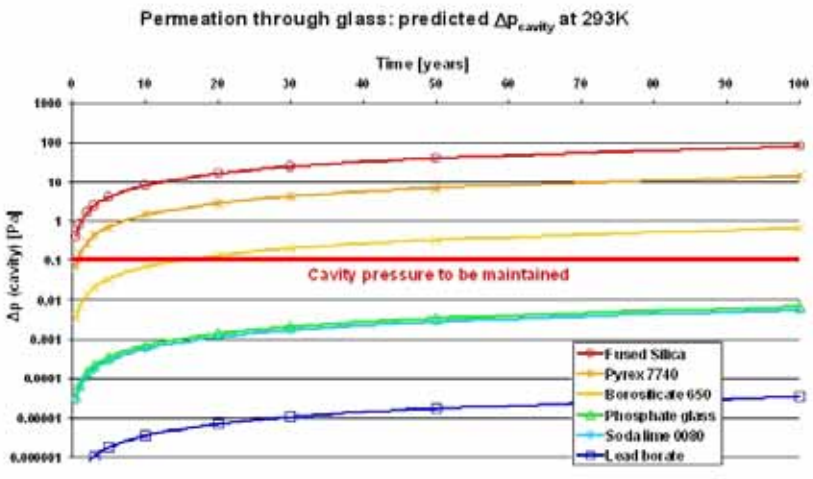


Fig. 1: Predicted time-dependent cavity pressure increase for various glass types due to gas permeation through the panes

Note that the pressure increase is indirectly proportional to the vacuum gap distance but independent of the glazing dimension, since both, gap volume and total amount of gas permeated through the glass sheets, increase linearly with the glazing area.

Depending on the type of glass used, the calculated pressure increase varies by approximately six orders of magnitude !! It seems like a providential act of nature that soda lime glass (which is rather identical in its chemical composition with soda lime glass used in the float glass production process) combines low cost and the second lowest gas permeability of the six glass types shown here. This is because of the large amount of network modifiers (Na, K, Ca, Mg) present in such a glass. Quartz, Pyrex and borosilicate glasses are inept for use in vacuum glazing.

Quantitatively we conclude that gas permeation through the glass panes is in no way limiting the performance of evacuated glazing if 6mm float glass is used for its manufacture with an estimated perssure increase below 0.01 Pa after 100 years. Even under worst-case scenario conditions, i.e. assuming one pane at a constant temperature of 80°C and the second at 20°C, the estimated pressure increase after 30 years would be below 0.1 Pa. Whether a real vacuum glazing unit could actually withstand thermomechanical stresses associated with this worst-case temperature difference is an entirely different question.

Long-term outgassing and photofragmentation in vacuum glazing is a subject which is still poorly understood. Outgassing of evacuated glazing assemblies fabricated by the glass solder method has been extensively discussed in the literature [6,7]. This work is closely related to outgassing effects known from ultrahigh vacuum (UHV) science and technology and can be summarized as follows:

- Upon heating or UV exposure, adsorbed small molecules such as H_2O, CO und CO_2 will outgas over time, if the glazing surfaces were baked prior to, during and after evacuation through a pumpout tube at a temperature of 150°C.
- No outgasing of small molecules will occur over time if the glazing surfaces were baked at temperatures above 300°C during the evacuation process.
- Glazing surfaces are contaminated with organic compounds and small molecules instantaneously if they are exposed to ambient air (shown by XPS).

Therefore, long-term outgassing is effectively suppressed, if the glass panes are heated to 300°C during sealing and evacuation. Our edge sealing approach is based on a sealing process in a high vacuum (HV) environment at a temperature of ~300°C which eliminates the problematic of future outgassing or photodesorption of small molecules.

A far more complex problem which has not been discussed in the literature is the photoinduced fragmentation of large organic molecules such as tensides, long-chain hydrocarbons, silicones etc. Such species are omnipresent in the ambient air and adsorb irreversibly on glass surfaces. Even if only a small coverage of such adsorbates is found on glass surfaces, decomposition of such species could lead to significant pressure increases (> 1Pa) over time. We therefore propose that the bull be taken by the horns or in other words that large organic molecules adsorbed on the glass surfaces be removed by means of a suitable surface cleaning method. The German Pro-VIG vacuum glazing project [8] employs a similar cleaning strategy prior to introducing the glass panes into the HV chamber.

In order to test the usefulness of various surface cleaning methods, we carried out a model study with the goal to demonstrate the cleaning efficacy. For this purpose we covered the low-ε coated side of a commercial window glass (Silverstar NT, Glas Trösch AG) with a well defined amount of a tenside molecule, sodium dodecyl sulfate (SDS). The surface coverage of the model contaminant SDS was analyzed by means of time-of-flight secondary ion mass spectrometry (TOF-SIMS). We found that a 5 minute UV/ozone cleaning treatment completely removed the model contaminant adsorbate. The relative SDS signal intensity dropped by over 4 orders of magnitude and was at the level of the instrument noise after the UV/ozone cleaning treatment. With this study completed, we are confident, that such a cleaning step significantly reduces the risk of pressure buildup due to photofragmentation reactions due to large organic adsorbate molecules.

To conclude, we believe that if all three sources of pressure buildup are kept in check (i.e. at a level below 0.1Pa after 30years), the use of chemical getters may become dispensable.

d) Development of a hermetic edge sealing process

In the 2007 annual report we pointed out the importance of the three following criteria necessary for an economically and thermally feasible sealing technology for vacuum glazing:

- low process temperature (to prevent damage to low-ε coatings and loss of tempering)
- fast method (for economical large-scale production)
- sealing in vacuo ("seal and forget" process eliminates evacuation through pumpout tube)

We present here the basic principle and selected experimental results of this sealing method which was filed for patenting in 2008. The underlying concept is that of anodic bonding, a joining method which can produce mechanically strong connections between an insulator and a metal without the need for sample pretreatment or activation [9].

Motivated by the potential usefulness of this technique for the problem at hand, we began by fabricating glass/metal/glass sandwich composite sealing specimens. Our first experiments were based on the soda-lime glass / Al system which is widely described in the literature [10]. A first series of experiments were carried out under standard atmospheric pressure without inert gas protection. The effect of the bonding parameters, such as bonding time and applied voltage was crudely monitored. With relatively little effort we were able to obtain the first 3cm by 3cm leaktight edge seal prototypes by bonding a coherent gasket made from 30μm thick Al foil.

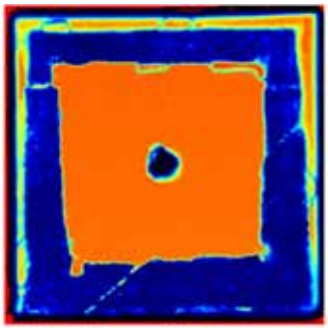


Fig. 2: Ultrasonic image of 250μm Al sheet bonded 5cm by 5cm specimen showing contact defects (hairline cracks)

Shortly thereafter we discovered the limitations of the Al gasket method namely that thin foils can be bonded uniformly whereas for thicker Al sheets which would be to define a typical vacuum gap of approximately 250μm did not produce leaktight seals, even for small 5cm by 5cm bonding specimens. The explanation for this behavior is that the material stiffness increases in a nonlinear fashion with increasing foil / sheet thickness and therefore renders a uniform surface contact of the gasket on both glass panes around the entire seal perimeter virtually impossible.

The next solution approach was to use tin-based solder alloys as the seal material with the advantage that in the molten state they would perfectly adapt to both glass surfaces thus eliminating the uniform contact difficulties. Unfortunately, we were unable to produce a mechanically stable anodic bond with conventional tin based solders. In order to reduce the amount of surface oxidation of the tin solder we built a HV bonding setup which allowed for anodic bonding experiments to be performed at typical pressures of $7 \cdot 10^{-4}$ mbar. Even though experiments with commercial tin solders *in vacuo* showed reduced surface oxidation, we were unable to produce a stable anodic bond with glass.

It was only by coalloying an activating component to the tin solder that we succeeded in producing mechanically strong bonding specimens. By extruding a wire from liquid solder and overlapping it to form a gasket we were then able to produce our first leak tight anodically bonded samples based on tin solder. Figure 3 below shows a sample which was fabricated by liquid phase anodic bonding of an activated tin-solder wire. Despite obvious defects in the form of bubbles within the solder seal, the sample proved leaktight within the limit of detection of the leak testing device ($<10^{-10}$ mbar $\cdot$ l $\cdot$ s^{-1}).

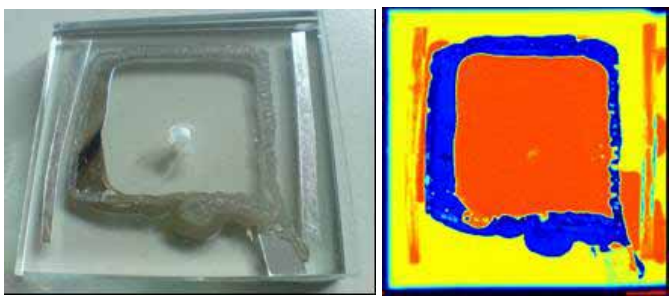


Fig. 3: Photographic (left) and ultrasonic (right) images of a leak tight 5cm by 5cm bonding specimen obtained via the liquid phase activated tin solder anodic bonding method

e) Trial experiments for fabricating whole glazing assemblies

In 2007 we began with the planning of a HV chamber designed to fabricate 0.5m by 0.5m evacuated glazing prototypes. This chamber was constructed and tested in the spring of 2008. A figure of the chamber and its interior is shown below in Figure 4.

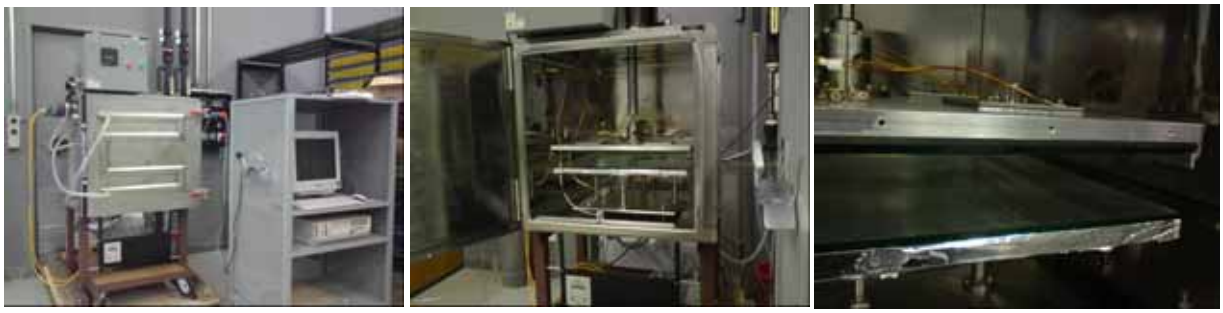


Fig. 4: Images showing the entire prototyping system including controls (left), the interior of the HV chamber (center) and the heated Al electrodes with mounted glass panes (right).

Briefly the chamber consists of two heated electrodes (1.6kW heating power per electrode) and various thermocouples for temperature measurement. The upper electrode is hanging from the top and can be raised and lowered by a linear feedthrough. For a typical experiment, two glass panes are cleaned and positioned with a solder wire frame placed on the bottom pane and contacted. The initial separation between the two plates is set to 15 cm. Once the chamber is sealed and pumped to a base pressure of $2 \cdot 10^{-4}$ mbar, the temperature is slowly raised (~ $1.5°C \cdot min^{-1}$) to the desired sealing temperature, typically 270-320°C. Then the upper plate is lowered onto the lower one by means of the linear stage and the anodic bonding process initiated by applying a voltage between the Al electrodes and the liquid solder seal. Once the bonding is complete, the bonding voltage is switched off together with the heating for the electrode plates. The sample is then allowed to cool to room temperature with the vacuum pumps still on, the chamber vented and the prototype sample removed from the system.

The major problem related to the prototype manufacture was the lack of activated tin-solder wire. The raw solder material was produced in the Empa Joining and Interface Technologies Department (124) in the form of ingots. Those were then remelted in a home built solder extruder specifically designed and built for making solder wire from this material (Fig. 5). Initial attempts to manufacture solder wire and various optimizations led to somewhat improved solder wire output. Howvere, the wires obtained in this way were usually short pieces (10 - 20cm) which had to be combined with a solder iron and contained significant amounts of surface impurities such as oxides. Efforts to reduce the amount of oxide formation such as transfer into a inert gas purged glove box yielded little to no improvement.

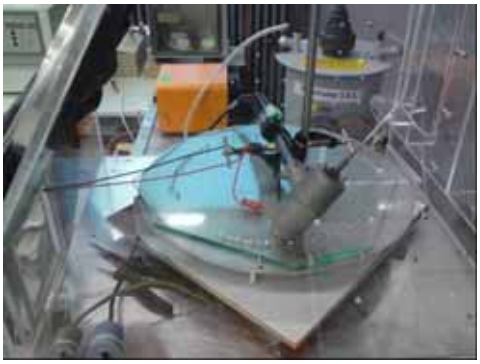


Fig. 5: Manufacture of solder wire used for prototype edge sealing from activated solder ingots by means of extrusion of liquid metal in an inert gas atmosphere

Fig. 6: First 50 cm by 30 cm sealing prototype manufactured in the large high vacuum chamber.

Figure 6 shows our first VG sealing prototype made using the technology developed by us. The seal is coherent along the entire perimeter but shows several large defects and is certainly anything but hermetic. Unfortunately we were unable to produce additional samples due to the lack of a reproducible technology to produce our own solder wire in the quantities needed for such an endeavor. Future research efforts will be centered around improving the solder dispensing technology.

Nationale Zusammenarbeit

Empa-internal collaboration with Dr. N. Bosco (Dept. 124) as part of this project in 2008 was centered around the edge-sealing problem, setup and testing of the prototype assembly vacuum system as well as conducting preliminary mechanical characterization on edge seal test specimens. Dr. Bosco left Empa in June of 2008 to return to the USA. A collaboration with Glas Trösch AG is planned for the follow-up project and is planned for 2009.

Internationale Zusammenarbeit

Currently there are no international partners linked to this project.

Bewertung 2007 und Ausblick 2008

The vacuum glazing project has made good progress again in 2008, however our progress in the field of prototype manufacture was less than expected due to shortage of personnel and unexpected complications with the solder dispensing and wire manufacture. A substantial amount of effort was spent on creating trying to manufacture long, coherent pieces of activated solder wire, however without beeing able to solve the problem once and for all. The large vacuum chamber was set up and tested and performs as expected. Our achievements in the field of surface cleaning model studies and service life prediction are such that we will shortly publish a scientific article on the various factors affecting service life in evacuated glazing.

To conclude we have met all of the goals set for this year except for the prototype manufacture. We only produced a single 30cm by 50 cm sealing prototype, and the lack of sufficient quantities of (oxide free) solder wire prevented us from fabricating additional prototypes. However we have been granted additional EKZ project funding to investigate the solder dispensing problematic as a separate main task and we are looking forward to this work which will start in 2009. With our combined efforts addressing the solder dispensing problematic, there seems a reasonable chance of success of the Empa vacuum glazing project for the near future. Provided that we can find a viable solution for the solder dispensing problematic, a KTI project for upscaling to a continuous production pilot plant with a leading Swiss glazing fabricator Glas Trösch AG is in planning for 2010- 2011.

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