RELIABILITY OF PMMA FOR CPV LENS APPLICATIONS

Birgit Fassbender[1], Jochen Ackermann[1], Peter D. Colburn[2], Peter A. Marks[3], Peter Battenhausen[1] and Uwe Löffler[1]
[1] Evonik Industries AG, Darmstadt (Germany)
[2] Evonik Cyro LLC, Wallingford CT (USA)
[3] Evonik Cyro LLC, Sanford ME (USA)

1. Introduction

Polymethyl methacrylate (PMMA, PLEXIGLAS®[1]) has been used successfully in manifold outdoor applications like automotive lighting, signage & outdoor lighting, architectural glazing, aircraft glazing and transparent noise barriers over decades since its invention more than 75 years ago in Darmstadt, Germany. Its outstanding optical properties and unsurpassed weathering resistance in combination with robust handling make PMMA an attractive raw material for various solar energy applications and especially for optical systems in concentrating photovoltaic (CPV). With the overall photovoltaic (PV) market growing in the gigawatt range up to now the PV market dominates the building-integrated and rooftop applications, whereas solar farms are using CPV products (Kurz 2008). CPVs have gained interest since harvesting technique of direct vs. global radiation by tracking the sun has lowered the cost for each produced kWh (Miller et al. 2009). In this specific application the lens material is exposed around-the-year to extreme outdoor weather conditions of the Sunbelt region, which requires an excellent heat (temperatures of up to 85 °C) and UV resistance. CPVs in general use mirrors or lenses to focus UV-Vis and IR light onto a small area as little as 1 cm² of active semiconductor PV material based on a combination of group III and V periodic table elements (Fraas, Knechtli 1978). These so called multi-junction cells show very high cell conversion efficiencies of over 40.7% (King et al. 2007).

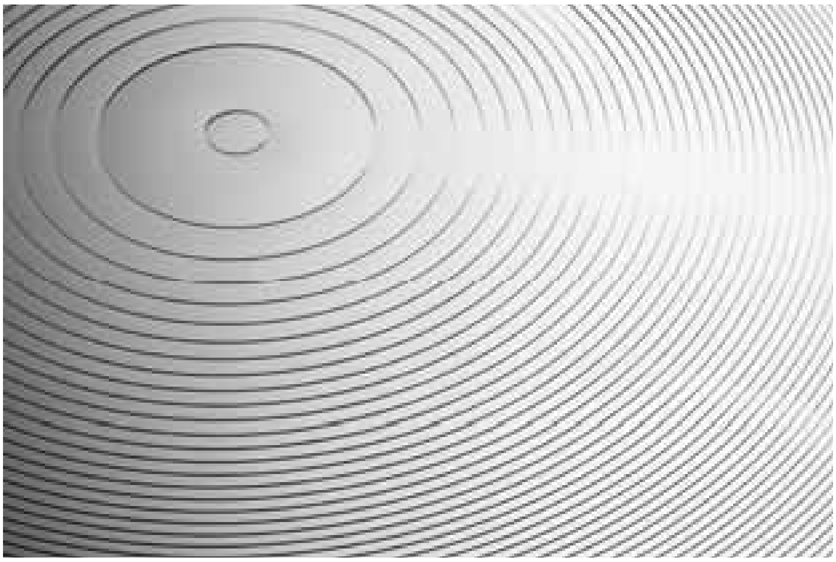

Fig. 1: Single Fresnel lens made from PLEXIGLAS® – diameter 250 mm, focal length 400 mm, thickness 3.5 to 8.5 mm. (Evonik Industries AG)

Beside of being very effective they are very expensive in production. Thus, the multi-junction solar cells in terrestrial applications should be used in combination with solar concentrators. The key for a decisive cost

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[1] Evonik Industries AG is a worldwide manufacturer of PMMA products sold under the PLEXIGLAS® trademark on the European, Asian, African and Australian continents and under the ACRYLITE® trademark in the Americas.

reduction in the production of electric power has to be the choice for low cost elements for optical concentration and sun tracking. Beside its good aging and optical characteristics, PLEXIGLAS® is adaptable to diverse processing techniques such as casting, extrusion, molding and thermoforming. Additionally, these techniques can form accurately dimensioned micro-structures being essential for high quality Fresnel lens production (Raihart, Schimmel 1975).

Ongoing CPV prototype development is governed by three major points: Performance, cost and reliability. Performance is optimizing the optical efficiency, cell cooling, and performance losses associated with manufacturing imperfections, soiling, tracking errors or thermal expansion/contraction. Cost implements the use of inexpensive components and the ease and the automation of assembly. Reliability investigates the long term degradation of optics or other loss of alignment, loss of adhesion or breakdown of bonds between cell and the optics and heat sink, etc. (Kurz 2008). Especially in the point of optical efficiency and reliability PLEXIGLAS® plays an eminent role and becomes more and more used in the CPV applications.

The state of the art CPV systems automatically track the movement of the sun across the sky to within 0.2° of accuracy. Such precision enables them to achieve efficiency levels of 20 to 24% in terms of electricity production, compared to 15 to 16% for modules with conventional silicon cells coming along with a overall cost reduction per kWh (Kinsey et al. 2010). Hereby, the weight of the panel has to be taken into account since using tracking systems consumes some of the produced energy. Therefore, the energy consumption of the system shall be as low as possible to run the system even more effective. The main competitor for PLEXIGLAS®, special silicone-on-glass (SoG) by having more than twice the density meaning twice the weight of PMMA increases the tracking costs. Costs of maintenance also have to be taken into account when calculating the energy payback of a solar cell. This value used to be over 20 years in the 1970’s while CPVs can do this in our days within less than a year (Peharz, Dimroth, 2005).

2. Findings about the behaviour of PMMA in solar applications

PMMA is synthesized by radical polymerization of methyl methacrylate (MMA). The polymer chain is of pure aliphatic structure and sterically protected by very stable ester groups.

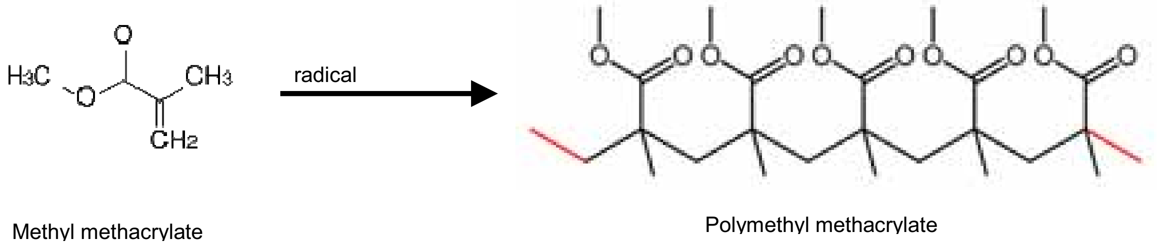

Fig. 2: Reaction scheme showing the radical polymerization of MMA.

Comonomers (methyl or ethyl acrylate) enable heat processing by inhibiting depolymerization. Additives (lubricants, plasticizers, stabilizers, colorants, fillers, …) help to adjust dedicated properties. Due to its thermoplastic nature it can be easily processed by injection molding, compression molding or extrusion. Among other transparent plastics (like e.g. polycarbonate) PMMA shows a well balanced portfolio of properties for outdoor applications. The main reasons in favour of using PMMA as plastic material for CPV applications (Fresnel lenses, mirror materials) are as follows:

- adjustable transmission (esp. in the UV region) enabling either prolonged lifetime of cell and lens (compared to UV transmitting grades) or increased module efficiency (compared to UV-blocking grades)
- outstanding optical properties like high transmittance, low reflectance, low dispersion and low scattering losses

- design freedom due to its excellent processing performance by injection molding, extrusion and hot-embossing
- excellent resistance to weathering and UV radiation leading to superior yellowing behavior and stable transmittance levels over long periods
- robust in handling due to high impact strength and light weight compared to inorganic glass
- possible recycling for readjusting into the production or cracking into monomers for repolymerization

3. Performance & Efficiency

3.1 Adjustable Transmission

In literature there are different definitions of efficiency in PV applications, among solar cell or lens efficiency. Apparently, optimizing the cost connotes optimization of the overall efficiency. In CPV applications the III-V multi-junction solar cell is the expensive building block and defines the spectral width and thus the remaining parts have to be matched e.g. the transmission range and optical efficiency of the lenses to guarantee the maximum possible light harvesting. In general, pure PMMA shows a high transmittance in the UV wavelength range starting from 250 nm and consequently can be used e.g. in solarium lamp coverage. For standard use, PMMA is usually modified with UV absorbance molecules to protect from aggressive UV radiation (c.f. Figure 3 grey dashed curve). In special cases also the use of standard PLEXIGLAS® in Fresnel lens applications might be of interest.

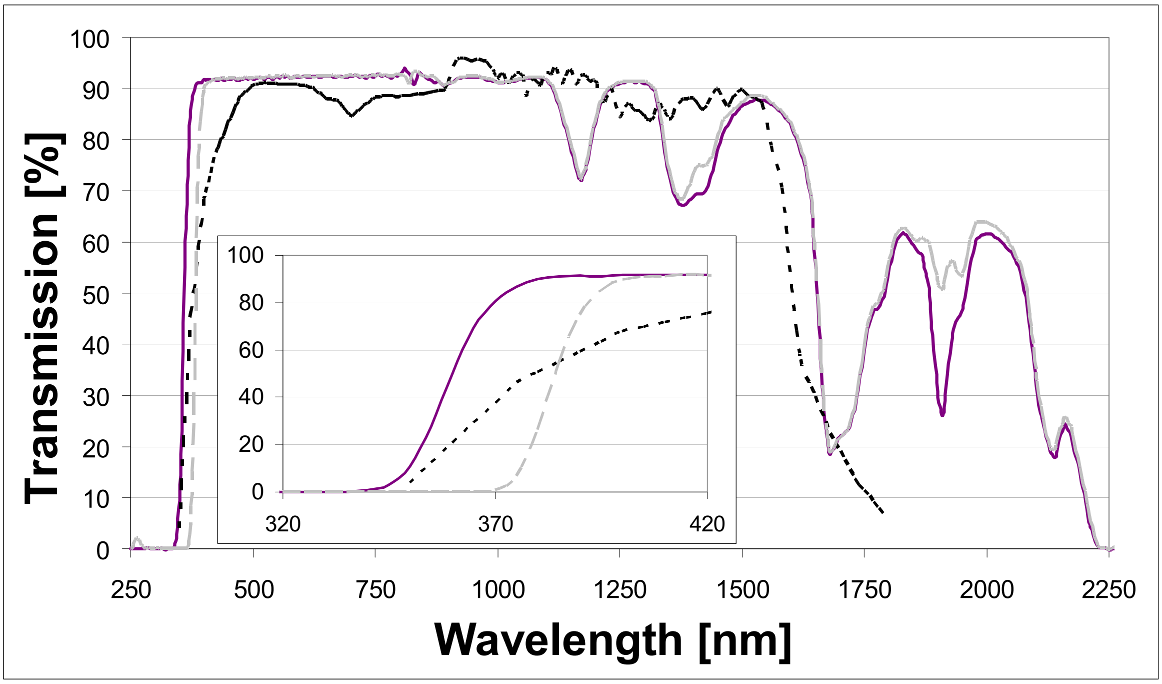

Fig. 3: Transmission data of UV-blocking standard PMMA grade, thickness of 3 mm (grey dashed curve), PLEXIGLAS® Solar IM20, thickness of 3 mm (purple curve), and the III-V multi-junction cell spectral response (black dotted curve). For UV-Vis, very good transmittance data are achieved, in the visible area 92.03%. Amplification shows the difference between standard PMMA and PLEXIGLAS® Solar IM20 in the UV range.

For the application in CPV the special grade PLEXIGLAS® Solar IM20 was developed where the onset of high transmittance was shifted specifically to a lower wavelength range. This alteration matches the transmission spectrum in the UV range with the absorption of III-V multi-junction PV cells resulting in optimum light quantum harvesting (c.f. Figure 3 purple curve and black dotted curve – amplified part). Here, the flexibility of PMMA is benefit since it can be tuned to the right wavelength in the UV range so even the smallest part of the solar energy can be used, meaning that also for future solar cells PMMA can be adjusted to the optimized wavelengths. The transmitted IR radiation of PMMA is less than in glass which is not disadvantageous at all. The photon energy is gained in the UV-VIS range and IR radiation apparently impedes the heating of the panel and hence the loss of solar cell efficiency (Miller et al. 2009).

3.2 Optical Efficiency

Good optical efficiency of Fresnel lenses depends on the surface structure and the chosen material. The ideal structure focuses theoretically 100% of the light on the PV cell. In reality this is never the case and all imperfections decrease the percentage of the focused beam. The two main reasons are due to thermal expansion of the material and imperfection of the Fresnel structure based on the fabrication.

The optical efficiency of PMMA and silicon SoG based Fresnel lenses were compared at constant temperature (20°C) and the real conditions (higher temperature) and were measured at different sunbelt sites by Hornung et al. 2011. Resulting that at 20°C SoG performs better then PMMA but as soon as the thermal influence defocuses the center of the beam the PMMA performance is comparable or even better to the SoG. PMMA shows at all measured sites an optical efficiency of 82.8±0.5% at 20°C and drops just to 81.8% under thermal influence while the loss in SoG can be up to 2% (Hornung et al. 2011). The overall conclusion is that PMMA and SoG showed in the operation temperature range a good and constant behavior. The higher transparency of SoG in UV region can be evaded by using PLEXIGLAS® Solar IM20 instead of the standard PMMA (c.f. Figure 2 grey curve). In terms of thermal defocusing the PMMA lenses show less variation than conventional SoG lenses which makes them more reliable. The smaller delocalization also could be essential in future Köhler design Fresnel lenses and no efficiency drops were observed for PMMA by Minano et al. 2010. Another advantage to assure the long term optical efficiency of PLEXIGLAS® is that lenses do not show as SoG the effect of delamination. The edge and the so called ‘sunburst’ delamination were observed in SoG structures of the PVB (poly vinyl butyral) type. The most common cause of this type of localized delamination is the use of clamping devices on the edges (Davies and Cadwallader 2003).

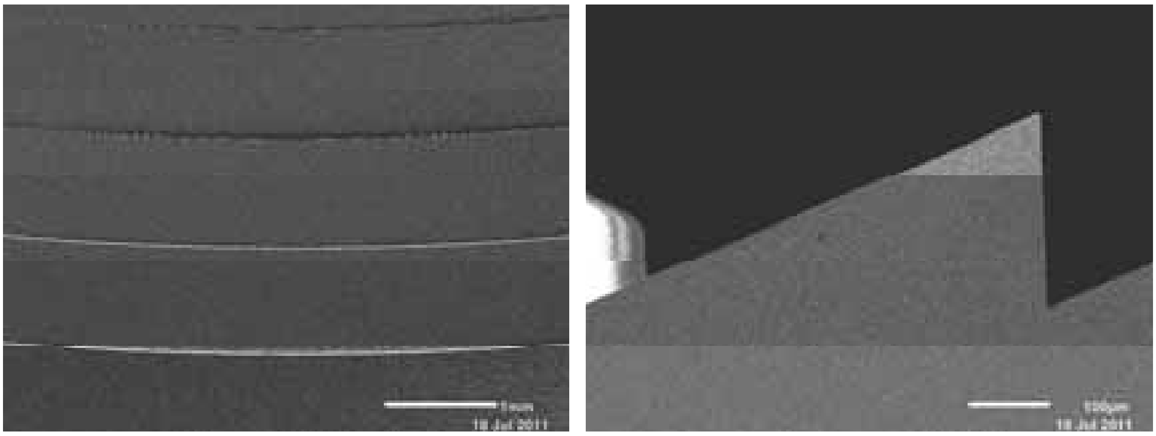

Fig. 4: REM micrograph shows the hot-embossed PLEXIGLAS® film lens. Left side is the top view and right side the breaking edge - view of the cross section.

The second decisive point for efficiency is the precision of the Fresnel lens structure. The REM micrograph in Figure 4 shows the top view in a distance of 3 cm to the center at the left side and a breaking edge cross section at a distance of 8 cm to the center at the right side. The tooth length D changes as a function of the distance changing from 860.1 μm to 605.0 μm in the window from 3 to 5 cm distance to the center of the lens. The tip radius was determined in the same window being 1.8 μm. To compare the quality of different Fresnel structures with different tooth sizes and at different positions on the lens the relation d/D is commonly used, where d is the tip radius. While the relation d/D in the range between 1% and 10% comprises medium definition structures, 0.5%, as is the case for the presented Evonik style, is in the range of a highly defined structure. For the optimal efficiency the draft angle shall be 90° but due to production this cannot be reached with hot-embossing. Thus, an obtained angle which is bigger than 88° is fairly good. Resulting in the average optical efficiency of > 86% at 660 nm and > 88% at 400 – 750 nm. The quality highly depends on the structure and therefore on the production method and its potential to mold an exact structure. Over the years different techniques were developed to produce the Fresnel structure on polymers such as PLEXIGLAS®.

3.3 Design freedom

3.3.1 Fabrication methods

Large quantities of Fresnel lenses for CPV power plants require a fully automated production process at short cycle times. Therefore, multiple production methods such as diamond turning, laser writing, lithography and e-beam writing are not efficient enough. Others, based on master tools are more profitable in terms of production time cycles. Here, especially injection molding, hot-embossing, and lamination are commonly used for the mass production of PMMA Fresnel lenses.

3.3.2 Injection molding

In general injection molding technologies using PMMA molding compounds have been proven to be capable of producing optical parts not only in high amounts but also in high quality. Such lenses showed a peak-to-valley deviation of less than 10 µm, nearly no internal stress and very low and homogeneous edge radii in the order of 5 µm (Luce, Cohen 2010). Here the advantage is that standard production equipment can be applied, and the cycle times are lower than can be reached with the hot-stamping of entire sheets. Production capacities essential for CPV application for solar power plants are in a range with an output of 10 ktons per year. This output requires an automatic process and low cycle times for lens production. Furthermore, Luce and Cohen demonstrated that the cost from a commercial point of view for small numbers is lower for hot-embossing, but for high quantities somewhere above 100.000 parts, injection molding is more competitive. For Fresnel lenses combined in panels compression injection molding is needed to reproduce the structure on a high quality level.

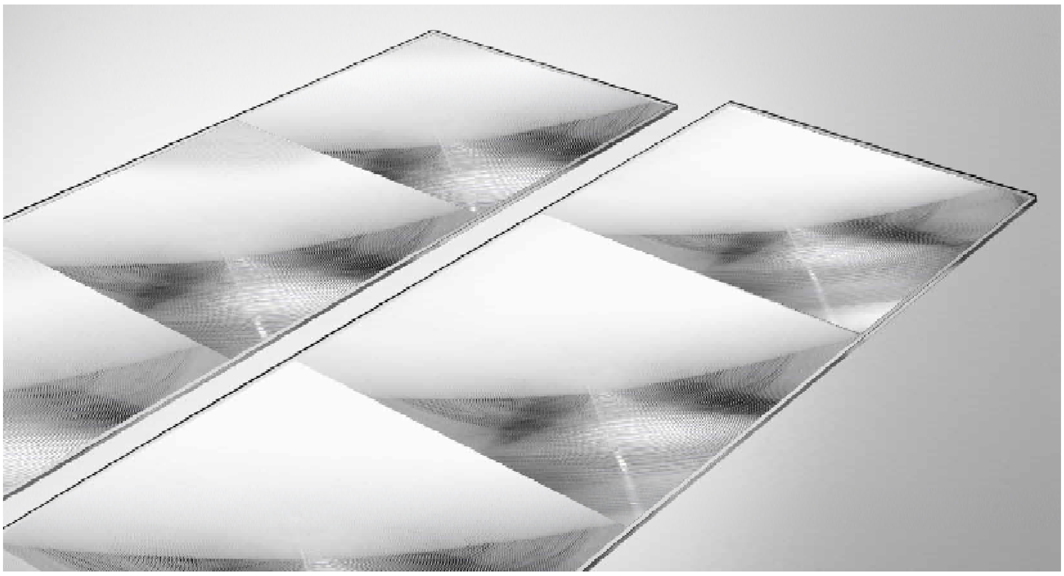

Fig. 5: Fresnel lens panels produced by a hot-embossed film being laminated onto a PLEXIGLAS® supporting sheet (Evonik Industries AG)

3.3.3. Hot-embossing

Hot-embossing is a production method replicating precise micro-structures onto plastic substrates (i.e. sheets or films). It has become a popular process since the creation of master molds with micro-structures became relatively easy. Nevertheless, the entire cycle usually takes more than ten minutes and the complete sample has to be heated above the glass temperature (T_g) and cooled down again. Therefore, this technique is rather time consuming for mass production and additionally causes high energy costs. Nonetheless, hot-embossing has become widely used for micro-structure fabrication with applications in optical sensors and biochips mainly because of the good and reproducible replication of the sharp features (Chang and Yang 2005). An alternative provided by Evonik Industries AG and its partners is to micro-structure just a film which is laminated in a second step onto a sheet. Here apparently the energy costs can be kept to a minimum and the processing speed can be accelerated when using an inline processing set-up.

3.3.4. Lamination

The idea of laminated Fresnel lenses was based on the plastic-on-glass fabrication and was first realized using a silastic rubber mold (Jebens 1980). Lamination combines the high structural quality of the hot-embossing and the higher output of injection molding. Thus, lamination for increasing quality is a desired alternative. Jebens showed 1980 that beside plastic-on-glass also plastic-on-plastic can be profitable using lamination. For PLEXIGLAS® micro-structured films being thermally laminated onto PLEXIGLAS® sheets no extra glue, such as PVB, silicone or TPU is needed which eliminates the major plastic-on-glass problem of potential delamination and thus minimizes the probability of system failure. This combination of techniques enables production of panel sizes as desired and consequently, the focus of all lenses in one panel is interconnected and no focusing problems occur due to further alignment (see Figure 5). The latter problem was overcome by using optimized glue in between the lens and the metal setup of the solar cell. Lamination inline with coextrusion renders the possibility to produce a Fresnel lens system with a potential third top layer making the surface facing the sun even more weather resistant.

3.4 Outstanding weatherability

3.4.1 Degradation mechanisms

Generally, pure PMMA shows high UV stability due to weak absorption in the UV region. However, even PMMA can experience degradation by different mechanisms: oxidation of residual monomer, chain scission and subsequent oxidation of low molecular fractions because of intense UV radiation combined with heat and humidity, oxidation of low molecular weight fractions from processing at high temperatures, mechanical cracking based on high stress levels. All of these effects are based on two major processes, photo- and thermal- degradation. These degradation mechanisms are the major impacts on PMMA's long term stability. Photo degradation refers to defects caused by high energy (UV) radiation and elevated temperature leads to thermal degradation. A broad overview of degradation mechanisms and its resulting molecules was provided by Miller and Kurtz 2011.

3.4.2 Photo degradation

The amount of photo degradation reactions in PMMA is relatively low compared to most other polymers being the base for the unbeatable weatherability of PLEXIGLAS®. The photo stability of PMMA is based on the fact, that little light is absorbed and thus can cause damage in the molecular structure. This low amount of UV absorption in PMMA is a result of the chemical structure. The homolytical scission of C-C and C-H bonds need 365 and 419 kJ/mol accordingly which are equivalent to the wavelengths of 335 and 285 nm which are not absorbed (Torikai et al. 1990 and Shirai et al. 1999). For the π–π^* transition of the carbonyl group in a non conjugated system, a wavelength of 190 nm would be necessary which is not part of the sun spectrum. Therefore, the main degradation is based on the forbidden which means underrepresented n-π^* transition of the carbonyl group. The mechanism is based on a quantum absorption and transfer of an electron to the non bonding π^* orbital which is the starting point for further reactions occurring at wavelengths around 380 nm (Torikai et al. 1995). The resulting reactions can be distinguished between Norrish type I and II. The β-scission of the polymer chain, a type I reaction, accelerates the degradation and causes a reduction in molecular weight and can be detected via e.g. gel permeation chromatography (Dickens et al 1984). While type II results in chain scission type I can provoke a manifold of small molecules which differ in presence or absence of oxygen (Grassie 1973). These reactions can be caused by residues such as additives, initiator or non reacted monomers.

3.4.3 Thermal degradation and oxidation

Normal temperatures do not play any roll in thermal oxidation of PMMA, even the elevated temperature in the accelerated weathering at 65°C does not show any effect on the structure (without the combination of UV irradiation). Also extreme conditions like the exposure of PMMA to pure oxygen at 120°C prove the stability of the polymer. Only some rare and unstable head-to-head combinations in the backbone are weak points in the PMMA structure at less than 200°C. The Vicat temperature of the PLEXIGLAS® Solar IM20 is around 110°C which defines the short term operation temperature of the Fresnel lenses which have to be form stable to keep the focus on the small PV solar cell. The temperature range which might cause thermal oxidation is not accessed in the CPV application of the primary lens. Higher importance than the chain oxidation is the increasing transportation of oxygen into the sheet or migration of monomer out based on the higher operation temperature (Dicens et al. 1986). It was shown by Furneau et al. 1980 that independent of the sun facing side the degradation of a polymer sheet under atmospheric conditions is symmetric and depends only on the distance to the surface as a result of the amount of oxygen which can access e.g. the additives for oxidation.

3.4.4 Countermeasures

To produce more stable PLEXIGLAS®, e.g. for Fresnel lens applications the pure PMMA is modified to assure a longer stability in the extreme application of CPV systems. A common method for UV protection is to use UV – absorbers which have to be chosen carefully and do not affect the intended applications (Zweifel et al 2009). Under UV irradiation the weak chain links are methacrylates suffering from some β-scission while acrylates preferably recombine after homolytic separation again (Grassie 1973). On the other hand the α-CH groups in the acrylate polymer backbone are predetermined for H-abstraction followed by auto-oxidation (Hatada et al. 1993). Thus, for a higher UV stability a copolymer of both monomers forms an even more stable PLEXIGLAS®. Another alternative approach is to cross-link the methacrylate chains (Lomakin et al 1993).

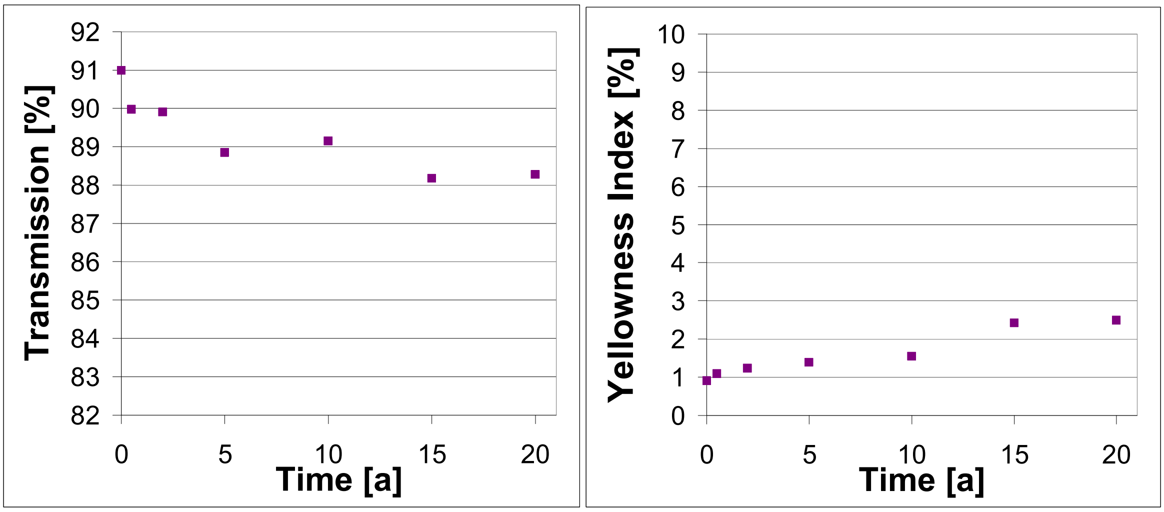

Fig. 6: Transmission data and yellowness index of standard PLEXIGLAS® of 9 mm thick sheet over 20 years exposed at Darmstadt (Germany) from 1971 to 1991.

3.4.5 Long term stability of PLEXIGLAS®

Due to reflection losses of approximately 4% at each surface the absolute transmittance in the UV-Vis wavelength of a virgin PLEXIGLAS® sheet is 92. The effective transmittance strongly depends on the meant application and thus on the exact composition of the PMMA, i.e. the amount of copolymers and additives. Additionally, the transmittance value depends at a minor rate on the thickness of the sheet as shows a sheet of 9 mm thickness in Figure 6 being 91% in the virgin state. The transmittance loss is not as eminent as it is the case of inorganic float glass where a change of up to 17% can be observed (Pilkington 2002). In an outdoor application, the transmittance of PLEXIGLAS® can be affected due to weathering. Notably, PMMA is known for its good stability for long time periods up to 25 years (Evonik 2011).

The outdoor performance over long time periods has been investigated by Röhm GmbH since the early 1940ies and is still under investigation today by its successor company Evonik Industries AG showing that the transmittance loss can be rather low: only 2% over a time period of 20 years (Schreyer 1970) which can be supported by some later test series showing a loss of 2.8% transmittance and an increase of the yellowness index to 2.5%– see Figure 6.

Tab. 1: Transmission loss of low Fe glass, float glass and PMMA in Rapperswil due to soiling and degradation after 20 years. Total loss is the sum due to soiling and degradation (Ruesch et al. 2008)

Sample	Number of samples	Soiling loss [%]	Degradation loss [%]	Total [%]
PMMA	6	7.0 – 8.3	0.6 – 1.7	7.3 – 8.6
Low Fe Glass	8	4.8 – 8.3	0.1 – 8.8	6.3 – 17.3
Float Glass	8	4.2 – 10.3	0.7 – 6.7	7.7 – 12.1

These results recently were supported by the SPF study (Ruesch et al. 2008) using among PLEXIGLAS® samples provided by Röhm (today Evonik Industries AG) and exposing them under intensive solar irradiation at an elevation of 1500 m in Davos (Switzerland) and moderate solar irradiation at Rapperswil (Switzerland). The amount of irradiation – such as it is required for CPV Fresnel lens or mirror systems – in Davos was 312 MJ/m² at a positioning angle of 60° facing south which is comparable with a sheet placed at an angle of 5° in Bandol (South France) with 313 MJ/m². Because of the higher altitude a higher UV irradiation was obtained in this latitude as compared to central Europe. It was shown by Ruesch et al. 2008 that the transmittance for PLEXIGLAS® dropped over 20 years hardly in Davos in average from 83.5% to 82.5% (c.f. Figure 7). The value of 83.5% transmittance depends on a composition which was not optimized for optical purposes. The second exposure site was Rapperswil (Switzerland) just 400 m above see level but next to Zurich city and its industries. With 236 MJ/m² at an angle of 60° this site can be compared with the amount of UV irradiation which occurs in Darmstadt (Germany) (214 MJ/m² at an angle of 5°).

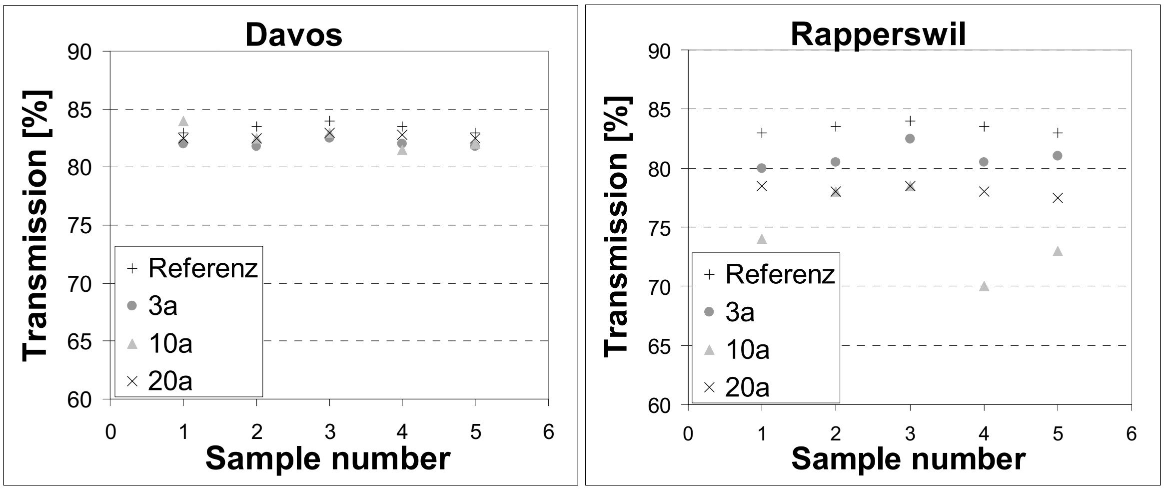

Fig. 7: Transmission data of PMMA sheets and PLEXIGLAS® provided by Röhm GmbH (today Evonik Industries AG); measurement without prior cleaning; exposed over 20 years at Davos (Switzerland) and Rapperswil (Switzerland). Data adapted from Ruesch et al. 2008.

These two data sets show that surface soiling more affects the transmission than UV damage. In detail Ruesch et al. 2008 found that the total average transmittance loss of PMMA was approximately 7.3 -8.6% while 7.0 – 8.3% was due to soiling (Table 1). The soiling loss could be eliminated by cleaning the sheets with ethanol or soap and after 20 years just 0.6 – 1.7% loss of transmittance was due to degradation (Table 1). This also explains the drastic loss in transmittance in Rapperswill after ten years. Here the researchers reported that it did not rain for several months before testing while after 20 years the transmission experiments were conducted shortly after a strong rain fall. Furthermore, they did not report any observable scratches as a result of the cleaning. The comparison of PMMA with low Fe glass and float glass in Table 1 shows that PLEXIGLAS® is not only comparable but on average even better than glass especially under the assumption that CPVs are cleaned regularly. Hence, PLEXIGLAS® Fresnel lenses for CPV in solar parks are located in the dessert and far away from environmental pollution. Consistent cleaning is necessary for all kind of CPV materials to obtain a constant and the maximum of transmission since in a desert self cleaning by regular rain can be neglected. Thus, the transmission loss of PLEXIGLAS® can be reduced to the degradation loss which is pleasantly low.

3.5 Handling & Recycling

3.5.1 Handling

Transmission loss as discussed above is a vital criterion for solar lens applications. One further issue is the loss based on light scattering as a result of surface pitting (Raihart, Schimmel 1975). PMMA is more prone to scratching as compared to conventional inorganic glass. Thus, scratch-resistant coatings are added to increase scratch resistance. In general, PLEXIGLAS® as a plastic compound has good scratch resistance compared to polycarbonates and can be further improved by using a scratch resistant coating (see Figure 8). Combined with the low brittleness the PLEXIGLAS® solution is a good alternative to inorganic glass.

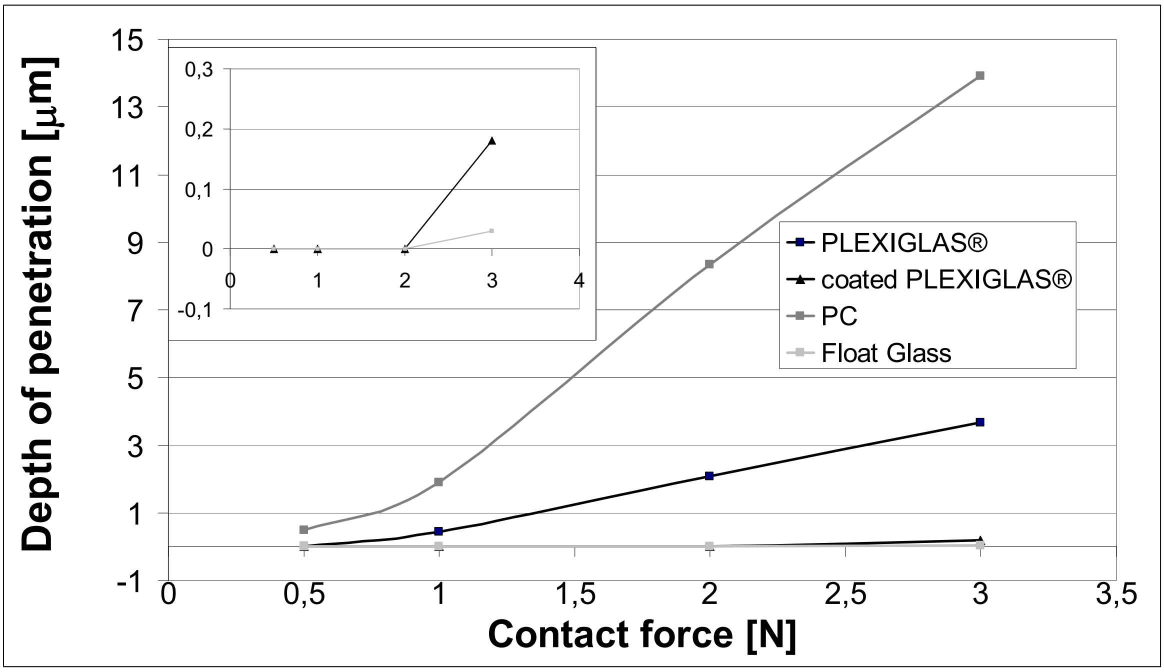

Fig. 8: Erichsen scratch-hardness test of standard PLEXIGLAS®, coated PLEXIGLAS®, polycarbonate (PC) and float glass. The measurement was conducted with a Universal scratch tester 413 from Erichsen equipped with a scratch diamante of 90µm tip radius angular speed of 5 rpm.

3.5.2 Recycling

Very few polymer materials are recovered, reconstituted, and/or upgraded, and then fed back into the system with the same material inherent properties as in their first lifetime – a process known as “closed-loop recycling”. Clean thermoplastic PMMA waste or scrap can be recycled without cracking. Instead it is heated in a reactor under coinstantaneous mixing and grinding above the glassing temperature and under the melting temperature (105°C < T < 160°C) and can be used once more e.g. for extrusion (Patent 2008). Thermoelastic and thermoplastic PMMA can be reprocessed to recover the different acrylic monomers. MMA is one of the rare monomers which can be recovered from the polymer by processes which are usually referred to as “cracking monomer” or “monomer crackback” meaning depolymerisation by introducing chipped scrap to a heat transfer medium where depolymerisation occurs at around 500°C. The obtained purity can be increased if needed by additional steps of distillation to more than 99% (Evonik 2011). Thus, using PMMA helps to save natural resources and saves money since 5% recycled PMMA lowers the total cost of the raw material up to 7% (Charmondusit, Seeluangsawat 2009).

4. Conclusions

Because of its adjustable optical and weathering properties PMMA is the leading plastic material for the use in CPV Fresnel lens or mirror systems. PMMA has shown a good track record in long-term outdoor applications – and in some real CPV installations. It is important to note that PMMA is not simply PMMA – one has to choose the right PMMA formulation and the right processing conditions to safeguard long-term outdoor applications under heavy solar irradiation conditions. Evonik Industries AG helps to make the right choice and provides not only the optimized molding compound with PLEXIGLAS® Solar IM20 for the production of e.g. injection molded Fresnel lenses but also a combination of hot-embossing and lamination technique based on an optimized PLEXIGLAS® grades to produce high quality laminated CPV lens panels.

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