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DEVELOPMENT OF VACUUM GLAZING WITH ADVANCED THERMAL PROPERTIES

Annual report 2007

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SUMMARY

The vacuum glazing project at Empa has met all of its goals with considerable success this year. Most importantly, a promising approach was found for the edge-sealing which poses the main obstacle for a practical realization. Using the process we were able to seal small glass samples within 90 seconds at 300˚C producing leak-tight and mechanically strong joints. This process is fast, simple and most importantly completely vacuum-compatible and will set a new standard in the field of large area glass/glass sealing. This technology is currently being patented and has a great market potential in the vacuum glazing field. The fabrication of 0.5m · 0.5m prototype glazing units with this sealing technology is planned for spring 2008.

A second focus is centered around model calculations on heat transfer and mechanical properties. Those calculations were completed and the results published. It was shown that a double vacuum glazing with two low-ε coated glass panes and a cavity pressure on the order of 10^{-2} Pa will have a center of glazing U-value of 0.2 - 0.5 $W \cdot m^{-2} \cdot K^{-1}$. According to our calculations, a similarly constructed triple vacuum glazing would achieve center of glazing U-values below 0.2 $W \cdot m^{-2} \cdot K^{-1}$ providing the thermal insulation performance of 20cm of Rockwool in a mere 2cm of total thickness and with the added benefit of substantial solar gains because of its optical transparence. For small glazings (0.5m · 0.5m), significant heat loss by conduction through the edge seal were predicted, however in the case of larger windows (> 2m · 2m), these edge-effects become negligible. A finite element mechanical study suggested large deflections of highly insulating glazing when joined mechanically at the edge. For a 3m · 3m thick double glazing (glass thickness = 6mm), the model predicted a central deflection of ~3cm when subjected to a temperature difference of 50˚C.

Finally, a model for the total pressure balance in an evacuated cavity was established taking into account three sources to the total pressure buildup namely leakage through the edge seal, the glass surfaces and desorption/photofragmentation of small, mostly carbon-based molecules. The first two contributions are mostly due to diffusion of He-molecules and can be kept at levels on the order of 10^{-2}Pa after 30 years by choosing proper materials and parameters. Surface cleaning of the glass is necessary to minimize the pressure increase caused by photofragmentation of hydrocarbons under UV irradiation and to minimize the amount of chemical getter material which is necessary to trap small organic molecules produced by such photofragmentation processes. A separate study on the role of the carbon surface chemistry was initiated to elucidate the underlying mechanisms.

Project goals

In Switzerland more than 40% of all energy is consumed in buildings, mainly for heating purposes. Windows constitute weak links in the building envelope that exercise a major impact on heating energy demand. The heat transfer mechanisms in gas-filled glazing cavities include radiative exchange between the glass sheet surfaces, convection and gaseous conduction. The application of two low-emissivity coatings ($\varepsilon = 0.04$) lowers the thermal conductance due to radiation between the glass pane surfaces to roughly 0.1 $Wm^{-2}K^{-1}$. Even when fill gases such as argon, krypton and xenon are used, thermal conductance due to convection and conduction cannot be reduced significantly below 1 $Wm^{-2}K^{-1}$. However, if the cavity is evacuated to approximately 10^{-4} mbar [1],[2], heat transfer by convection and gaseous conduction becomes negligible. In an evacuated glazing assembly, the total heat transfer rate is determined by radiation and, even more importantly, conduction through support pillars required to bear the atmospheric load on the external glass sheet surfaces. By evacuating the cavity, centre-of-glazing heat transfer rates are achievable that are two to five times lower than those of gas-filled cavities. This translates into energy savings of similar magnitude of installed vacuum glazing compared to state-of the art double glazing for large glazing surfaces.

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 > 20 years and by means of processes that are scalable to industrial dimensions and that can be realized cost-effectively. En route to realizing this goal, the following milestones will be achieved:

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

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

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

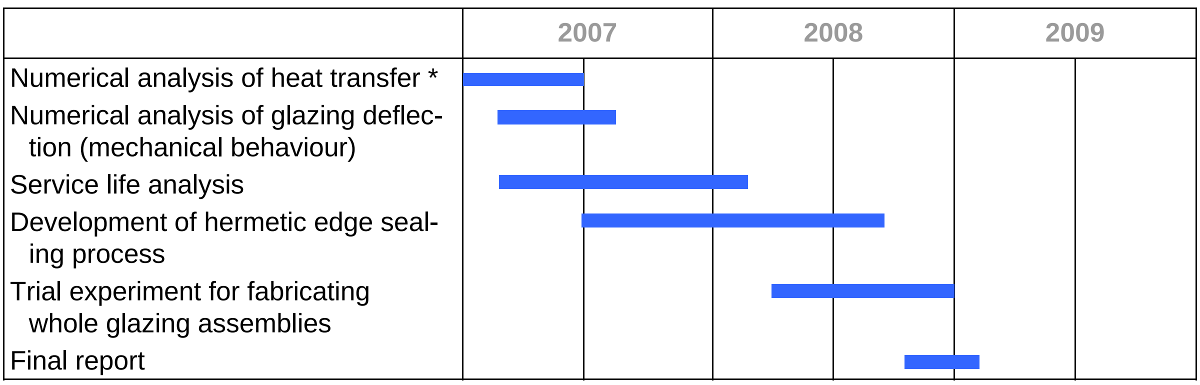
(iv) service life analysis based on leakage measurements of prototype assemblies and simulations

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

FINAL GOAL: Successful completion of the proposed program will result in demonstration of a new technology superior to existing concepts. The envisioned glazing will possess a center-of-glazing thermal transmittance of 0.5 $W/(m^2 \cdot K)$ or less, a value which is significantly lower than currently available evacuated glazings. Our prototype shall demonstrate the ability to maintain that reduced thermal transmittance over extended periods of time. At the same time our prototype will be more aesthetic than other competitors products since it does not require the unsightly evacuation port.

Research efforts and accomplishments

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



a) Numerical analysis of heat transfer

In the initial stage of the vacuum glazing project which was initiated by Dr. H. Manz, a numerical parameter study was carried out with the goal of estimating the total thermal transmittance as a function of various parameters. According to our models, heat transport through convection and conduction by gas molecules becomes negligible (Λ_{Gas}< 0.05 $W \cdot m^{-2} \cdot K^{-1}$) at pressures below 10^{-2}Pa (10^{-4}mbar) [2]. If both glass panes are decorated with low-IR emissivity coatings, heat loss by radiative energy transfer Λ_{Rad} is of a similar magnitude (~ 0.05 $W \cdot m^{-2} \cdot K^{-1}$). At this point, heat conduction through the support pillars (which take up the atmospheric pressure acting on the glass) and the edge seal becomes the predominant mechanism of heat transport. Depending on the dimensions, material and separation of the pillars, we predict center of glazing U-values of 0.2 – 0.5 $W \cdot m^{-2} \cdot K^{-1}$.

The effective U-Value (U_{eff}) in a glazing is always higher than its center of glazing value because of edge effects. For small glazings (0.5m · 0.5m) heat conduction through the edge seal strongly influences the total glazing performance, whereas for large ones (> 2m · 2m), U_{eff} is close to the center of glazing value. Hence, vacuum glazing units should be of a certain minimum dimension to meet their energy efficient reputation.

Additional calculations on triply-glazed vacuum glass [3] predicted super insulating properties of such constructs with center of glazing U-values of > 0.2 $W \cdot m^{-2} \cdot K^{-1}$. If available at competitive prices, such systems could become an attractive alternative to conventional external building insulation with the added advantage significant solar gains on the façade during the cold season. For reference, a typical exterior building insulation of 20cm of Rockwool has a U-value of about 0.165 $W \cdot m^{-2} \cdot K^{-1}$.

Our results are consistent with similar studies published by other research groups within the vacuum glazing community. Publication of our results concludes our efforts on the heat transfer modeling and meets the final project goals defined in the project proposal.

b) Numerical analysis of glazing deflection (mechanical behavior)

Mechanical stresses can pose a significant problem for vacuum glazing possibly leading to weakening of the edge seal or affecting the integrity of the glass [4]. A finite element method was used to predict deformations produced by uneven thermal expansion of a hot and a cold window pane of a vacuum glazing consisting of two glass sheets, an edge seal and support pillars. Fixed-frame boundary conditions were assumed. For small glazing dimensions (0.5m · 0.5m), the predicted center of glazing surface deflection increases in a roughly linear fashion with increasing temperature difference between the hot and the cold side Θ. Typical surface deflections for such small units and a temperature difference Θ of 50K are on the order of 2mm. For large (3m · 3m) units however, the dependence of the deflection amplitude on Θ is no longer linear but follows a power-law. The predicted center of glazing deflection amplitude for a 3m · 3m vacuum glazing and 6mm thickness of the panes is almost 30mm. This confirms the increasing problematic of thermomechanical stress of rigid-edge-sealed superinsulating glazings with increasing dimension. These results were submitted for publication and the manuscript is currently under reviewing. This concludes the thermomechanical modeling part of this project.

c) Service life analysis

A key issue associated with the practical realization of evacuated glazing is to maintain a cavity pressure of approximately 10^{-2} Pa over a typical service life of three decades. A model for analyzing service life will therefore was developed which takes into account the three main contributors to pressure buildup inside the cavity:

- leakage of molecules through the edge seal,
- gas permeation through the glass panes,
- long-term outgassing behaviour [5]: 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.

The sum of all three sources of pressure buildup given above must be known within a reasonable margin of error in order to choose the appropriate amount of getter material (the sink) necessary to chemically trap the desorbed molecules.

The contribution of the leakage through the edge seal is a property of that said seal and can be measured for a given test specimen by means of a leak detector. Given a 2m · 3m vacuum glazing with an evacuated cavity spacing of 250μm (volume = 1.5l), a leak rate of 10^{-12} mbar·l·s^{-1} (10^{-13} Pa·m^3·s^{-1}) will result in a pressure increase Δp of 6.3 ·10^{-2} Pa over 30 years. According to the VIG project (see literature) report, the benchmark for the edge seal is < 10^{-12} mbar·l·s^{-1}. This value is in agreement with our prediction, however it is worth mentioning, that the tightness requirements depend on the cavity volume and hence change somewhat with the window geometry.

The permeation of gases through different types of glass was calculated based on a thermally activated diffusion model. Diffusion rates for all relevant gaseous components in ambient air were computed based on the relative abundance of each species. An Arrhenius model was used to determine the effect of temperature. Because of its small size and huge diffusion rate constant compared to other gases, Helium accounts for most of the pressure increase (> 90%) due to gas permeation. The model also predicts vast differences in the permeabilities of different glasses ranging over more than six orders of magnitude. For a glazing consisting of two 6mm soda lime glass 0800 sheets (reasonably close in its chemical composition to float glass), a pressure increase of 1.9 ·10^{-3} Pa after 30 years was predicted for a 250μm vacuum gap if kept at 25˚C. Note that the pressure increase from diffusion through the glass panes is independent of the glazing area. If the same glazing were kept at 60˚C for 30 years, a pressure increase of 1.5 ·10^{-2} Pa would ensue, almost ten times the value at 25˚C. These results clearly show, that at elevated temperatures, the diffusion of gases (mostly He) through the glass panes can become service-life limiting. It is important to point out that the calculations were made with both panes at the same temperature level. A more practical calculation would take into account a built-in configuration with one pane exposed to ambient and another to hot or cold external conditions. Also, a more accurate prediction could be made when taking into account temporal variations (night/day and seasonal) of the outside temperature. Finally, commercial vacuum glazing units will be decorated with low-ε coatings which could cause a lowering of the effective diffusion rates compared to uncoated glass.

The last source of pressure increase due to desorption of water and various hydrocarbon species. To date this still remains a largely unknown factor and requires special attention [6]. Assuming that water can be removed entirely by a combination of initial surface treatment and gettering, the main lifetime limitation would likely be imposed by photofragmentation of carbon-based adsorbates (mostly aliphatic fatty acids, alcohols, amines etc.). Such molecules are omnipresent on each surface with prior exposure to ambient air and contamination cannot be avoided. Like in the case of water, removal prior to assembly is the first line of defense. Cleaning of such carbon based contaminants is a ubiquitous problem in the semiconductor industry [7] and many processes are known to complete this task. UV/ozone cleaning may be a mild method to remove carbon contamination and reduce the potential for future photofragmentation by UV light. If we assume surface where carbon-based adsorbates have been removed using state-of-the-art techniques, the residual amount of carbon is on the order of 5% of a monolayer coverage. If this carbon were to photodecompose COMPLETELY into ethylene, we estimate a maximum pressure increase of 15Pa from this gas evolution (for the same window geometry as given above). Without any surface cleaning, complete conversion of hydrocarbons to ethylene could raise the pressure inside the gap by up to 6mbar or 600Pa !!! Even though this sounds alarming, there is no immediate cause for concern: Little is known about the specific photochemical mechanism of such fragmentation processes and about their respective kinetic rates. Complete fragmentation into small molecules could is not likely over a 30 year life cycle. Hence this worst case scenario-estimation is in no way close to reality. We have initiated a separate research project to further elucidate cleaning and photo-fragmentation aspects of surface contamination by hydrocarbons.

Having discussed the three sources of pressure buildup inside the cavity over a life cycle, it must be pointed out that the use of getter (a chemical "trap" which capture gaseous molecule which are about to populate the vacuum space) materials will allow a significant prolongation of the service life. Chemical getters are metallic alloys which have a high surface affinity to many small molecules (H_20, CO, CO_2, ethylene, H_2 etc.) and are able to capture them. Chemically inert molecules such as He or Ne however cannot be trapped by chemical getters. Hence, the leak rates through the glass panes and the edge seal which will be dominated by He-permeation cannot be compensated for by such getters. Hydrocarbons on the other hand can be trapped by appropriate getter materials. Such materials are relatively expensive and their amount should be kept as low as possible inside the glazing. This again emphasizes the importance of eliminating the source of small molecules to a largest possible extent by properly cleaning the surface prior to the glazing assembly.

Our modeling calculations are in their final stage, results will be published soon in appropriate scientific journals. After completion of the carbon-surface chemistry subproject, we expect to make more quantitative predictions about amounts of surface carbon after various industrially scalable cleaning treatments [8] and about photodecomposition rates at known UV-irradiance levels. This will allow quantitative predictions of the cavity-pressure time dependence in any vacuum glazing unit based on a set of know input parameters.

d) Development of a hermetic edge sealing process

Probably the most important aspect of this work is the development of a proper edge sealing technology. Despite the fact that there are many research groups worldwide working on a similar tasks, the edge seal is the main obstacle for a practical realization of vacuum glazing. Probably the most widely used approach nowadays is sealing of two glass panes with a glass solder. There are many disadvantages associated with this concept which we want to improve on with our innovative sealing method. Glass solder sealing requires high temperatures (450˚C - 500˚C) applied over periods of several hours. Such high temperatures are not compatible with low-ε coatings and tempered glass, which from the start disqualifies such a window from ever reaching U-values below 1 $W \cdot m^{-2} \cdot K^{-1}$(radiative heat transfer becomes performance limiting). In addition it is a non-vacuum proof technology which means that the seal is made first and the glazing is evacuated later through a pumpout tube which adds an extra step in the production process and is not practical for large glazings and the required cavity pressures.

We would like to point out the importance of the following three key requirements for an ideal edge sealing method, namely

- low process temperature [9](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)

After much trial and error and many hours spent in the laboratory, we have found a very promising sealing method which meets all of the above criteria. Recently, we have demonstrated the fabrication of a 5cm · 5cm test frame which was sealed within 90sec below 300˚C in a vacuum chamber at a base pressure of $6 \cdot 10^{-2}$ Pa. The seal formed in this way was shown to be leaktight (leak rate below detection limit of the instrument < 10^{-10} $mbar \cdot l \cdot s^{-1}$) and mechanically very strong. Patenting of this technique is pending which is why we renounce to provide further information about the method at this point. We are very excited about this technique and we believe it is as close to an ideal solution to the edge-sealing problem as there can be. This completes the quest for an ideal sealing method and we are now focusing on the upscaling of this technique to fabricate larger prototype glazings.

e) Trial experiments for fabricating whole glazing assemblies

This part of the project will start in March 2008 and is currently in planning.

National collaboration

Empa-internal collaboration with Dr. N. Bosco (Dept. 124) as part of this project in 2007 was mostly centered around the edge-sealing problem and planning of a prototype assembly vacuum system. Patenting of this technology is pending and for proprietary reasons, close collaboration with industry has not been possible until now. Collaboration with ETH Zürich is envisioned in the near future with the opening of a joint Empa/ETH PhD position in 2008. A CTI project with a major glass manufacturer is planned for 2009.

International collaboration

Currently there are no international partners linked to this project. However we are in contact with vacuum glazing project leaders at Grenzebach GmbH, ZAE Bayern and Fraunhofer Institutes [10],[11]. Future collaborations are in planning.

Assessment 2007 and outlook on 2008

2007 has been a very successful year judging by the advances in this project. Of central importance are our achievements in the field of the edge-sealing which will be transferred into a proprietary technology. A patent related to that technology is currently in its filing stage. Our modeling calculations (on both thermal and mechanical properties of the glazing) were completed in time and have been or are currently being published. Also a complete model of the cavity mass balance (pressure balance) was established showing the influence of various parameters on the expected service life. This model will be published in 2008 pending some initial results of carbon surface cleaning studies. To conclude we have met all of the goals set for this year and are on schedule. We are excited about upcoming trials to fabricate prototype glazing units for which we have already purchased and modified a large vacuum chamber. Prototyping and optimization of the sealing method for larger glazings will be the main focus of research during the upcoming year. Insulation performance and mechanical as well as thermal stability of our prototypes will be tested once they can be produced with confidence.

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