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DURABILITY OF POLYMERIC GLAZING MATERIALS FOR SOLAR APPLICATIONS

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ABSTRACT

The economic viability of solar collector systems for domestic hot water (DHW) generation is strongly linked to the cost of such systems. Installation and hardware costs must be reduced by 50% to allow significant market penetration [1]. An attractive approach to cost reduction is to replace glass and metal parts with less expensive, lighter-weight polymeric components. Weight reduction decreases the cost of shipping, handling, and installation. The use of polymeric materials also allows the benefits and cost savings associated with well established manufacturing processes, along with savings associated with improved fastening, reduced part count, and overall assembly refinements. A key challenge is to maintain adequate system performance and assure requisite durability for extended lifetimes. Results of preliminary and ongoing screening tests for a large number of candidate polymeric glazing materials are presented. Based on these results, two specific glazings are selected to demonstrate how a service lifetime methodology can be applied to accurately predict the optical performance of these materials during in-service use. A summary is given for data obtained by outdoor exposure and indoor testing of polyvinyl chloride (PVC) and polycarbonate (PC) materials, and an initial risk analysis is given for the two

materials. Screening tests and analyses for service lifetime prediction are discussed. A methodology that provides a way to derive correlations between degradation experienced by materials exposed to controlled accelerated laboratory exposure conditions and materials exposed to in-service conditions is given, and a validation is presented for the methodology based upon durability test results for PVC and PC.

INTRODUCTION

Polymeric glazings offer significant potential for cost savings both as direct substitutes for glass cover plates in traditional solar collector systems and as an integral part of all-polymeric systems. A review of polymeric solar collector systems development efforts is provided in [2]. Cost savings result from lower base material costs, lower manufacturing costs, and lower costs associated with shipping, handling, and installation. Glazings must have high hemispherical transmittance across the solar spectrum and must be able to survive at least 10 yr of exposure to service conditions including solar ultraviolet (UV) light and operating at temperatures of 0 to 90°C. They must also retain mechanical integrity (e.g., impact resistance and flexural rigidity) under these harsh environmental stresses. The emphasis of current efforts is to identify new or improved candidate glazings and to evaluate their optical and mechanical durability during exposure to actual and simulated in-service conditions.

Recently, several reviews of candidate polymeric glazing materials have been undertaken [3-5]. These were guided by the expectation that advances in the polymer manufacturing and materials industry would allow identification of potential new and improved collector glazing candidates. An international collaborative effort surveyed commercial producers of advanced polymer materials in the United States, Europe, and Japan [3]. The most promising class of polymers was fluoropolymers. These have excellent thermal and optical durability, but are expensive and are limited to use with thin-film collector designs. Film products such as Tefzel[®] (ethylene-tetrafluoroethylene copolymer; ETFE), Duralar[®] (also an ETFE), Halar[®], Teflon[®], and Kynar[®] exhibit very high

spectral transmittance, and many have sufficient tear resistance to be considered as collector glazings [4]. Suitably UV-stabilized polyetherimide (PEI), polyimide (PI), and polycarbonate (PC) were also suggested for consideration, although PI is quite expensive. Preliminary exposure test data for several dozen polymeric glazing materials being screened were reported in [4]; most materials identified by [3] were included. These exposures have continued and further results are discussed herein. Another complementary review also surveyed potential polymeric glazing materials [5]. In addition to twin-walled PC, fluoropolymer films, and multilayered polyethylene (PE) films under test, consideration of polyurethane films, silicones, enhanced acrylics, clay-filled thermoplastics, and polycyclohexylethylene (PeCHE) was also recommended.

Screening tests have revealed the more promising candidates, along with glazing materials that have failed. The most common mode of failure has been yellowing of the glazing material. These have generally included non-fluoropolymer thin-film materials (PET and PE, including UV-stabilized versions), and non-UV-stabilized PC constructions. Additional (less common) modes of failure include materials developing a cloudy white opaque appearance, temperature-related deformation and/or discoloration, and physical damage caused by hail and other natural weathering events. Materials that have maintained high solar-weighted hemispherical transmittance values (>90%) after more than 2 years outdoor and accelerated exposure include: Kynar[®], Duralar[®], Tefzel[®], and Halar[®], and PC with UV-screening layers.

Polycarbonate has high optical clarity and excellent impact strength. However, it will yellow during UV exposure and become brittle. Recently, stabilized versions of PC have been developed. For example, Bayer has two products designated APEC 9351 and APEC 9353. The first is a thermally stabilized formulation, which is offered for a maximum continuous-use temperature of up to 180°C, and the second is stabilized for UV exposure and elevated temperatures. General Electric has incorporated an integral UV-screening coating into a number of its Lexan products. Because PC has excellent initial properties and is available in a variety of forms (e.g., sheet or channeled) suitable for use with solar collectors, it has been studied extensively as a

promising glazing candidate. Parallel test results for PVC serve as a control, because it is known to weather poorly.

DURABILITY EXPOSURE TESTING

From 1993-2002, numerous samples of PVC and PC materials were exposed to accelerated lifetime testing in laboratories and to the outdoor environment at test sites located in Europe and in the United States by colleagues participating in the IEA Task 10 Working Group. The details of these test results are provided in [6]. Samples of PC and PVC, along with other candidate polymeric glazing materials, were subjected to in-service outdoor and accelerated laboratory exposure conditions.

Outdoor Exposure Testing

Outdoor testing was carried out in Switzerland at the Institut für Solartechnik (SPF), in Germany at the ISE in Freiburg, and at three sites in the United States: Golden, CO; Phoenix, AZ; and Miami, FL. A precise and detailed knowledge of the specific environmental stress conditions experienced by weathered samples is needed to allow understanding of site-specific performance losses and to permit service lifetime prediction of candidate glazings. Consequently, each operational exposure site is fully equipped with the appropriate meteorological and radiometric instrumentation and data-logging capability.

The materials tested are those intended for use in solar thermal flat-plate collectors. Thus, the samples for outdoor exposure were attached to mini-collector boxes, as illustrated in Figure 1. To simulate the elevated temperature that collector covers are exposed to, the “mini-collectors” are made of solar selective-coated stainless steel (solar absorptance = 0.86 ± 0.03 ; thermal emittance = 0.14 ± 0.03). A thermocouple is affixed to the glazing material to monitor sample temperature, and a reflective light shield hood is used to prevent direct heating of the thermocouple.

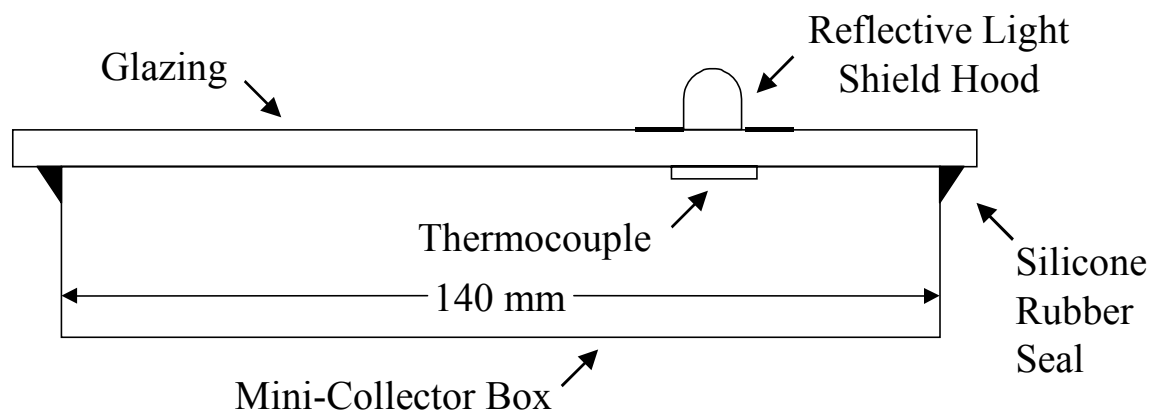


Figure 1: "Mini-collectors" used for outdoor exposure of transparent cover

The samples prepared in this way were exposed to the ambient climate at locations in Europe and in the United States, facing south at an inclination angle equal to the latitude of the site. The spectral hemispherical transmittance of all samples was measured prior to exposure. After some time, some of the samples were remeasured and exposed again without any cleaning. Other samples were measured before and after cleaning and then exposed again. Solar-weighted transmittance values integrated over the solar spectrum (τ_{sol}) and between 400-600 nm ($\tau_{400-600}$) are computed as degradation indicators. The bandwidth 400-600 nm is useful because degradation of optical transmittance of many polymeric glazing materials is most pronounced in that spectral range.

Representative $\tau_{(400-600)}$ data are plotted in Figures 2 and 3 for APEC 9353 PC and Duroglas PVC, respectively. The sawtooth appearance in these figures is indicative of two measurements after the same time of exposure, with the lower value being before cleaning and the higher value after cleaning. The data points represent average values of typically 2-3 replicate samples. Figure 2 shows a loss in $\tau_{(400-600)}$ of about 5% per year for samples of APEC 9353 exposed outdoors in Europe and the United States. Duroglas, a PVC glazing material, degrades very rapidly when exposed to UV light. Figure 3 presents outdoor test results for Duroglas exposed in the United States and Europe.

Samples of Duroglas exhibited between 25%-40% loss in $\tau_{(400-600)}$ after only one year of exposure, depending upon the outdoor site.

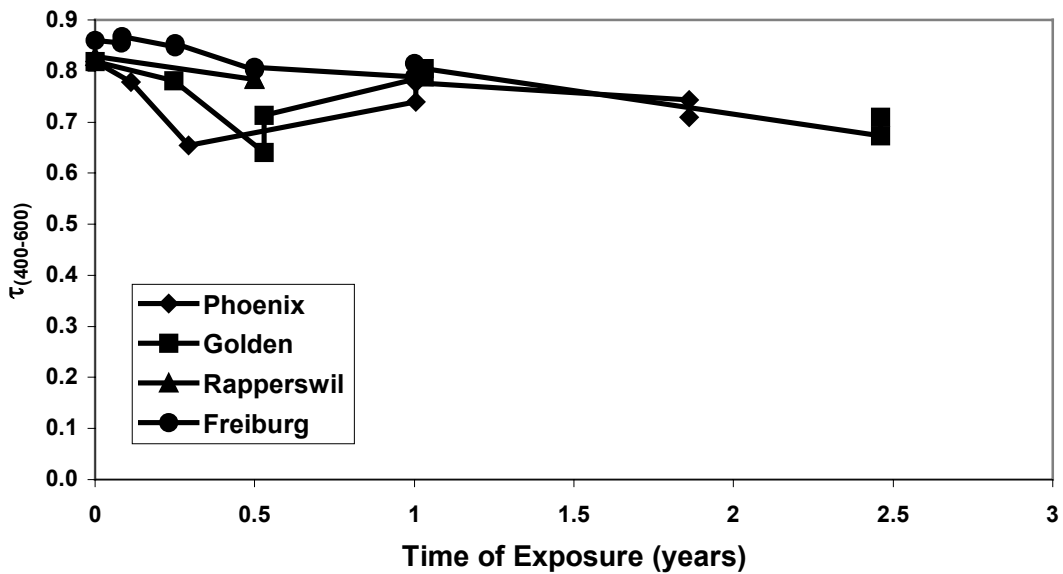


Figure 2: Outdoor exposure test results for APEC 9353 PC

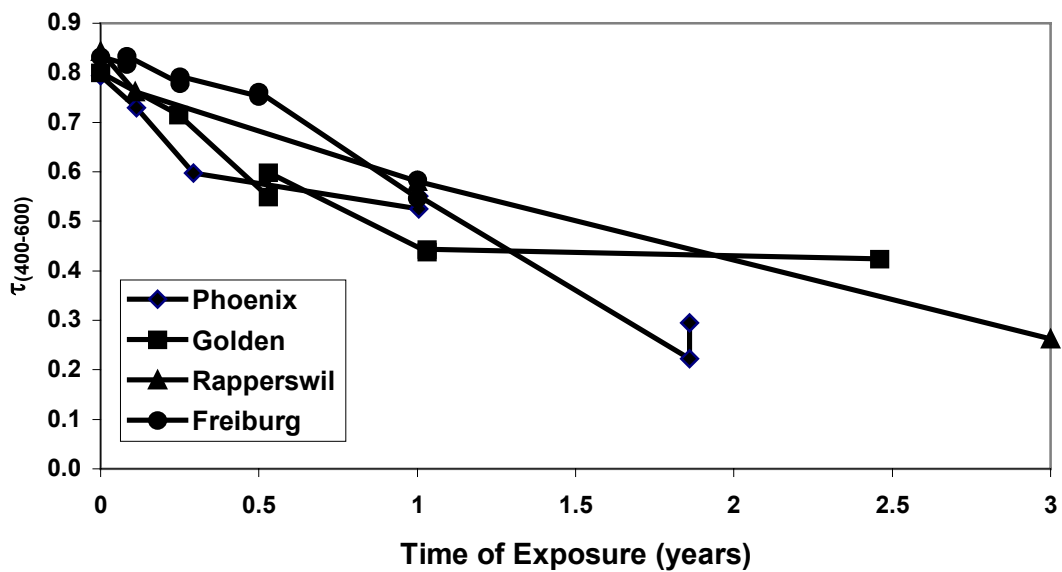


Figure 3: Outdoor exposure test results for Duroglas PVC

Accelerated Laboratory Exposure Testing

Accelerated indoor testing was carried out with different types of test equipment available at the participating laboratories. Several test protocols were performed using corresponding types of exposure chambers. In the first type,

UV exposure was combined with various combinations of elevated temperature and a defined level of relative humidity (RH), i.e., 60°C/80% RH, 80°C/40% RH, and 50°C/95% RH. These tests were performed in climatic cabinets in Rapperswil and Freiburg with an unfiltered metal halide (HMI) lamp as a light source. The intensity of the irradiation compared to an air-mass (AM) 1.5 solar spectrum is about three times as much UVA and seven times as much UVB. Some irradiance occurs at wavelengths below a terrestrial AM 1.5 spectrum. In the second type of exposure test, an Atlas Ci5000 Weather-Ometer[®] (WOM) was operated at 60°C, 60% RH, and an irradiation level of about twice an AM 1.5 solar spectrum throughout the UV and visible portion of the spectrum. In the final test protocol, an Atlas XR35 WOM – SPART 14 test was used. The SPART 14 test procedure was originally developed for clear coats in automotive paint systems. This weatherability test includes acidic rain spraying. In test method SPART 14, which is a modification of SAE J1960 [7], the Xenon arc light source is filtered through borosilicate filters and has an irradiance level of 0.5 W/m² at 340 nm; this corresponds to an intensity of roughly 1.4 times an AM 1.5 spectrum. The test cycle comprises (a) 40 min of light only; (b) 20 min of light with water sprayed on the front surface of the sample; (c) 60 min of light only; and (d) 60 min of no light with water sprayed on the back surface of the sample. Every 14th cycle, the water used to spray the front of the samples is acidic, with a pH of 3.2. The black panel temperature and relative humidity during light periods are 70°C and 75%, respectively. The chamber temperature and relative humidity during the dark periods are 38°C and 95%.

An exposure time of 1000 h (~6 weeks) in the SPART 14 test is estimated to correspond to about 1.3 years of outdoor testing in Miami, Florida, for automotive paints. Thus, 4000 h of SPART 14 testing corresponds to about 5 yr outdoors in Florida. However, one can assume that the temperature of an automotive coating will be at least 10°C higher than for transparent, low light-absorbing glazing materials. Consequently, the acceleration factor for the glazing can be estimated to be a factor of 2 higher. Accordingly, 1000 h of artificial weathering corresponds to 2.5 yr outdoors.

Highly accelerated exposure testing of selected samples using natural sunlight was also performed at NREL [8]. Parallel testing with the relevant stress factors of UV, temperature, RH, and acid spray at different levels was intended to allow the sensitivity of materials degradation to these factors to be quantified, and allow damage function models to be evaluated. These in turn can be used to compare the time-dependent performance of these materials with measured results from in-service outdoor exposure.

The values obtained for $\tau_{(400-600)}$ are plotted in Figures 4 and 5 for APEC 9353, and Duroglas, respectively, after the different types of exposure. Figure 4 shows that results for APEC 9353 exposed in the SPART 14 chamber are in good agreement with Ci5000 data. However, exposure of APEC 9353 in the unfiltered metal halide chambers is much more severe than in the Ci5000 and SPART 14. With the unfiltered metal halide light source, a ~15% loss in $\tau_{(400-600)}$ occurs after only 25 days, whereas it took roughly 100 days for an equivalent loss to occur in the Ci5000 and SPART 14. This may be explained by low-wavelength (below 300 nm) irradiation associated with the metal halide light sources.

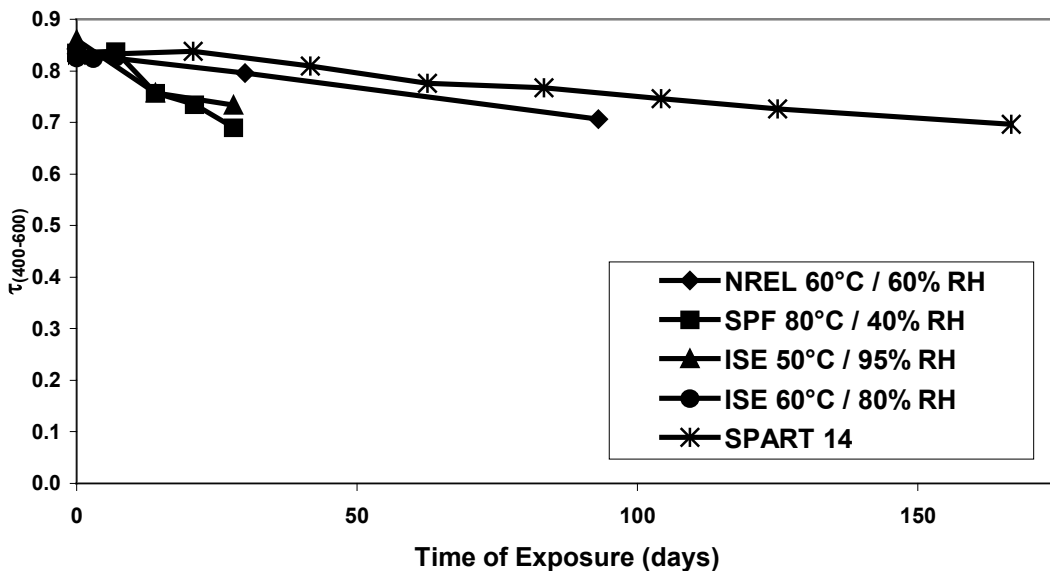


Figure 4: Accelerated exposure test results for APEC 9353 PC

Accelerated exposure test results for Duroglas PVC are provided in Figure 5. Exposure of Duroglas in the unfiltered metal halide chambers at a variety of temperature and RH conditions produced rapid degradation. Results for Ci5000 WOM exposures were less severe, although precipitous degradation did occur in fairly short time periods. Duroglas exposed to the SPART 14 chamber conditions results in considerably less degradation than for the unfiltered metal halide and Ci5000 chamber exposures.

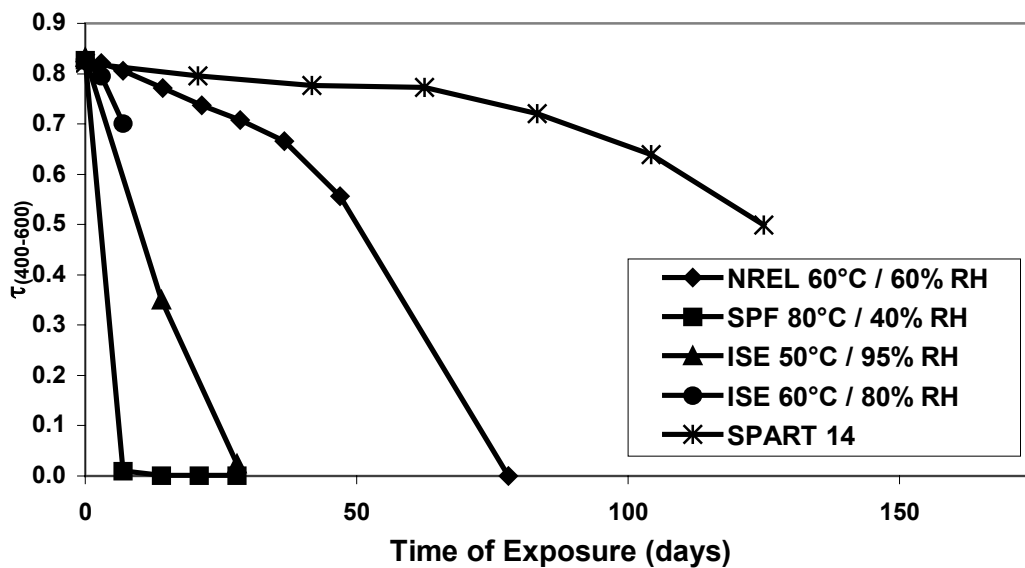


Figure 5: Accelerated exposure test results for Duroglas PVC

The approach developed by the IEA Working Group on Materials for Solar Thermal Collectors [9] is applied below to PVC and UV stabilized PC coverplate materials to illustrate how the general methodology can be used to assess the durability of polymeric-type materials.

ANALYSIS OF DURABILITY RESULTS FROM ACCELERATED AGING

Using artificially aged samples from screening tests, changes in the key functional properties or the selected degradation indicators are analyzed with respect to the associated changes in the materials. The analyses were made to identify the predominant degradation mechanisms of the materials. Possible mechanisms of degradation of the PC glazing were assumed to be (a) photooxidation (PO), (b) thermal oxidation, and (c) combined photooxidation

and hydrolysis. From the screening tests, it was concluded that only photooxidation contributes significantly to the service life of the glazing. A suitable time-transformation function is:

$$a_{PO} = \frac{\left(I^p \cdot e^{-E/kT} \right)_{acc}}{\left(I^p \cdot e^{-E/kT} \right)_{ref}} \quad (1)$$

where I is the intensity of photoreactive light, T is temperature, E is an activation energy, p is a material dependent constant, “acc” is accelerated test conditions, and “ref” is some set of reference conditions (e.g., in-service conditions).

For the PVC glazing, degradation mechanisms that could reduce the service life were assumed to be (a) dehydrochlorination, (b) photooxidation, and (c) thermal aging. For (a), the mechanism is a chain reaction type because hydrogen chloride formed from the dehydrogenation reaction also acts as a catalyst for this reaction. The reaction is consequently difficult to model mathematically in a simple way. Thus, it is difficult to express the rate of degradation in terms of a time-transformation function. The best time-transformation function for the PVC degradation was the same general photooxidation time-transformation function used to model the degradation of the PC glazing (Eq. 1).

During life-testing, PC and PVC glazing materials were exposed to the various accelerated conditions discussed above. Hemispherical transmittance measurements were made to characterize the loss in optical performance of the glazing materials during these exposures. Performance-versus-time data were thereafter used to determine the parameters of the time-transformation function (Eq. 1). The results, obtained from a subset of the data accumulated, are shown in Table 1. Values of the derived activation energies, E , are reasonable for photo-thermal degradation mechanisms. The value of $p \sim 2/3$ for PVC indicates that some shielding or rate-limiting reactions occur and do not allow all photons to participate in degradation. For the UV-stabilized PC

sample, the value of $p \approx 1$ indicates that exposure of this material follows strict reciprocity. Thus, all incident photons fully contribute to the degradation reactions, even at elevated levels of irradiation.

TABLE 1
Coefficients derived from accelerated exposure
for the tested polymeric glazing materials

Polymer Glazing	p	E (kcal/mole)
PVC	0.669	8.440
UV-Stabilized PC	1.093	6.688

VALIDATION OF METHODOLOGY

If it is assumed that the rate in transmittance change is constant if the surface temperature and the UV-light intensity are maintained at the same values during the time interval Δt_i , then the transmittance change $\Delta \tau_i$ may be expressed as

$$\Delta \tau_i = A(I_{UV})^p \Delta t_i e^{-E/kT} \quad (2)$$

using the time-transformation function shown in Eq. 1. The parameter A is a constant independent of surface temperature and light intensity, but it is material dependent. It may be determined from the same series of aging tests used to determine the activation energy E and the parameter p . For $\Delta \tau_i$ equal to the mean hemispherical transmittance between an even narrower bandwidth (400 and 500 nm; chosen to accentuate the region over which degradation occurs, resulting in a more highly sensitive degradation indicator), the values of A were estimated as $2892 \text{ (MJ/m}^2 \text{)}^{-1}$ for PVC and $5.497 \text{ (MJ/m}^2 \text{)}^{-1}$ for UV-stabilized PC.

By integrating Eq. 2, Eq. 3 is obtained.

$$\Delta\tau_i(t) = A \int_0^t [I_{UV}(t)]^p e^{-E/kT(t)} dt \quad (3)$$

Applying Eq. 3, the expected transmittance after different time periods of outdoor exposure may be estimated.

Using the values of the coefficients E and p from Table 1 and the time-monitored values of sample temperature and UV irradiance, the loss in performance was predicted for both the PVC and the UV-stabilized PC as exposed outdoors in Golden, CO, and Phoenix, AZ. Predicted values were then compared with measured data for these materials exposed at these sites. The results are shown in Figure 6. The time-dependent changes in the weathering variables result in the irregular shapes of the predicted curves. Excellent agreement is evident between the measured and predicted data. Thus, the phenomenological approach to data analysis is validated (i.e., obtaining model coefficients from accelerated test results and then using these coefficients to predict time-variable in-service degradation).

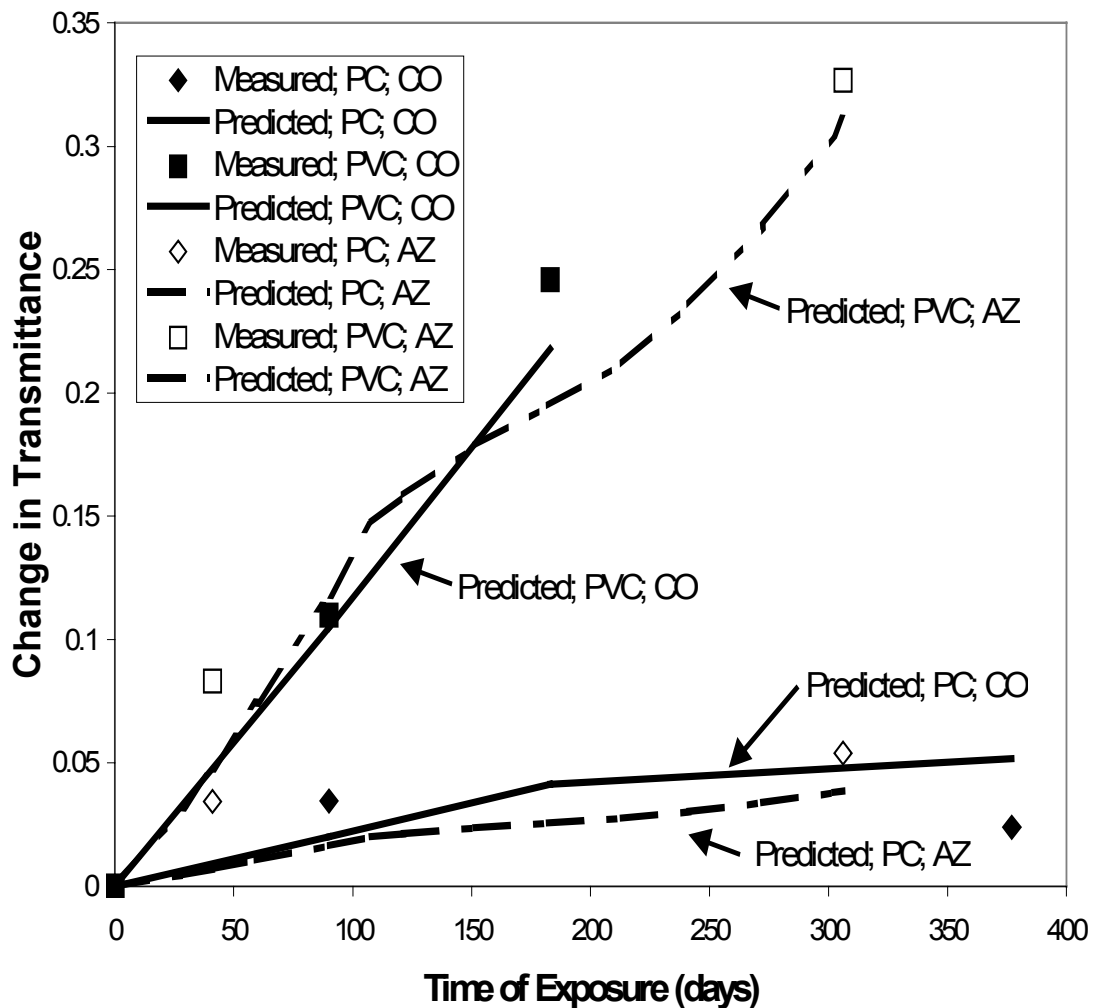


Figure 6: Comparison of the actual and predicted changes (loss) in hemispherical transmittance between 400-500 nm of PVC and PC for exposures of up to 380 days at Golden, CO and Phoenix, AZ.

CONCLUSIONS

Durability test data for both accelerated laboratory conditions and outdoor in-service conditions have been presented for PC and PVC glazing materials. Some of the accelerated exposure data were used to demonstrate how to derive damage functions that allow prediction of performance degradation. This methodology also allows the effect of multiple stress factors to be modeled. The usefulness and validity of this approach has then been confirmed by comparing predicted results with measured data for samples exposed to variable outdoor conditions. Consequently, highly abbreviated testing times at elevated stress

conditions can be substituted for long-time exposures at lower stress levels. The procedure developed allows much shorter development cycle times for new materials and allows improvements to be identified and readily incorporated into new products.

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