ABSTRACT

Insulating glass (IG) units significantly are contributing to fuel savings and the reduction of energy waste. Thus, the challenges of environmental care and green house gas (CO₂) reduction are supported by high-quality IG.

IG sealants have to meet manifold requirements such as ease of handling, efficiency, safety, cure profile, adhesion, elasticity, durability, noble gas or moisture vapour transmission rates (MVTR) and environmental friendliness. Noble gases used in IG units are Argon (Ar), Krypton (Kr) or Xenon (Xe). Sulfur hexafluoride (SF₆) performs very similar. Argon widely is used due to availability and price.

Polymeric raw materials for IG sealants (secondary seals) such as Polysulfides, PUR or Silicones strongly contribute to the success of today's IG systems. Among these polymers, Polysulfides have by far the largest market share in Europe (ca. 82%). The reason for the long success story of Polysulfide-based IG sealants is mainly the fact that customer expectations technically and economically are fulfilled for major IG applications. Niche applications like frameless overhead glazing or structural glazing are settled by other polymeric raw materials.

Keywords: Insulating glass, IG, IG units, sealants, energy saving, polymer, Polysulfide

1. INTRODUCTION

Driving forces for the development of today's multi-functional high-quality IG units with service life expectations of more than 30 years have been three major challenges:
- Reduction of energy wasting
- Noise control
- Comfort improvement.

The invention of float glass technology by Sir Alistair Pilkington in the fifties of this century made lots of high-quality flat glass panes available, starting multi-pane IG unit development. Heat transmission factors at this time were reduced from 6,0 W/m²K (single pane) to 3,2 W/m²K (double pane, air-filled), a big step forward. Today, a heat transmission factor of 0,4 W/m²K is possible in high-end IG units.

Different edge-sealing systems were developed, grew, survived or died out. The leading system which today is used world-wide for edge sealing can be characterized as following:
- Dual-sealed units
- Primary seal, PIB-based
- Secondary seal with elastomeric sealants

The edge seal strongly influences the quality of the whole IG unit. It is responsible for fogging control and copes with mechanical, chemical and thermal stresses. Certain reasons are responsible for the success of dual-sealed IG units with mainly Polysulfide-based secondary seals:
- Elastomeric properties
- Noble gas - and MVT rates
- Adhesion to substrates
- Chemical resistance
- Application and handling

In addition, new requirements for environmental friendliness, occupational handling safety, waste reduction and recycling are adding up to the existing list. Polysulfide-based IG sealants are meeting every requirement thanks to the versatility of this polymeric raw material base.

2. REQUIREMENTS FOR ENERGY SAVING

Energy saving has become not only an important economical factor but additionally it is a stringent requirement for environmental protection. International climate control conferences are proving the actuality of CO₂ and other green house gases reduction by efficient energy saving measures. One way to save natural energy resources is the thermal insulation of buildings, residential homes and offices. With respect to heat transfer through window panes, a significant reduction of heat transmission coefficients k was achieved by the introduction of multi-pane IG units (table 1).
defined niche markets (Table 3): 95 %). Other systems based on Butyl Hot Melts or primary and an elastomeric secondary sealant (ca.

IG units with air filling or single-pane units have no filling improves k_{IG} significantly. Traditional uncoated IG units with air filling or single-pane units have no chance to meet the requirements. Additional measures like noble gas diffusion factors k shall be limited. Two categories for new and old buildings exist. For windows, a total number k_{w,Feq} was introduced. If the building is a new one, k_{m,Feq} shall be ≤ 0,70 W/m²K. This number includes solar energy gainings from the infrared part of the sunlight spectrum (k > 780 nm). The whole window as an assembled part of the building has to be considered (IG unit, frame).

If the building is an old one, k_{w} shall be ≤ 1,80 W/m²K. This number is reflecting energy loss only through the IG unit. According to table 1, only IG units with low E-coated glass are able to meet this requirement. Additional measures like noble gas filling improves k_{w} significantly. Traditional uncoated IG units with air filling or single-pane units have no chance to meet the requirements.

3. EDGE SEALS IN EUROPE

Modern multi-functional IG units in Europe almost exclusively have edge seals made of a plastic primary and an elastomeric secondary sealant (ca. 95 %). Other systems based on Butyl Hot Melts or prefabricated Butyl Strips are only used locally in defined niche markets (Table 3):

<table>
<thead>
<tr>
<th>Year</th>
<th>Technology</th>
<th>k_{v} [W/m²K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;1950</td>
<td>S.p.*, traditional glazing, no coating</td>
<td>6,0</td>
</tr>
<tr>
<td>1960</td>
<td>D.p.*, air filled, no coating</td>
<td>3,2</td>
</tr>
<tr>
<td>1970</td>
<td>T.p.*, Ar filled, no coating</td>
<td>2,2</td>
</tr>
<tr>
<td>1978</td>
<td>D.p.*, pyrolytic hard coat., air filled</td>
<td>1,8</td>
</tr>
<tr>
<td>1984</td>
<td>D.p.*, sputtered soft coat, Ar filled</td>
<td>1,3</td>
</tr>
<tr>
<td>1994</td>
<td>D.p.*, magnetron sputtered coat., Ar/Kr filled</td>
<td>1,0</td>
</tr>
<tr>
<td>1997</td>
<td>T.p.*, two magnetron sputtered coat., Xe filled</td>
<td>0,4</td>
</tr>
</tbody>
</table>

* S.p.= Single pane; D.p.=Double pane; T.p.=Triple pane

Table 1: Development of heat transmission coefficients k_{v} for IG units in Europe over the years

In Germany recently new energy saving regulations were introduced by the federal government, effective 1.1.1995 (Table 2):

<table>
<thead>
<tr>
<th>Part of building</th>
<th>Max. heat transmission coefficients k_{v} [W/m²K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Walls</td>
<td>New buildings: k_{w} ≤ 0,5, Old buildings: k_{w} ≤ 0,5</td>
</tr>
<tr>
<td>Roof</td>
<td>k_{r} ≤ 0,22, k_{r} ≤ 0,30</td>
</tr>
<tr>
<td>Floor</td>
<td>k_{f} ≤ 0,35, k_{f} ≤ 0,50</td>
</tr>
<tr>
<td>Windows</td>
<td>k_{w,Feq} ≤ 0,70, k_{w} ≤ 1,80</td>
</tr>
</tbody>
</table>

Table 2: Requirements for residential buildings according to Germany's new energy saving regulations as of 1.1.1995.

For different parts of a building, heat transmission factors k shall be limited. Two categories for new and old buildings exist. For windows, a total number k_{w,Feq} was introduced. If the building is a new one, k_{m,Feq} shall be ≤ 0,70 W/m²K. This number includes solar energy gainings from the infrared part of the sunlight spectrum (k > 780 nm). The whole window as an assembled part of the building has to be considered (IG unit, frame).

If the building is an old one, k_{w} shall be ≤ 1,80 W/m²K. This number is reflecting energy loss only through the IG unit. According to table 1, only IG units with low E-coated glass are able to meet this requirement. Additional measures like noble gas filling improves k_{w} significantly. Traditional uncoated IG units with air filling or single-pane units have no chance to meet the requirements.

4. FUNCTIONS OF PRIMARY SEALS IN IG UNITS

Primary seals almost exclusively are made of Polyisobutylene (PIB)-based formulations. PIB has thermoplastic properties and is hot applied on the spacer bars by automatic lines. Also hand-application is possible with heated extruders or at room temperature with prefabricated strips. A new approach is a Hot-Melt system, replacing the entire spacer bar by extruded PIB/molecular sieve formulations (TPS system). However, typical IG units today still are made with metal or plastic spacers bars and PIB primary seal.

Table 4: Functions of primary seals in IG units

- Fixation assistance during unit assembly
- Moisture vapour barrier (MVTR very low)
- Gas transmission rates very low
- Protection of gas spaces in IG units

PIB-based primary seals are used in combination with secondary seals without incompatibility problems. Compatible polymer bases are Polysulphides, PUR or Silicones which are the most important ones for IG units. An important condition is a properly formulated secondary seal with sufficient reserves in polymer base, avoiding plasticizer migration, bleeding or staining.

5. FUNCTIONS OF SECONDARY SEALS IN IG UNITS

Secondary seals are established today as the result of the selection and evolution process in IG seals mentioned above. They are structural adhesives, binding the glass panes in multi-pane IG units (MIG) together. Secondary seals are commonly two-component sealants mixed before application, applied by robot or hand. They are forming rubber-like end products by chemical reactions of the base (A)-component and the accelerator (B)-component. Their backbone is an organic polymer
base, e.g. Polysulfide, PUR or Silicone. Thermoplastic hot-melt sealants are rarely used because of their high Shore A hardness at low temperatures. In niche applications, 1-part products are used, but their curing time generally is too long for fast delivery of the finished IG units (several days). 2-part IG sealants are curing within 1-4 hours after application.

Functions of secondary seals

- Structural bond of glass panes and spacers
- Protection of gas filled spaces from influences like
  - moisture vapour penetration
  - chemical attack (cleaning fluids, glazing products)
  - liquid water penetration (rain, condensation)
- Low MVTR and gas diffusion rates
- Stress relaxation
- Adhesion to glass and spacer substrates
- Durability

Table 5: Functions of secondary seals in IG units

The formulations of secondary seals in IG units are based on know-how developed by the manufacturing companies, but similarities in general principles do exist for two-component IG sealants.

Table 6: Key raw materials and functions for elastomeric secondary seals in IG units

6. POLYMERS FOR SECONDARY SEALS

Historically, the first synthetic rubber base was Polysulfide, found by the German chemists C. Löwig and S. Weidmann (1840). In 1924, Dr. J. C. Patrick rediscovered Polysulfides again in Kansas City (USA) and patented his results under the trade name "Thiokol". After different uses were found for solid and liquid Polysulfide rubbers, edge sealing of IG units was started in the fifties of this century. Therefore with Polysulfides 40 years of practical experience are existing in IG sealant production. Today Polysulfides dominate the application in secondary seals, followed by relatively small shares for PUR, Silicone and Hot Melt sealants. Other systems like Epoxy / Polysulfides or 1-part sealants have only marginal importance.


Table 8: Key properties of polymeric bases for secondary seals in IG units.
For Polysulfides, the profile of properties is at a level between good and excellent ratings without major disadvantages. Additionally, Polysulfide-based sealants for IG units are very tolerant against mistakes in production and forgive variations in temperature and mix ratio to a large extent. Mixing tolerances of up to 20 % for A- and B-components do not affect final cure properties and do not influence sealed IG unit performances. This unique property is hardly achievable by competitive systems.

Part A and B do have very similar viscosities, which supports mixing. There is no sensitivity of part A or B against atmospheric moisture or oxygen. Drums can be handled open and storage is possible without any additional requirements, as for temperature as for humidity. Polysulfide-based IG sealants are non-sagging and easy-flowing in automatic lines. Their cure profile is sharp and speedy after a variable open time. Cure reactions after mixing of part A and B can be stopped easily by freezing and e-started again at room temperature. Any crystallisations of ingredients are not possible during application.

Overall handling advantages as described strongly had contributed to today’s world-wide success of Polysulfide-based IG sealants.

PUR-based IG sealants are performing quite similar but with a few exceptions. Mixing ratios have to be kept strictly to get good cure profiles. Tolerances of max. ± 10 % are realistic. Noble gas diffusion is by a factor of 3-4 higher compared to Polysulfide, reducing service life with respect to heat transmission. MVTR is slightly better, compared to Polysulfide. Big problems have been observed with cured and crystallized iso-cyanate-based B components. Chemical resistance and UV resistance is lower due to residual double-bonds in the PUR backbones.

SIR-based IG sealants are used in applications where other polymers have difficulties. Their biggest advantage is the outstanding UV resistance. Structural glazing, skylights and frameless overhead-glazing are typical fields for SIR-based IG sealants. Additionally, applications at temperatures below -50°C are only possible for SIR. Problematic is the adhesion to substrates after constant water immersion. Noble gas transmission rates with a factor of 20-40 higher than Polysulfides are a hindrance for their use in gas-filled IG units.

Thermoplastic Hot Melts are difficult to compare with elastic polymer bases like Polysulfides, PUR and Silicones. However, in special applications it is possible to use Hot Melts with success.

7. NOBLE GAS-FILLED IG UNITS

Because of their excellent data for moisture and noble gas transmission rates, Polysulfide-based secondary seals can offset possible leaks in PIB primary seals, just in case of not properly made IG units. This is important facing a background of rapidly growing shares for noble gas-filled IG units and dropping numbers of standard IG units.

In 1996, ca. 80 % of IG units in Germany have been made as “special types”. They have been produced either with low E-coated float glass or with noble gas-filled spaces or with a combination. Polyurethane-based secondary seals are performing similar, but IG units with SIR-based secondary seals do not regularly pass the requirements of DIN 1286, part 2 or pr EN-1279-3 on noble gas loss rates. Loss rates of 1 per cent / year are regarded as still acceptable, which are regularly met by PIB/POLYsulfide-based dual edge seals.

<table>
<thead>
<tr>
<th>Noble Gas Gas</th>
<th>Number of IG units (Rosenheim 88-90)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Loss Rates [%/a]</td>
<td>Polysulfide</td>
</tr>
<tr>
<td>≤ 0,5</td>
<td>2</td>
</tr>
<tr>
<td>≤ 1,0</td>
<td>11</td>
</tr>
<tr>
<td>≤ 2,0</td>
<td>2</td>
</tr>
<tr>
<td>≤ 10,0</td>
<td>--</td>
</tr>
<tr>
<td>&gt; 10,0</td>
<td>--</td>
</tr>
</tbody>
</table>

Table 9: Statistical survey on gas loss rates for Argon gas filled IG units. Rosenheim testing institute results 1988-1990.

With loss rates of 1 % / a, Feldmeier and Schmid anticipate service lives for IG units of 20 years. Polysulfide/PUR-sealed units with loss rates of regularly less than 1 % can be expected to offer service lives of 30-40 years by this calculation. Noble gas loss rates are only one parameter of many, determining IG unit service lives, but a period of 30-40 years correlates with practical experiences.

In case of PIB primary seal leaks, leaking corner keys in the spacer bars or not properly sealed gas fill openings, the secondary seal thickness is responsible for offsetting noble gas loss rates or moisture penetration. With Polysulfides, thicknesses can be limited to 3-5 mm. PUR-and especially SIR-based secondary seals have to be made 10 - 30 % thicker (3,5 - 6,5 mm) to get similar safety reserves, which is a cost factor. However, noble gas loss rates of less than 1 % can be achieved in principle with every secondary seal system if the PIB primary seal is not leaking. With single-sealed units, consistently this target is not to achieve.

8. ENVIRONMENTAL ISSUES

Elastomeric 2-part sealants for IG units are complex blends of polymers and various chemicals. They have to be treated with care in their uncured state. For Polysulfides, Polyls and Polysiloxanes as key polymers in IG sealants no requirements for labelling according to Germany’s hazardous substance decree are existing. Thus, base components (A) remain unlabelled. For curing agents (B), the situation is different. Isocyanates (TDI, MDI) have to be labelled as hazardous substances (Xi, Xn or even

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also silicic acid esters. Manganese dioxide ($\text{MnO}_2$) pastes, required for Polysulfide bases are not regarded as hazardous substances in Germany if $\text{MnO}_2$ contents are below 25%. Above this limit, labelling as hazardous substance (Xn) is necessary.

Occupational sensitization or allergic reactions are not expected and have not been observed with Polysulfide-based IG sealants. Isocyanates, epoxy resins or silicic acid esters in contrast are known as possible sensitizers and allergens after occupational exposures. This has to be considered. Additionally some IG sealant bases for isocyanate cure are containing mercury accelerators at levels of more than 250 ppm.

During production of IG units, small amounts of cured and uncured IG sealants are generated as waste. In Germany, uncured sealant bases have to be treated as hazardous waste and must be handled accordingly (waste treatment key number 55 907). The correct treatment e.g. is done in plants suitable for chemical waste incineration or in ground-sealed landfill areas for hazardous substances. Cured sealants are not restricted and presently are deposited in landfill areas for non-hazardous household wastes or incinerated together with household waste (waste treatment key number 55 908).

For IG sealant bases (A) recycling systems have been established. Empty drums, big-bags and liners containing residual products are collected, transported to the sealant manufacturer and reworked. Thus, wastes are already reduced to an absolute minimum. For Polysulfide-based sealants, this recycling system additionally is applicable to the accelerator (B). In case of PUR or SIR sealants, bases frequently are crystallized or cured and are not suitable for recycling.

Recently a method was developed to recycle cured IG sealants based on Polysulfide and to use them again as raw material for new sealants. This step closes the loop for total recycling of IG units.

9. CONCLUSION

Fuel saving as an actual environmental challenge is supported by highly developed IG units in Europe. “Special unit types” are gaining market share, equipped with low-E-coatings and/or noble gas-filled spaces.

80% of German IG units were “special types”.

Leading principle for manufacturing of IG units is the dual-seal technology. PIB-based primary seals are generally used. Their main functions are:

- Fixing of glass panes and spacers during production
- Reduction of moisture vapour and noble gas transmission rates.

As secondary seal, elastomeric sealants are widely used. Base Polymers are (in order of importance) Polysulfides, PUR, SIR. Hot Melt Butyls are used occasionally.

Main functions are:

- Structural bond to glass panes and spacers
- Protection of gas space from various stresses (mechanical, chemical, moisture).

Dual-sealed IG units have service life expectations of more than 30 years. An important condition is a proper primary seal without interruption and leaks. 82% of the secondary seals are made with Polysulfide because of very good general properties and easy handling. SIR- and PUR-based sealants are used in other cases, e.g. if UV-resistance or structural glazing is required.

Handling and application of IG sealants is done by hand or with automatic lines. Occupational risks are very small due to non-hazardous base components. Accelerators for Polysulfide-based IG sealants in Germany are not regarded as hazardous substances at $\text{MnO}_2$ contents below 25%.

IG units will continue to develop. Future fields of improvement are the introduction of silicon panels for solar cell energy production. Improvements of low E-coatings are expected and the share of noble gas-filled IG units will grow constantly.

Sealants for dual edge-sealing will be present in the next millennium for successful IG unit production.

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