DEVELOPMENT AND OPTIMIZATION OF BACK SURFACE ACRYLIC SOLAR MIRRORS TO ASSURE 25 YEAR LIFETIME PERFORMANCE

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Dedicated to science and the pursuit of progress.
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Development and Optimization of Back Surface Acrylic Solar Mirrors to assure 25 year lifetime performance

Abstract

by

MYLES MURRAY

0.2 Abstract

Use of materials without sufficient track records, in technologies that perform outdoors for long periods, may result in premature degradation or outright failure; however many unproven material systems have not been evaluated for their lifetime performance. In addition, they can present opportunities to reduce system costs without negatively impacting performance. Lifetime and degradation science (L&DS) is used to inform material choice, by implementing accelerated weathering protocols alongside periodic evaluations cross-correlated using a stress-response framework, to mitigate risk of premature performance loss. L&DS was implemented for Replex Plastics in development of a novel, low cost, solar-grade mirror with a 25-year performance. New performance metrics for solar mirrors were developed and used to determine which of a set of acrylic substrates should be used in the construction of these solar-grade mirrors. Two formulations of protective coating were evaluated and possible degradation modes for aluminum back-surface reflectors were determined. Accelerated weathering protocols that induce damage from UV light, temperature, humidity and corrosive environments were performed on bulk materials and components. Analysis of the bulk material transmission, mirror
reflectance, and scattering functions were correlated with accelerated weathering protocols for this technology development endeavor.
1 Introduction

1.1 Lifetime and Degradation Science: Applicability to Renewable energy

A typical method for determining the cost of a renewable energy technology relies upon two metrics: the cost of the system, the initial performance of the system. In Photovoltaics, cost (C) and performance (P) are typically tied together. Cells, modules, arrays and assemblies are typically rated in terms of $/W, a cost/performance metric. While this metric is useful for comparing the cost of PV systems, it does not necessarily reflect the value of the energy that the system will produce. The value of energy produced by a PV system is highly dependant on two more metrics: the local solar resource and the durability of the system. The durability of a system (P(t)) represents the time dependant nature of performance 1.1. If a system undergoes a loss of performance over time then

\[ P(t) = P_i - R_d t \]  \hspace{1cm} (1.1)

where \( R_d \) is the degradation rate of the system. Typically degradation rates are calculated after a number of full systems have been fielded. Crudely, the value of electricity
generated is calculated by dividing lifetime system costs (initial investment plus operation and maintenance costs) by total energy provided by the system over its life (equation 1.2). If all costs and market trends are counted for, this value is called the levelized cost of electricity (LCOE) (equation 1.3).

\[ Value = \frac{cost(\$)}{electricity\ produced(kWh)} \quad (1.2) \]

\[ LCOE = \frac{\Sigma cost}{\int P(t)} \quad (1.3) \]

Therefore, when comparing systems of different generation types, LCOE is a function of lifetime, degradation rate, initial cost, operation cost and performance, shown in figure 1.1. Performance is incumbent on the conversion efficiency of the technology and the available resource. It is important to note that for PV systems the degradation rate and the available resource may be linked, as sunlight is one of the factors inducing degradation while concurrently dominating system performance through lifetime.

In order to mitigate risk associated with fielding large numbers of full systems in multiple locations for validation purposes, lifetime and degradation science can be implemented in order to make appropriate material choices early on in development.

1.2 Lifetime and Degradation Science: Linking Mechanisms to Performance

Service lifetime prediction (SLP), a common context in which to discuss material and component lifetime, requires knowledge of the interactions of bulk materials, interfaces, and interactions with the environment\textsuperscript{2,3}. Because typical PV lifetimes are longer than
product development cycles, waiting for real-time testing is not a viable option. Determination of the primary degradation modes and determining meaningful ways to accelerate those degradation modes can be helpful. SLP is primarily concerned with mitigating risk of early failure and warranty claims, where early failure is characterized by falling below a preset performance parameter$^{4-6}$. 

Figure 1.1. It is the intersection of Cost, performance and reliability that determine the cost of electricity
A recent U.S. Department of Energy report\textsuperscript{7} identified L&DS as a critical meso-scale scientific challenge. The challenge of understanding degradation phenomena that occur on nanometer length scales and femto-second time scales, but evolve to be critical elements in bulk behavior and performance over decades spans over 12 magnitudes of order in length and 24 orders of magnitude in time as shown in figure 1.2. Linking degradation mechanisms to material performance is the essence of L&DS. Implementing L&DS for PV Technologies is identified as a critical scientific challenge for robust
adoption of PV. The 25-year lifetime performance of PV requires a better understanding of the degradation mechanisms in PV materials, components and systems. Typically failure of PV components is attributed to biological attack, material incompatibility, and sustained or periodic stresses. While most materials for PV are considered biologically inert, material compatibility must be carefully considered when designing systems for long lifetimes. Small differences in the thermal expansion coefficient for example, can lead to periodic stress through the system's lifetime. Weathering leads to degradation when in-use conditions include atmospheric gases, pollutants, diurnal and annual thermal cycles, exposure to ultraviolet (UV) radiation and humidity. Rain, hail and wind loads can induce cyclic mechanical stress on a system of varying stress profiles.

In selecting materials and components for light harvesting, stability of the bulk substrate is of paramount importance. Interfacial interactions can also be an important consideration, as the free energy of atoms at interfaces is typically higher than those in bulk materials, increasing reaction rates for thermodynamically drive reactions. While a significant effort is underway to determine the appropriate protocols for SLP of PV systems, a solar mirror is a somewhat simpler system and therefore presents fewer challenges. A test protocol for determining bulk acrylic stability for 25 year lifetimes was devised. Three acrylic formulations were evaluated, and one was selected for the solar grade mirror. With that information, metallization composition and protective coatings were evaluated for their performance in a series of accelerated weathering protocols exposing the objects to high intensity UV, heat, humidity and corrosive environments.
stress-response (R(S)) framework for comparing dissimilar materials’ response to variant exposure conditions was developed to link performance through time to environmental stress 12–14.

By developing metrics, metrology, and tools to quantify, compare, and cross-correlate the response of PV systems, components and materials to a variety of stressors such as UV radiation, humidity and temperature variation for both accelerated and outdoor testing, we can link stresses to observed responses in a R(S) framework and determine quantitative rates of degradation 15,16. L&DS requires the development of quantitative degradation mechanisms and rates for degradation and failure modes, so as to enable quantitative lifetime projections. Lifetime and degradation science (L&DS), based on a stress and response R(S) framework, is being developed that links the intensity and net stress to which materials, components and systems are exposed to the responses observed and the degradation and damage accumulation over lifetime. This R(S) framework can encompass single factor, multifactor and cyclic environmental stressors including solar irradiance, temperature and humidity, which can cause degradation over time. The R(S) framework allows for the cross-correlation of the materials response and degradation rates with numerous applied stress intensities and net stresses and allows determination of the effects of stressors, even at accelerated intensities, with applicability to multiple service conditions.

At the same time, an R(S) framework permits incorporation of data determined over many stress conditions and corresponding to many different responses and its cross-correlation to produce a more complete picture of a material’s or system’s stress and response when compared to the traditional acceleration factors approach, which is always in reference to one set of service or application conditions. The R(S) framework
Introduction

requires the use of quantitative metrics to be used such as induced absorbance to dose (IAD) which is defined as the rate of photodarkening or photobleaching of a material as a function of radiation dose. In the R(S) framework, all of the available responses of a material are integrated to provide a comprehensive scientific understanding of the material.

1.3 Thesis Overview

As a case study for L&DS and the R(S) framework, optimization and characterization of Poly (methyl-methacrylate) acrylic back surface mirrors was performed. Multiple accelerated weathering protocols were used to inform choice of substrate composition, metallization and coating systems.

1.3.1 Literature Review

A summary of technology trends, previous work on solar mirrors and the solar mirror marketplace helps to show the position of Replex Plastics and the benefits of their technology platform. A brief overview of the L&DS technology development pathway shows where information in this report fits into the product development.

A literature review of the degradation mechanisms of acrylic PMMA used for solar grade products was used to determine appropriate testing procedures, and inform substrate selection. Comparisons between other weathering protocols, radiation exposures show the need for product testing and validation.

A review of damage evolution and typical metrics for discussing mirror performance was used to determine appropriate testing procedures, and inform substrate selection. Weathering protocols and lifetime performance prediction for current industrial solar
mirrors show the need for such a weathering program to ensure good lifetime performance.

1.3.2 Substrate Optimization

Details of experimental exposures and metrics developed to enable the cross-correlation of weathering protocols to real world conditions. Three exposures of three grades of acrylic are performed and results of experiments presented.

1.3.3 Mirror Performance Optimization

Methods and metrics for discussing mirror performance are compared. Details of three experimental exposure protocols and results are presented.

1.3.4 Discussion

Comparisons between rates and degrees of weathering are discussed. Results are interpreted in the context of solar mirror performance.

1.4 Personal Scientific Contributions

Personal contributions to this body of work were extensive. Initially, acquisition, setup, and maintenance of exposure and evaluation equipment was a priority. Design of experiment, determination of safe operation conditions and sample acquisition were important and extensive. Continual monitoring of exposures paired with analysis of results led to publication of this report. Most of the spectroscopy and data analysis was performed personally with the help of Dr. Bruckman. Student researchers were overseen as they performed scattering analysis and other tasks. While the lion’s share of exposure were
performed at Case Western Reserve University, some were performed at Replex’s head-
quarters in Mt. Vernon, Ohio. Presentations, publications and posters discussing this
work at various conferences can be found in Appendix 2.
2 Literature Review

2.1 Solar Mirrors

Solar mirrors have taken a prominent role in harnessing solar energy. Concentration photovoltaic (CPV) designs using cassegrain optical concentrators shown in figure 2.3 and parabolic dish concentrators in figure 2.5 are among candidate applications to reduce the delivered cost of PV power. Concentrating solar power (CSP) utilizes either flat mirrors mounted on heliostats, linear fresnel reflectors, or reflectors arranged in a parabolic dish or parabolic trough mirrors to focus the sun's energy on a power conversion unit, usually heating a working fluid for a turbine, but sometimes on the hot end of a stirling engine. Solar mirrors may also find use in day-lighting applications, accelerated outdoor weathering testers and other building integrated products that require long lifetimes.

Mirrors for power generation have any number of different characteristics, determined by their end use application. All applications rely on high hemispherical reflectivity. For some CSP applications, long throw lengths are required for optimal performance making specular reflectance of paramount concern. For optical elements with shorter throw lengths, like low concentration PV (LCPV) and mirror augmented PV (MAPV) applications some low angle scattering is acceptable. Spectrally filtering
mirrors offer some additional benefits when mirror augmentation is implemented on systems that typically operate without mirror augmentation. For flat-plate PV devices, exceeding nominal 1-sun exposure can induce degradation associated with higher operating temperatures and UV induced degradation\(^{22}\). Since typical flat-plate PV devices do not efficiently convert UV and IR light to electricity, spectrally filtering mirrors can be used to enhance device performance without inducing or limiting additional degradation.

Industrial solar mirrors, such as ones made by FLABEG are second surface glass mirrors. Thickness varies with application from 1-5mm. Silver metallization allows for up to 95% reflectance on thin substrates. Tempered glass has adequate radiation durability, but is a high cost material, which is fairly difficult to form. Additionally, the chemistry of silver thin films requires the deposition of a thin film of copper, which acts as a tie layer.
promoting adhesion of the silver metallization to the substrate\textsuperscript{23}. Silver also tarnishes easily in the presence of small amounts of sulfur, meaning that the protective coating used for silver mirrors must be hermetic and impervious to weathering stress\textsuperscript{24–26}.

Figure 2.2. Compound Parabolic Concentrator, Photo Kara Shell
Figure 2.3. Parabolic Dish Reflectors for CPV applications, Photo Laura Bruckman

Figure 2.4. Flat mirrors on heliostats for CSP power generation, Photo Wikimedia Commons
Replex Plastics Inc., a manufacturer of thermoformed acrylic and polycarbonate domes and mirrors for automotive and security applications leveraged and existing partnership with Dr. Roger French at Case Western Reserve University to move from their existing product to a solar-grade product. Because the technology under development is embedded in the company’s current product, the flexibility of the existing manufacturing line, and the ability to create the novel product without need for an expanded production line, development of this new product represents access to new markets without additional capital investment. Because solar mirrors are a component of many systems, but not consumer products on their own, solar mirrors represent a high-value addition to the Replex line of products.
While there are barriers to market entry and some well established competitors, low-cost, formable mirrors may enable new technology applications where existing products are not viable. The real challenge for market acceptance is convincing customers that the product is a viable candidate for 25 year-performance. The acrylic substrate used for most Replex products has a manufacturers warranty of 10 years, necessitating an exploration of choices for solar grade mirrors, meanwhile choice of back surface coating and metallization must be optimized for lifetime performance.

### 2.2 Technology Development

The typical technology development is a linear process depicted in 2.6. The first step of technology development is to target identity and concept development. After a detailed design is made and device specifications are set, prototype development begins. As prototype generation occurs, process design is needed to meet specifications on the larger scale before manufacturing trials begin. Photovoltaic technology elements to be sold into the market are required to pass qualification testing before they can be used in grid-tied applications.

Reliability engineering is thought to fall into three main statistical regimes: infant mortality, random failures, and failure at the end of life depicted in figure 2.7. Initial performance qualification testing, represented by UL 1703\textsuperscript{27}, IEC 62108, IEC 61215, and IEC 61646, allows for reasonable confidence intervals when mitigating infant mortality and can sometimes be useful in predicting random failures in PV modules, which are failures that occur imminently or randomly after deployment due to process and manufacturing defects. Lifetime performance, however, requires knowledge of the power
It is thought that by performing highly accelerated stress tests, degradation modes of materials can be elucidated and with confirmation from outdoor testing, lifetime performance can be predicted. Early insights into material stability, degradation phenomena
and mechanisms can save money, provide product confidence, and give rise to continuously improving product durability. Using an L&DS cyclic approach 2.8 a library of exposed materials, components and systems alongside high quality analytics can give prognostic insights into system durability providing the confidence that is needed for new technology adoption.

2.3 Substrate PMMA Acrylic Weathering

Polymers of many types are vulnerable to UV degradation because the highest energy UV radiation observed in the solar spectrum has energies that surpass all but the strongest
carbon bonds. In order to protect these polymers, various formulations of UV absorbers, hindered amine light stabilizers (HALs), and radical scavengers have been employed to stabilize materials exposed to UV degradation. A common class of UV absorbers are hydroxyphenyl benzotriazole compounds such as Tinuvin™ made by BASF, which are usually used at concentrations between 0.1 and 1 wt. % and protect the polymer matrix by being sacrificially degraded by the UV light. The degradation of numerous grades and formulations of PMMA, acrylic polymers, has been studied under a wide variety of UV light sources, including mercury vapor lights emitting at 253.7 nm.
nm\textsuperscript{32–36}, xenon arc lamps filtered to match AM 1.5\textsuperscript{37}, and \textsuperscript{60}Co emitted gamma radiation\textsuperscript{32}. Additionally, studies of the wavelength dependence of the photodegradation have been performed using glass filters\textsuperscript{37}, monochromators\textsuperscript{38,39}, and with different light sources emitting at 300 nm and 350 nm\textsuperscript{33}.

Indoor exposures have been performed under vacuum and in air, with mass loss metrology and gaseous degradation products being identified by mass spectroscopy\textsuperscript{33,35,36}. Production of degradation products and viscosity averaged molecular weight distributions have been used to determine the quantum efficiency of UV light in degrading PMMA\textsuperscript{32,35,36,39,40}. Others have performed outdoor test with flat plate exposures and with accelerated tracking exposures\textsuperscript{37,38,41,42}. Optical characterization of degradation in acrylics has been done using colorimetry\textsuperscript{37,42}, optical density\textsuperscript{39,40}, UV spectroscopy\textsuperscript{35–37,39,40,43}, electron spin resonance spectroscopy\textsuperscript{32} and Fourier transform infrared spectroscopy\textsuperscript{38,41,43–45}. Surface degradation of acrylics has been studied using atomic force microscopy\textsuperscript{43}, scanning electron microscopy\textsuperscript{41} and contact angle measurements\textsuperscript{43}. Mechanical testing has been used to characterize the effects of UV degradation on PMMA mechanical properties\textsuperscript{42,46}. Other studies have been done to analyze the thermal decomposition and depolymerization of PMMA associated with mass loss and increased photodegradation\textsuperscript{47}.

Degradation of PMMA by ultraviolet light is thought to occur when high energy incident radiation initiates radicalization of an ester side group leading to $\beta$-scission of the polymer backbone and decomposition of the radical end to products including H\textsubscript{2}, CO, CO\textsubscript{2}, CH\textsubscript{3}OH and HCOOCH\textsubscript{3}\textsuperscript{36}. The loss of the ester group creates a greater free volume in the PMMA, increasing likelihood of creep and dimensional instability\textsuperscript{48}. Chain scission that occurs from exposure to solar radiation is predominantly caused by UV radiation with wavelengths shorter than 320 nm\textsuperscript{38,39}. This process is accompanied by an
increase in yellowing in many experiments. The color centers responsible are identified as free radicals trapped with the bulk material until they become neutralized by oxygen or other radiation.

Typically, UV exposure of materials can be performed with a xenon arc light source, which can be filtered to closely match the shape of the solar spectrum for air mass (AM) 1.5, a standard representing real-world exposure at 37° latitude. AM 1.5 exposures can be performed at multiple levels of irradiance by using concentrating optics to focus the light beam. This enables proper weathering acceleration because spectral matching between outdoor conditions and indoor exposures excludes degradation induced by radiation outside of typical environmental conditions. However, due to their lower operating costs, fluorescent exposure of materials using UVB-313, UVA-340, or UVA-351 lamps are often used as a less expensive way to provide insights into material performance over time. UVA-340 lamps radiate light that closely matches the AM 1.5 spectra in the region from 280 to 360 nm when the spectra radiance peaks at 340 nm with a value of 0.30 W/m²/nm.

2.4 Mirror Weathering

Metrics for comparing reflector performance are poorly developed. Manufacturers typically quote a minimum percent reflectance value. Because reflectance is wavelength dependent, details of the wavelength selected for analysis can be obfuscated by manufactures. For solar mirrors, "solar weighted reflectance" is a common metric used to compare mirrors of similar construction. This metric integrates reflectance spectra with the solar spectra AM1.5. This quantity represents the amount of incident solar radiation which is reflected from a surface and its typically quoted as a single number in
terms of percent. Because of the various applications for solar-grade mirrors, this quantity also hides some important information. The solar spectral distribution shows radiation from the UV (280 nm) to the infrared 2500 nm. However, not all wavelengths are of equally useful. Useful wavelengths are those whose energy can be converted into work. Therefore, while solar weighted reflectance can be useful for CSP mirrors, where light is reflected and its energy is transferred to a working fluid and nearly all of the light is collected, it is not a good metric for photovoltaic electric generation, because electron-hole pair generation has a threshold energy below which absorbed photons will not generate useful power. Since thresholds and conversion efficiency vary by absorber type, there are no current useful metrics for comparing mirrors intended for use with photovoltaics.

Of additional concern is the definition and differentiation of specular and diffuse reflectance. Specular reflectance is defined as the amount of reflected light that is directly transferred to a target receiver surface. If well collimated light is incident on the reflector, an acceptable cone-angle (in mrad) around the pointing vector is defined to be the specular cone. Light that is reflected, but scattered outside this cone is deemed diffuse reflectance. This can be due to microroughness of the mirror surface and hazing of the substrate or topcoat. For optical elements with long throw-lengths, like heliostat mirrors for CSP, high quality mirrors with a specular cone of 2-4 mrad are used. For PV applications throw lengths are usually shorter (typically ranging from .1-1m) and as such a larger acceptable cone can be defined.

Loss in specular reflectance attributed to weathering has generally been found to be proportional to loss in total hemispherical reflectance. Corrosion induced loss in total hemispherical reflectance of the reflective layer typically occurs before specularity degrades. Because total hemispherical reflectance is relatively easier to measure compared
to specular reflectance and because it is the predominant contributor to loss in specular reflectance during weathering, it is the performance parameter that is routinely used.

The 2-plane reflectance 2.9 diagrams of aluminum and silver show the obvious performance gains allowed by silver metallization. Lower interfacial absorptance and higher overall reflectance are obvious benefits of silver metalization. Being the most conductive metal, a silvered reflector with a thickness of 60nm or greater can reach 98% reflectivity. While less aluminum is needed for good reflectance, 60nm reaches full opacity, the maximum reflectance is near 90% at 550nm.

While silver surfaces are fairly resilient to oxygen attack, silver is prone to tarnish in the presence of even minute concentrations of sulfur. Silver sulfide and similar compounds increase absorptance of silvered surfaces. While oxygen attack of aluminum mirror surfaces is common, since the oxidation product (Al$_2$O$_3$) does not contribute to absorptance some oxidization is acceptable.
Adhesion failure can severely limit performance of solar mirrors. Bonding between silver and PMMA is weak, particularly in the presence of moisture. Optically transparent, inorganic interlayers can improve the adhesion between PMMA and silver. Strongly acidic oxide layers like SiO$_2$, (and perhaps Cr$_2$O$_3$) may be expected to bond well to basic PMMA. Interlayers deposited between the silver and PMMA to improve adhesion have had some success.

2.5 Damage Evolution in Solar Mirrors

2.5.1 Metallization roughening

A thin alumina passivation layer, 4-5nm thick, forms on the deposited aluminum surface before coating and protects the aluminum film from oxidization. However, because atoms at interfaces have higher Gibbs free energy, thermal energy can drive reactions and vibrational stress can cause rearrangement of atoms. Moisture and electrolytic solutions can further speed oxidization by enabling charge transfer. A back surface mirror reflector can only be as smooth as the substrate on which it is deposited. Acrylic sheet is made to ASTM D4802 which prescribes the allowable roughness of standard sheet. Significant deviations from this can occur when an object is thermformed, but acrylic surface roughness is still typically much smaller than metallization thickness, and hence reflectance is dominated by mirror finish quality. Surface roughening can lead to geometric absorptance–self-shadowing– or scatter if RMS roughness is large compared to wavelength as depicted in figure 2.10. Metallization roughening in mirrors has been studied in the presence of corrosive environments, when subject to
high intensity laser exposure\textsuperscript{\ref{57}, VUV\textsuperscript{\ref{58}} and EUV\textsuperscript{\ref{59}}} exposure, under particle bombardment\textsuperscript{\ref{60}} and when fielded\textsuperscript{\ref{61}}. The effects of surface contamination have been investigated\textsuperscript{\ref{62,63}}. Investigations of polymer reactivity and mirror performance have been performed\textsuperscript{\ref{64}} showing that interfacial energy is a significant driver in mirror degradation.

Accelerated lifetime testing for mirrors have been devised for CSP\textsuperscript{\ref{17,54}}, CPV\textsuperscript{\ref{65,66}} and EUV Lithography\textsuperscript{\ref{67,68}}.

Photon and contaminant induced silver corrosion seems to play a major role in the corrosion of silver films\textsuperscript{\ref{69}} but aluminized mirrors do not appear to have the same susceptibility to tarnish.

Development of first surface aluminum mirrors on glass substrates has been performed\textsuperscript{\ref{70}} with good durability results. Sol-Gel protection of first surface aluminum mirrors has been investigated\textsuperscript{\ref{71}} and appear to mitigate concerns over cleaning protocols for first surface aluminized mirrors.

If oxidization or corrosive attack occurs, pit formation, cavitation, crack formation and other surface defects can evolve leading to increased masking-shadowing. Differences in the refractive index of aluminum oxide and acrylic can cause parasitic absorption to increase, or cause an increase in scatter.

\section*{2.5.2 Protective coatings}

Two proprietary, lead-free formulations of protective coating developed by Replex for solar grade mirrors were used to coat otherwise identical samples. Typical coating compositions use a balance of pigments, solvents, additives and resin. TiO\textsubscript{2} and ZnO provide opacity and increase resistance to yellowing, mildew and UV degradation. Coating resins provide adhesion, durability and resistance properties. Appropriate resin choice
is imperative to durable coating systems, most coatings can last 25+ years in industrial applications before failure occurs and metallization corrosion is dramatically increased\textsuperscript{72}.

Replex mirror coatings were in one instance a point of failure for transportation mirrors. This was determined to be due to a formulation change of fleet-vehicle washing solutions. The new solution was very aggressive against the coating leading to deadhesion.
and loss of reflectance. While new coatings have improved resistance to such chemical attack, such a case obviates the necessity for careful consideration of the washing procedures for any solar-grade mirror.
3 Experimental Substrate (PMMA Acrylic) Weathering

3.1 Specimen Selection and Characterization

Three formulations of PMMA used in the present study are multipurpose (MP) and UV transparent (UVT) and UV absorbing (UVA) acrylic samples. Each formulation contains different amounts of the same UV stabilizer. These acrylic samples were provided by Replex Plastics. MP grade acrylic is used in the security and transportation industry as a substrate for mirrors and housings and holds a warranty that it will not lose more than 3% of its light-transmitting ability in 10 years. UVT acrylic is used for daylighting applications that require full spectrum light, but is not highly radiation durable. UVA acrylic was expected to be the best candidate for solar-grade mirrors, because it contains a significant amount of stabilizer and is nearly (99%) UV opaque at thicknesses greater than $\frac{1}{2}$ mm. UVA acrylic may be a useful product for LCPV applications, as it absorbs most of the UV light responsible for photodegradation of PV encapsulants at the cell level. The thickness of all the acrylic samples was approximately 3 mm.
3.2 Simulated Solar Exposures

Exposures were performed with a Q-Labs QUV Accelerated Weathering Tester (Model QUV/Spray with Solar Eye Irradiance Control) and with a Newport 1.6 kW diverging beam solar simulator (Model 92190) with a 13X irradiance concentrator (Model SP81030-DIV). By sequential measurements of optical properties after different steps of exposure dose (or exposure doses), it is possible to determine the relationship between solar irradiance (the stress) and induced degradation rates of optical properties (the response) of these materials, along with the form of the dose dependence of the response (linear, exponential, sub- or supra-linear). The 18 day of concentrated Xenon-Arc exposure is equivalent to radiant exposure of 8.6 years of tracked exposure in Phoenix, Arizona. Additional exposure of UVA samples beyond the 18 day exposures were performed, as degradation was not apparent after the 18 day exposure.

The QUV UVA-340 exposures had eight of each type of acrylic samples so as to provide sufficient statistics. A representative sample was removed after each dose step and the baseline sample was never exposed. The first dose step was 284 hours and three subsequent dose steps were 220 hours. After 948 hours of exposure exposure steps were extended to 1000 hours each. There was only one sample for each type of acrylic exposed in the Newport solar simulator at 50.4 kW/m². Each dose step was 72 hours. The
baseline abs/cm and YI measurements were performed on the acrylic samples prior to exposures.

### 3.2.1 QUV Accelerated Weathering Tester

The QUV accelerated weathering tester uses customized fluorescent lamps to expose samples to high doses of UV radiation which simulates different types of damaging environmental stress conditions. The exposures were performed with UVA-340 lamps, which emit radiation between 280 and 400 nm. This closely matches the AM 1.5 spectrum at wavelengths shorter than 360 nm, where much of the damaging radiation exists in the solar spectrum.

The QUV was run using ASTM G154 Cycle 4 (1.55 W/m$^2$ nm$^{-1}$ at 340 nm at 70$^\circ$C) without the condensation step for 21 days. By setting the QUV peak intensity to 1.55 W/m$^2$ nm$^{-1}$ at 340 nm, a higher level of damage can be induced, making this a single-factor accelerated test compared to outdoor weathering. This level of UV radiation is 5.2 times higher than the intensity of AM 1.5D at 340 nm. While the integration of the QUV spectra gives a much lower total dose, the ratio of light in the damaging range from 280-360nm is much higher. Only 1.92% of AM 1.5 radiation falls into the UVA-340 exposure.

By correcting the TMY3 data for this factor the 1944 hour exposure is equivalent to 2.3 years of tracked outdoor exposure in Arizona. This was calculated with Equations 3.2 and 3.1 where $E_e$ is spectral irradiance for each source (W/m$^2$ nm$^{-1}$), $\lambda$ is wavelength in nm, $E_e$ is irradiance (W/m$^2$) and $H_e$ is radiant exposure to wavelengths between 280 nm and 360 nm (J/m$^2$).

$$E_e = \int_{280}^{360} E_e \lambda d\lambda$$

(3.1)
Figure 3.2. AM 1.5D spectra with UVA-340 at 0.32 W/m² nm⁻¹ (left), AM 1.5D with with UVA-340 at 1.55 W/m² nm⁻¹ (center) and AM 1.5D at 48.4 kW/m² and with UVA-340 at 1.55 W/m² nm⁻¹

\[ H_e = \int E_e d t = \int_0^t \int_{280}^{360} E_e \lambda d \lambda \] (3.2)

3.2.2 Diverging Newport Solar Simulator

The Newport xenon arc solar simulator was used to irradiate material samples with spectral matched AM 1.5 radiation using a Newport 13X high flux concentrator accessory with the solar simulator, irradiance levels of 50.4 kW/m² were achieved, making this a single-factor accelerated test compared to both UVA-340 and outdoor exposure. Continuous power monitoring with a Newport power meter (Model 1918-R) was used to monitor the irradiance making it is possible to quantify the irradiance and dose to which a sample was exposed. Integrated dose was calculated for total dose and UVA-340 (Equations 3.2 and 3.1). Compared with outdoor exposures in Arizona, using typical metrological year data (TMY3) supplied by NREL are about 9.1 GJ/m² per year, this 18 day test exposed samples to doses equivalent to approximately 8.5 years.
3.3 Evaluations for Stress & Response Framework

3.3.1 Cary 6000i with DRA-1800

An Agilent Cary 6000i spectrometer with a diffuse reflectance accessory-1800 (DRA) and an InGaAs detector was used to determine the Abs/cm for each acrylic sample. The spectra were acquired from 180-1800 nm every 0.40 nm with a scan rate of 48.00 nm/min and a spectral bandwidth of 2.00 nm in the UV-Vis and 4.00 nm in the NIR regions.

3.3.2 Hunterlab UltraScan Pro

The HunterLab UltraScan Pro was used to determine the YI of each of the acrylic samples. This instrument is a fast, high-performance color measurement spectrophotometer with a spectral range from 350-1050 nm with a 5 nm optical resolution.

3.3.3 Temperature characterization

The temperature was determined for the samples in the Newport solar simulator and the QUV accelerated weathering tester were determined with a Flir infrared camera (Model T300).

3.4 Response Metrics For Bulk Samples

Induced absorbance to dose (IAD) is a measurement of the change, at a particular wavelength, of the bulk optical absorbance per centimeter of a material and this analysis assumes uniform bulk absorption phenomena which may not be observed in certain samples \(^{73–75}\). Therefore IAD analysis is assumed to be independent of thickness. In these samples, preliminary evidence shows that photodegradation progresses from the light exposed surface through the bulk of the material. Therefore the IAD metric as used here
represents the average of the optical absorbance per centimeter through the thickness of the sample. IAD is a quantitative dose metric for photodarkening and photobleaching on a per unit dose basis. The per unit dose basis can be changed, so that IAD can be calculated per full spectrum dose or per UVA-340 dose. Incremental IAD, equation 3.3, is useful to track and identify transient phenomena, which tend to stop after a certain dose (e.g. phenomena associated with curing or photobleaching of impurities). While average IAD, equation 3.4, is useful to follow over large doses and long exposure times, since the total dose exposed increases, the contribution due to transient phenomena are reduced in amplitude and continuous degradation processes end up being the predominant observed process in the average IAD. If either calculation of IAD reports the same values for successive dose steps, then the material is behaving linearly, and if the same values of IAD are seen with two different exposures then reciprocity in irradiance is being obeyed.

\[
AverageIAD = \frac{Abs_f(\lambda)/cm - Abs_i(\lambda)/cm}{Dose_{total}}
\]  
(3.3)

\[
AverageIAD = \frac{Abs_{i+1}(\lambda)/cm - Abs_i(\lambda)/cm}{Dose_{i+1} - Dose_i}
\]  
(3.4)

IAD values were determined with the Agilent Cary 6000i with a DRA-1800.

### 3.4.1 Yellowness Index (YI)

Yellowness index (YI) as defined in the ASTM E313 is a colorimetric measure of yellowing. Because YI is measured over a broad wavelength range it is more sensitive than typical spectral measurements. YI is calculated from the transmittance values for 380-780 nm. Yellowness indices are also useful because they are closely linked to reduced optical
performance. YI can be determined by equations 3.5 with the HunterLab Colorimeter or with equation 3.6 with the Cary 600i:

\[
YIE_{313} = \frac{100(C_X X - C_Z Z)}{Y} \tag{3.5}
\]

where X, Y and Z are the CIE tristimulus values. For D65/10, Cx=1.3013, Cz=1.149851 or

\[
YIE_{313} = \frac{T_{620} - T_{420}}{T_{500}} \tag{3.6}
\]

where \(T_\lambda\) is the transmission at a given wavelength. Yellowing rates, the rate of change in yellowness as a function of dose, were determined as per equation 3.7.

\[
dYI = \frac{YI_{\text{final}} - YI_{\text{initial}}}{Dose_{\text{total}}} \tag{3.7}
\]

### 3.4.2 Metrics for Stress Characterization

In our quantification of exposure stresses, UVA-340 dose was chosen as an irradiance dose metric instead of the traditional TUV dose. UVA-340 dose is a useful metric because UVA-340 lamps have peak intensity at 340 nm and their spectral irradiance drops off quickly at wavelengths longer than 360 nm. UVA-340 dose includes the most damaging wavelengths of light that cause degradation of the PMMA samples. The wavelengths between 360-400 nm that are included in the TUV metric have not been shown to cause significant degradation and the spectral match to the light source is not close. Using
Table 3.1. UV Stressor Comparison

<table>
<thead>
<tr>
<th>Stressor</th>
<th>Full Spectrum Dose</th>
<th>TUV dose</th>
<th>UVA-340 Dose</th>
</tr>
</thead>
<tbody>
<tr>
<td>Newport Simulator</td>
<td>48.4kW/m²</td>
<td>1662W/m²</td>
<td>618W/m²</td>
</tr>
<tr>
<td>Hot QUV</td>
<td>84.5W/m²</td>
<td>84.5 W/m²</td>
<td>60.7W/m²</td>
</tr>
</tbody>
</table>

UVA-340 dose for calculating response rates is expected to show better correlation results of QUV accelerated weathering tester and Newport solar simulator exposures because TUV dose would inherently underestimate the amount of damaging short-wave UV light present in QUV exposure with UVA-340 bulbs shown in table 3.1.

### 3.4.3 Comparison of Stressors

While photodegradation mechanisms are many, complex and can be ill-defined, photodarkening, photobleaching and yellowing rates can be discussed without the complexities of explicit mechanistic insights. Higher amounts of total degradation were observed in all samples exposed in the Newport solar simulator, shown by the larger increases in optical absorbance for all samples. This was expected because of the fact that these samples saw much higher net stresses. However, when comparing responses of samples on a degradation per dose basis, shown with IAD values, QUV exposure showed a higher rate of degradation. The QUV accelerated weathering tester is an approximately 5X UV irradiance stress at 1.55 W/m²/nm at 340 nm of UVA-340, while the Newport solar simulator is approximately 50X full spectrum irradiance stress.

The IAD curves for dose steps, which are comparable in terms of total UVA-340 dose between the two exposures (figure 3.10), show that exposures in the QUV protocol has a larger IAD value in each region than an equivalent dose from the Newport solar simulator. This may be due to the effect of the elevated chamber temperature as summarized in Table 3.2. In the UVT acrylic, three times more photodarkening occurred near
Figure 3.3. The conditions used for the accelerated UV exposure were of different stress intensities and net stress values.

<table>
<thead>
<tr>
<th>Exposure</th>
<th>Chamber Temperature</th>
<th>Sample Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>Newport Solar Simulator</td>
<td>Uncontrolled</td>
<td>50°C</td>
</tr>
<tr>
<td>QUV</td>
<td>70°C</td>
<td>67°C</td>
</tr>
</tbody>
</table>

Table 3.2. Temperature Characterization for UV Exposure

the fundamental absorption edge and six times more photodarkening occurred from 350-500 nm. In the MP acrylic, equivalent photodarkening occurred near the fundamental absorption edge and three times more photodarkening occurred from 375-450 nm when comparing the two stressors. However, the stabilizer package shows a much higher level of photobleaching in the QUV exposure protocol, which supports the assumption that the QUV protocol induces more degradation, and that this effect may be due to the higher sample temperature. Sample temperatures are given in table 3.2.

3.5 Comparison of MP and UVT PMMA grades

IAD values for each of the two PMMA grades for each exposure show that the IAD values were higher for UVT acrylic compared to MP acrylic in both stress conditions as seen
in Table 3.3. MP shows features in all three regions, while UVT does not have distinct spectral features in region 2. In all MP and UVT samples exposed, there is significant photodarkening occurring in region 1. In QUV exposure of MP samples, there is some initial photobleaching in regions 2 and 2’; however, subsequent dose steps show photodarkening of the base polymer lowering this photobleaching and eventually leading to photodarkening in regions 2 and 2’. Region 3 is characterized by broad band increase in optical absorbance. This response is decreased by a factor of nearly 10 with the addition of the stabilizer package in MP grade acrylic.

In addition to the effects of temperature, another factor which may account for the increased rate of photodegradation in the UVT sample in the QUV is that the sample did not screen any UV light which passed through the sample. Because samples in the QUV were mounted on aluminum trays, degrading UV light may have reflected, passing through the sample a second time, while in MP samples the UV light was absorbed by the UV stabilizer package, and only lower energy light was reflected. This may account for the differences in IAD rates at the fundamental absorption edge.

The response ratios between the grades show that more degradation occurred in UVT in region 1 and 3 for both stress exposures. This relates to the fact that the UVT acrylic is less stabilized then the MP acrylic. The YI rate was also much higher in UVT than MP in the QUV exposure. Region 2 and 2’ could not be compared because there are not significant peaks in UVT related to the stabilizer package.
3.6 UVA weathering results

After 227GJ/m² of dose in the Xenon-Arc exposure, degradation has been observed in UVA acrylic. IAD values produced show what may be a small amount of stabilizer bleaching as a reduction in optical density near the absorber package edge. Confirmation of these results with longer exposures to evolve more damage have shown UVA to be highly radiation durable. While bleaching phenomena is undetectable, after 3.8GJ/m² UVA-340 dose yellowness has increased, shown in the IAD of UVA acrylic.

3.7 Graphical Representation of Results

The absorbance per centimeter, base 10 results of two different types of exposures on two formulations of acrylic PMMA, UVT and MP, are reported in figures 3.4 and 3.5, respectively. Baseline spectra of both formulations of acrylic show a fundamental absorption edge near 275 nm (region 1); however, MP grade acrylic shows two strong absorption peaks centered near 298 nm (region 2) and 339 nm (region 2') due to the absorber package, while both formulations are transparent to wavelengths longer than 380-780 nm (region 3), but IAD are calculated at 440 nm for region 3.

3.8 Summary of Bulk Material Comparisons

3.8.1 IAD Comparisons

The IAD values for the two exposures and PMMA grades of acrylic are summarized in table 3.3 for regions 1, 2, 2', and 3 at equivalent dose steps. UVT acrylic exposures in QUV and Newport solar simulator show the UV degradation of unprotected PMMA acrylic (Figure 3.7). Because this material does not contain a significant UV absorber package,
UVA-340 dose is highly effective at degrading this polymer. Photodarkening, is shown as a positive IAD curve and is observed at all wavelengths, but increases near the fundamental absorption edge.

MP acrylic exposures in QUV and Newport solar simulator show the UV degradation of UV formulated PMMA acrylic (Figure 3.5). This material which contains a significant
UV absorber package is highly protected from UV degradation. Photodarkening is again observed at the fundamental absorption edge, shown as a positive IAD values; however,
Figure 3.8. Spectral IAD of UVT Acrylic exposed to QUV weathering (left) and Newport Solar Simulator Exposure (Right)

Figure 3.9. YI values at each exposure step show near linear response with dose

both studies show some initial photobleaching of UV stabilizers and slight photodarkening in the visible region.
Figure 3.10. IAD comparisons at near equivalent dose steps in two exposures for UVT (Left) and MP (Right)

Figure 3.11. IAD of UVA Acrylic after 3.8 GJ/m² UVA-340 dose, equivalent to approximately 40 years of outdoor exposure
3.8.2 Yellowness Indices

Yellowness indices are reported for samples exposed to UVA-340 radiation (Figure 3.8). The yellowing rates for UVT and MP are 0.041 and 0.0035 1/(MJ/m²) of UVA-340, respectively. Yellowing rates for Newport solar simulator became non-linear after 2 dose steps, but yellowing rates in the linear region for UVT and MP are .0068 and .0014 1/(MJ/m²) respectively.

3.8.3 Yellowing

Changes in yellowness were observed in both MP and UVT acrylic formulations and appear to behave in a near-linear fashion with dose (Figure 3.8). Changes in yellowness of these materials show the power of YI as a response because it is very sensitive to small changes in the optical performance of these materials after exposure to these stresses. No yellowing was observed in UVA acrylic, indicating that the presence of such a high concentration of UV absorber has turned off or reduced this degradation mode effectively. Hazing of the samples was not observed. Miller et. al. suggested that yellowness and haze are two degradation mechanisms that occur in different classes of PMMA acrylic.

The yellowness that occurred in the samples in the QUV exposure occurred throughout the entire sample after the first dose of exposure. The samples in the Newport solar simulator showed the yellowness occurring predominantly on the first surface closest to the irradiation after the first dose. The yellowing proceeded from the first surface to the interior of the 3 mm sample after each subsequent dose.

This result agrees with the findings that yellowness occurs at the top surface and can be reduced with polishing. The increased depth penetration of yellowing products
in samples exposed in the QUV may be due to the fact that samples were backed with standard aluminum sample holders, which may have contributed to degradation of the samples. This allowed some light to be reflected from the aluminum holders through the samples, though the extent and wavelength dependence of this contribution has not been thoroughly investigated. Another possibility for the increase in yellowing in the QUV UVA-340 irradiance samples is the higher temperature of the stress conditions. It has been suggested that there is a relationship between UV radiation and temperature when discussing yellowing and that an increase in temperature can increase the yellowing rate. The samples in the Newport solar simulator were placed on a fused ultra-high purity silica photomask blank; therefore, there was very little reflection of light back through the sample and the temperature of these exposures was less than that of the QUV exposures. The equivalence of UV exposure of the two difference sources also be affected by filter and burner aging in the xenon arc lamp.

3.8.4 Stabilizer

While neither of these materials is guaranteed to have a 25-year lifetime in use at 1 sun irradiance for PV applications, the range of their response to stressors can inform material selection criteria for PV applications. UVT acrylic, lacking UV absorbers, can provide insights about the degradation of the base PMMA resin, while MP acrylic shows the effect that a moderate stabilization package can provide. The yellowing rates of these two materials show this sharp contrast, and MP acrylic shows a 10-fold reduction in yellowing rate. Similar observations can be made about response near the fundamental edge, where MP acrylic shows an approximate 10-fold reduction in photodarkening near the fundamental absorption edge.
Recent exposures of UVA have shown the yellowing phenomena in region 3 after 3.8GJ/m² full spectrum dose. IAD ratios show a full 9x reduction when compared with MP acrylic. This highly-dosed sample confirms the conjecture that since UVA acrylic has 10x more UV stabilizer than the MP acrylic UVA acrylic could last for 10 times longer than MP acrylic. Additionally, because of the high absorbance of UVA the damage accumulation proceeds in a region much smaller than for MP acrylic, reducing its total effect on transmission.

3.8.5 Response Ratios and Acceleration

When comparing the ratio of the IADs between the two exposures of equivalent doses, the QUV exposure has a much higher acceleration rate of degradation compared to the Newport solar simulator exposure (table 3.3). The acceleration rate in the significant regions for each grade of PMAA is different, which can be caused by test conditions, composition of material additives, degradation mechanism as well as temperature. The observation that a constant acceleration factor was not observed in the different degradation regions (1, 2, 3) and mechanisms demonstrates that the traditional search for a single acceleration factor applicable to a particular material exposed under differing stresses or stress levels is probably an unreasonable expectation. This supports the need for developing a more versatile R(S) framework in which to encompass stressors, stress levels and multiple responses.
Table 3.3. Comparison of IAD values at region 1, 2, 2’ and 3 for grades of acrylic

<table>
<thead>
<tr>
<th>Exposures</th>
<th>PMMA Grade</th>
<th>Region 1</th>
<th>Region 2</th>
<th>Region 2’</th>
<th>Region 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>QUV</td>
<td>UVT</td>
<td>0.019</td>
<td>0.0069</td>
<td>0.0029</td>
<td>0.00070</td>
</tr>
<tr>
<td></td>
<td>MP</td>
<td>0.0052</td>
<td>na</td>
<td>-0.0042</td>
<td>-0.0018</td>
</tr>
<tr>
<td></td>
<td>UVA</td>
<td>na</td>
<td>na</td>
<td>na</td>
<td>7.05 x 10^-5</td>
</tr>
<tr>
<td>NewPort Solar Simulator</td>
<td>UVT</td>
<td>0.0069</td>
<td>0.0021</td>
<td>0.00062</td>
<td>1 x 10^-7</td>
</tr>
<tr>
<td></td>
<td>MP</td>
<td>0.0036</td>
<td>0.0045</td>
<td>-0.0033</td>
<td>0.00011</td>
</tr>
<tr>
<td></td>
<td>UVA</td>
<td>na</td>
<td>na</td>
<td>na</td>
<td>1.8 x 10^-5</td>
</tr>
</tbody>
</table>

3.9 Observations from Stress-Response Framework

Use of a the R(S) framework is an important way to study, understand and cross-correlate lifetime and degradation studies of materials, components or systems. By studying degradation responses due to multiple types of exposure conditions and steps, we are able to understand linear, sub- and supra-linear and nonlinear responses over a wide range of stress levels, types of stressors along with combination of stresses and time varying stresses. Also, by comparing or cross-correlating the effects of multiple stressors to the response of a single stress exposure, we can understand the specific (combined or sequential) synergistic mechanisms at play in a material system. Significant understanding of the ways a material may behave over its service life can be understood by comparing results of real time exposures to the R(S) framework of the system that has been developed beforehand. In cases where unwanted degradation is observed, the stress and response framework can be used to understand and guide mitigation of these unwanted responses. Additionally, because a stress and response framework can include highly accelerated stress conditions, without assuming that these conditions map linearly to in-service exposures, useful information about end of life performance can be gleaned.
4 Experimental Mirror Weathering

4.1 Specimen Selection & Characterization

4.1.1 Substrate Selection

Back surface mirrors were made with the same three formulations of PMMA used in substrate studies. Multipurpose (MP) and UV transparent (UVT) and UV absorbing (UVA) acrylics were used as the substrates for these mirrors.

4.1.2 Metallization & Coating

An exploratory exposure of a silver mirror metallization on acrylic was performed for several months, but the silverized mirrors proved undurable. The focus of this work is on aluminized mirrors. Acrylic sheet was metallized with aluminum and coated with two proprietary single layer unleaded coating systems. These mirrors provided by Replex Plastics are UV-Reflective (UVR) Multi-purpose (MP) and UV Absorbing (UVA) respectively. UVA mirrors are expected to be robust, due to their high stability to UV light. The two coating systems will simply be referred to as "Grey Coat" and "White Coat." Two proprietary, lead-free formulations of protective coating developed by Replex Plastics for solar grade mirrors were used to coat otherwise identical samples. Typical coating compositions use a balance of pigments, solvents, additives and resin. TiO$_2$ and ZnO provide opacity and increase resistance to yellowing, mildew and UV degradation.
Coating resins provide adhesion, durability and resistance properties. Appropriate resin choice is imperative to durable coating systems, most coatings can last 25+ years in industrial applications before failure occurs and metallization corrosion is dramatically increased.

### 4.2 Characterization of Aluminum Thickness

Metallization of formed acrylic components was performed with physical vapor deposition. The deposition process uses high-purity aluminum which is fired on a tungsten heating element and passively transferred to the preformed substrate. Coating uniformity is therefore a matter of proper element placement. According to figure 2.9 aluminum metallization of 60nm of greater is enough to have high reflectance, low absorbance and complete opacity.

A mirror reproducability study was performed to determine the uniformity of metallization reflectance, and the minimum thickness. Minimum thickness was established using a mass-balance approximation. Using the difference in the bulk aluminum before and after firing and the area covered by the metalization, approximate mean thicknesses were determined. Table 4.1 shows the results of this thickness approximation, and table 4.2 shows the results of the round-robin reflectance measurements for uniformity.

It is clear from these measurements that the metallization is beyond opaque and provides reproducible reflectivity results on their standard line. Metallization thickness

<table>
<thead>
<tr>
<th>Quantity Aluminum evaporated/cycle</th>
<th>335mg/cane</th>
</tr>
</thead>
<tbody>
<tr>
<td>Filaments/cycle</td>
<td>15</td>
</tr>
<tr>
<td>Total Area coated</td>
<td>156000 cm²</td>
</tr>
<tr>
<td>Average Thickness</td>
<td>120nm</td>
</tr>
</tbody>
</table>

Table 4.1. Aluminum thickness determination
Table 4.2. Reflectance round-robin results

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of samples</td>
<td>82</td>
</tr>
<tr>
<td>Average reflectance at 550nm</td>
<td>87.5 % ± 1.52%</td>
</tr>
<tr>
<td>Deviation over single sample (550nm)</td>
<td>0.86 %</td>
</tr>
<tr>
<td>Maximum reflectance (550nm)</td>
<td>91.0 %</td>
</tr>
</tbody>
</table>

of 120nm also allows for some corrosion to take place without reduced performance, which may mitigate a potential degradation pathway.

4.3 Chosen Accelerated Weathering Protocols

4.3.1 Hot QUV exposure

"Hot QUV" or UV-only exposure of back-surface acrylic mirrors was performed with a Q-Labs QUV accelerated weathering tester, using UVA340 fluorescent lamps, that closely match the high energy UV spectral range of the solar spectrum. This exposure was conducted in accordance with ASTM G154 Cycle 4 without the condensing humidity step. Test conditions were 1.55 W/m²·nm⁻¹ at 340 nm and chamber temperature was controlled at 70° C.

4.3.2 ASTM G154 with QUV

Exposure of back-surface acrylic mirrors with UV and dark humidity was also performed with a Q-Labs QUV accelerated weathering tester. This exposure was conducted in accordance with ASTM G154 Cycle 4 with the condensing humidity step. Maximum irradiance, at 340 nm, was set at 1.55 W/m²/nm and chamber temperature was controlled at 70° C, condensing humidity was obtained at 50° C.
4.3.3 Q-Fog Salt-Fog Exposure

Salt-fog corrosion exposures were performed in a Q-Labs cyclic corrosion cabinet. The salt-fog solution was 5% NaCl and the cabinet temperature was 35°C as prescribed in ASTM B117. Salt-fog corrosion according to ASTM B117 does not correlate to any real-world exposure other than submersion in tidal water, but as a highly accelerated stress test may be useful for screening candidate reflector compositions. Induced degradation in such a corrosive environment is expected to cause some surface roughness and possibly delamination of the metallization.

Another protocol that may correlate better to real weathering conditions is the Prohesion cycle, which consists of one hour spray at ambient temperature and one hour dry off at 35°C, using a solution of 0.40

4.4 Evaluations for Aluminum Mirrors

4.4.1 Metallization Roughening

A thin alumina passivation layer, 4-5 nm thick, forms on the deposited aluminum surface before coating and protects the aluminum film from oxidization. However, because atoms at interfaces have higher Gibbs free energy, thermal energy can drive reactions and vibrational stress can cause rearrangement of atoms. Moisture and electrolytic solutions can further speed oxidization by enabling charge transfer.

A back surface mirror reflector can only be as smooth as the substrate on which it is deposited. Acrylic sheet is made to ASTM D4802 which prescribes the allowable roughness of standard sheet. Significant deviations from this can occur when an object is thermoformed, but acrylic surface roughness is still much smaller than metallization thickness and hence specularity is dominated by surface quality. Surface roughening
can lead to geometric absorptance–self-shadowing– or scatter if RMS roughness is large compared to wavelength.

If oxidization or corrosive attack occurs, pit formation, cavitation, crack formation and other surface defects can evolve leading to increased masking-shadowing. Differences in the refractive index of aluminum oxide and acrylic can cause parasitic absorptance to increase or cause an increase in scatter.

4.4.2 Protective Coatings

Failure of some test specimens were noted in ASTM G154 exposure and salt-fog exposure. In all cases delamination or deterioration of the protective coating began at the edge of the sample. These samples exhibited a decrease in reflectance through the area of the sample, not only at edges. An increase in scattering was also observed. Application of a protective edge seal is expected to mitigate this effect.

4.4.3 Reflectance

Specular and diffuse reflectance are impacted by various material and surface properties. Front surface crazing, bulk haze formation and metallization roughening can scatter light. As such the bidirectional scatter distribution function (BSDF), illustrated in figure 4.1 for reflectors can be determined, giving the distribution of diffusely reflected light around the specular cone. For a given definition of specularity, integrated BSDF should be the same magnitude as diffuse reflectance. In order to characterized reflectance of mirrors and develop metrics for mirror performance, full spectrum reflectance measurements were made in hemispherical reflectance mode, direct only mode, at selected wavelengths of interest. Bidirection reflectance distribution analysis was performed, and BSDFs were parameterized with the Harvey Shack method.
4.4.4 Transmittance

Transmittance of light through the aluminum acrylic mirror is assumed to be zero. If degradation of the protective coating occurs and corrosion proceeds through the thickness of the metallization, this assumption may need to be addressed. Coating failure is nearly identical to component failure for these solar mirrors, and if the selected coating system proves unreliable.

4.4.5 Development of BSDF for Back Surface Mirrors

In order to parameterize BSDFs so that they can be studied through time and used to model real optical systems, the Harvey-Shack approximation, equation 4.1 is used:

\[ BSDF = \frac{A}{B + |\beta + \beta_0|^g} \]  

(4.1)

In Equation 4.1, the \( \beta \) vector represents the projection of the unit vector of the scattering direction onto the tangent plane and the \( \beta_0 \) vector represents the projection of the unit vector in the specular direction onto the tangent plane. A, B and g are fitting parameters that allow the model to accurately portray the scattering data. A determines the height of the fitting curve, B determines the inflection point where the curve transitions from flat to sloped and g determines the slope of the roll-off. If g=0 the model is equivalent to a Lambertian distribution. For polished surfaces, values of g generally range from 1.5 to 3.5 with 2 to 3 being the most common and the value of B is very small with a range of 1x10^{-6} to 1x10^{-10}.

Parameterization allows for correlation of weathering exposure to degradation and metallization roughness, and is a good first order comparison for different mirror grades.
Figure 4.1. Pictoral Depiction of BSDF (From Wikimedia Commons)
presented here. BSDFs can also be integrated into optical modelling software like Trace-Pro as a surface property. This kind of information can be useful for end users who can define custom shaped mirrors and add the surface properties to simulate the effect that scatter has on system performance.

Since BSDFs capture the total reflection of light from an object they can be useful for finding cases of crazing or cracking in the substrate metallization. Scatter from 3-D features that are large when compared with analysis wavelength can behave as a diffraction grating with roughness in a one-dimensional, single frequency profile as seen in figure 4.2. This phenomena was not observed in samples subjected to aging, but such observation in field aged mirrors can be quantified.

4.4.6 Absorptance

Parasitic absorptance is inferred from the three measured quantities. Absorptance at analysis wavelength is not impacted by acrylic yellowing in UVA mirrors because there is no acrylic absorption at 635 nm.

4.4.7 Optical analysis

Optical analysis of these mirrors was performed with an Aglient CARY 6000i spectrophotometer with a diffuse reflectance accessory (DRA-1800) which can give measurements of specular, diffuse-only (=/-4°) and total hemispherical reflectance. Spectral reflectance measurements are reported for representative samples.

4.4.8 Scattering Functions

In order to measure bidirectional scatter distribution functions, the ScatterScope 3D produced by ScatterMaster and distributed by Lambda Research was used. Development of a scattered light analysis capabilities for back surface mirrors was performed
with Lambda Research and engineers at Scatterworks. The ScatterMaster suite of software performed parametric fitting of the BSDF into the Harvey-Shack (ABg) model for
scattered light and Harvey-Shack approximations were performed in Lambda Research's TracePro BSDF converter utility.

### 4.4.9 Development of a Mirror YI Metric

Metallization degradation can be decoupled from substrate degradation using mirror yellowness index \( M_Y I \). This metric, modelled after the traditional YI uses reflectance values instead of transmission values for its calculation (equation 4.2)

\[
R_{YI} = \frac{R_{630} - R_{420}}{R_{500}}
\]  

(4.2)

Where \( R \) is reflectance at a wavelength \( \lambda \).

### 4.5 Comparison of Mirror Grades

As expected, substrated degradation proceeded very rapidly for UVR mirrors. \( M_Y I \) values show that substrate degradation occurs to the greatest extent in UVR mirrors and 7.8 times slower in MP. There was virtually no change in \( M_Y I \) for UVA mirrors. Reflectance loss comparisons at 420, 500, 620 and 635nm are shown in table 4.3.
Figure 4.3. UVR mirror with exposure to "hot QUV" and ASTM G154 with condensing humidity

Figure 4.4. Multipurpose Mirror exposed to "Hot QUV"
4.6 Graphical Representation of Results

4.7 Summary of Mirror Durability Comparisons

For UVR acrylic back surface mirrors, there was a total reflectance loss in the 250-400 nm region (Figure 4.3) and an increase of total reflectance near 1412 nm for each subsequent dose of exposure. The diffuse reflectance increased with each dose of exposure. The MP
acrylic back surface mirrors showed total reflectance losses in the 255-295 nm and 380-425 nm regions (Figures 4.4 and 4.7) and an increase of total reflectance near 1412 nm for each subsequent dose of exposure. The diffuse reflectance of the MP acrylic back surface mirrors increases with dose. UVA back surface acrylic mirrors do not show a significant difference in the total reflectance after dose in the QUV. The increase in the diffuse reflectance seen in back surface mirrors exposed in the QUV with humidity show surface degradation and increased scattering occurred due to exposure to UV light.
4.7.1 Scattering Results

The BSDF is analogous to a Fourier transform of the sample's surface profile - higher frequencies of surface roughness translate to higher scattering angles. A perfectly flat
surface will show a straight line at the expected reflecting angle for a flat mirror, in our case, zero degrees. As a surface gets rougher, this straight line widens out because more light is scattering to higher angles. The UVR mirror that was exposed to 1000 hours of salt fog scattered more light at higher angles compared to an unexposed sample. There was an increase in scattering from roughening of the acrylic polymer surface after exposure to the corrosive environment. There was only a slight change in the BSDF for samples after the QUV exposure compared to the change from the salt fog. The output of the function is of the unit inverse steradians. It is the ratio of light at a given scattering angle to the total incident light per solid angle. By multiplying by the total incident light intensity the data can be transformed to show intensity per steradian. The peak of the function is at a scattering angle of zero; the majority of the light is still reflected in this case. However, we have removed this peak to better show the changes in scattering at large angles.
5 Discussion

5.1 Scientific Contributions

The mechanistic underpinnings of polymer degradation, stabilizer kinetics and aluminum oxidization have been well researched and discussed in the literature, and are not discussed here. By discussing the observable phenomena of degradation in terms of regions 1, 2 and 3 we are able to discuss degradation rates without discussing the particular degradation pathway. Relating highly different exposures in terms of net stress and stress level allows for exposure of materials to high levels of stress while accelerating the same fundamental phenomena.

5.2 Technology Development with L&DS

Technology development of back surface acrylic mirrors was undertaken. Appropriate substrate substitution was made to ensure the stability and performance of the system. Development of a library of exposed samples and metrics for analysis was performed so that field retrieved samples, outdoor exposed samples and


5.3 Informed Substrate Selection

The acrylic degradation study shows that stabilizer content is vital to performance. While UVT acrylic yellows and degrades fairly rapidly, the additional additive package in MP-grade acrylic delays the onset of rapid yellowing. Degradation of the UV absorber package is observed in initial UV exposures. As the stabilizer package is bleached from the substrate, the exposed surface appears to yellow fairly rapidly. In the Newport Solar Simulator, a change in the degradation mode occurs after approximately 100GJ/m$^2$ full spectrum dose which is equivalent to 8-12 years of tracked exposure in Arizona. After this point, an increase in yellowing rate is observed. As the material begins to yellow quickly, absorption causes elevated temperatures which can exacerbate UV degradation. UV degradation in MP acrylic was predominantly observed on the light-exposed side, which can be understood because absorption of this light is exponential through the samples.

UVA acrylic shows to be highly resistant to UV degradation. This is a direct result of UV absorber concentration. UVA acrylic was subjected to over 227GJ/m$^2$ of full spectrum radiation, equivalent to over 20 years of tracked exposure in Arizona. Degradation in UVA acrylic subjected to UV-only exposure was not observed.

5.4 Metallization and Mirror Coating Lessons

Aluminum metalization appears to be more robust than silver metalization used for most silver mirrors. While aluminum's second surface reflectance is somewhat lower, the introduction of more stable material system into the solar mirror market may be valuable to some product developers. Aluminum metallization does not tarnish and
darken the way that silver metallized systems do, and do not appear to require 2-layer nor lead-based protective coating systems for robust designs. The white paint coating system appears to be more robust when exposed to high levels of humidity and heat, but both coating schemes may be adequate for mirror stability.

5.5 Suitable Applications for Acrylic Back Surface Mirrors

Acrylic back surface solar mirrors developed by Replex Plastics are different from other solar mirror products. Glass substrates used for CSP mirrors provide mechanical rigidity required for long throw-length optics. Silver mirrors which are fairly standard in CSP and CPV applications offer a higher degree of reflectivity than aluminized mirrors. Aluminum does however outperform silver mirrors in terms of durability and ease of manufacture. A single layer protective coating has so far shown to be durable to accelerated weathering equivalent to 4.1 years of light with 650 hours of condensing humidity. Replex's Solar-Grade back surface mirrors are spectrally filtering, reflecting very little UV light and thereby providing a way to mitigate UV degradation associated with cell-level UV degradation for CPV applications.

5.6 Applications and Advantages of Acrylic Back Surface Mirrors with L&DS

For companies interested in developing low-cost high solar technologies that depend on long lifetimes, Replex solar-grade mirrors offer many benefits.

Acrylic substrates allow for a higher degree of flexibility while retaining high optical quality. Acrylic is a good material for these applications because it is lower cost than glass and easily formable enabling rapid product development cycles. UVA acrylic
has proven highly radiation durable Spectrally filtering mirrors may allow for use of traditional 1-sun materials for CPV applications because they do not introduce high UV loads.

Reflectance and BSDF analysis can allow for quantitative prediction of system performance over product lifetime because parameterization of degradation phenomena allows for quantitative optical modelling of systems through time. This can allow developers to know that their systems perform to their expectations throughout their lifetime.
6 Suggested Future Research

6.1 Continuation of Exposures and Analysis

Any prediction of future performance comes with risk. Accelerated weathering protocols are useful because they are repeatable. This allows for fair comparison of different types of material systems. If degradation observed in fielded materials and components matches the degradation observed after accelerated weathering protocols correlations can be drawn and predictions of future performance can be made with higher confidence.

Outdoor exposure can be performed on fixed angle weathering mounts, on trackers that follow the sun for maximum natural radiation exposure and on accelerated outdoor weathering testers. Accelerated outdoor exposures typically use reflectors to increase solar exposure past typical 1-sun levels. Though exposures of these types are typically run for 3-5 years, the value of weathering information increases as samples age and many exposures continue until failure. It is important when designing real-world testing protocols that multiple exposure sites are used. Exposures performed in Arizona are characterized as "hot and dry" and may correlate better to weathering data from the "Hot QUV" exposure, while weathering in Florida is considered "hot and humid" and may correlate better to QUV weathering with humidity. Weathering in temperate
climes, urban environments, near salt-water and in very wet locations may be important for determining the degradation modes in fielded systems. Environmental stress due to hail strikes, acid rain, salty conditions and extended rain may cause unforseen degradation outside the scope of this work.

Replex Plastics’ solar mirror samples are under real world weathering in Cleveland, Ohio but exposure in different locations is necessary for a better characterization of the durability of this system. Accelerated weathering protocols may also enable confidence in the accelerated weathering data presented.

6.2 Development of First Surface and Silver Mirrors

In order to enable efficient light harnessing in HCPV applications, spectrally filtering mirrors cannot be used. Because multi-junction devices are current limited by the most poorly performing junction, full spectrum light, that includes all of the UV, is needed. Development of mirrors used for HCPV applications would be enabled by employing the same technique, but using transparent protective coatings. Since there are many transparent coatings available, development of these mirrors would require a new set of exposure protocols.

Development of these technologies might also provide an opportunity to experiment with silver metallizations. Silver metallizations can enable a higher level of reflectance. Manufacture of silver mirrors relies on enhanced processing, multi-layer fabrication techniques and development of hermetic two-layer coating systems. Silvered acrylic mirrors have been developed, but interfacial interactions at the silver-PMMA interface have shown to be difficult to control, as such a new substrate may need investigation.
6.3 Application of Edge Seal

Degradation of the solar mirrors appeared to proceed from the edge of the sample, where the aluminization was in direct contact with the exposure environment. While this degradation so far has not caused failure, application of an edge seal can provide enhanced degradation resistance. Mirrors with an edge seal are currently under exposure and results will be correlated to the unsealed mirror samples used for this study.

6.4 Investigation of Mechanical Strength of Acrylic Substrates

All of the responses used for evaluation of these solar mirrors was focused on the optical quality of bulk materials and object dependant optical performance. While these metrics are critical for predicting performance for power applications, resistance to hail and wind loads may also degrade with exposure. Destructive tests that require cutting mechanical test specimens or performing hardness tests would provide this information, but would reduce the library size for future studies. Results of such testing may be warranted if mechanical strength is found to degrade over time.
EDUCATION
Masters of Science, Materials Science and Engineering
Case Western Reserve University, Cleveland, Ohio, Jan 2013
Bachlors of Arts, Chemistry
Hendrix College, Conway Arkansas, May 2008

COMPUTER SKILLS
Languages & Software: LaTex, Mathematica, Adobe Photoshop, Adobe Indesign, System Advisor Model (NREL), TracePro (Lambda RES), PROLith (KLA-Tencor)
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EXPERIENCE
Technology Development 2010-2013
Replex Plastics, Mt Vernon, Ohio
• Developed, characterized and performed lifetime prediction for Replex Plastics Solar-Grade Mirror
• Developed metrics for predicting future performance of Mirror-Augmented Photovoltaics

Roger French: 216-368-3655

PV Quality Assurance Taskforce 2011-2013
NREL PVQA Taskforce Working Groups 3,5
• Attended workshop at SPIE Solar in San Diego, CA
• Continued work in support of new quality assurance standards.

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Grant Writing/Support Winter 2010
Materials Science Engineering Dept. Case Western Reserve University
• Developed dynamic budgeting modelling tool for various research projects.
• Support role on grant writing & scoping, oversaw purchasing more than $1,000,000 in capital expenses, implemented initial 3D design of proposed system. Grant awarded 2011, Ohio Third Frontier Tech 11-060

Teaching Assistant Winter 2010
Materials Science Engineering Dept. Case Western Reserve University
• Applied Thermodynamics (2012)
• Optical Materials, Elements and Technologies (2011)
• Introduction to Materials Science and Engineering (2010)

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Environmental Monitoring Specialist 2009-2010
Cleveland Division of Air Quality
• Operation and maintenance of air quality monitoring equipment.
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Nursing Office Aid 2008-2009
Neighborhood Family Practice
Recognized need for, developed, and wrote mail merge program before implementation of EHR. Performed various duties including technical support, mailings and inventory control.

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Student Researcher
Hendrix College

- Procured water samples, used EPA guidelines to develop methods of analysis and sampling techniques for water quality analysis. Oversaw purchase and installation of new equipment and wrote operational protocol.

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VA Hospital Infectious Research Division Cleveland, Ohio

- Washed and autoclaved biological equipment. Grew cultures and tested for optical density, viability and protein concentration.

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- Supervised other student workers, worked alongside safety coordinator and lab manager, set up and dismantled undergraduate experiments.

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COMMUNITY
SERVICE
Poster Session Chair, IEEE Energy Tech
Volunteer at Weapons of Mass Creation Festival 2012
Detroit Shoreway Earth Day neighborhood cleanup
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Tim Hagan Citizenship Award for service to community 2004

EXTRA-
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Attended Governor’s Task Force on Energy Policy Columbus, OH 2011
Appointed Editor in Chief, Aonion Literary Magazine, Hendrix College 2007-2008
Participated American Collegiate Dance Festival Iowa City, Iowa 2008
Awarded Eagle Scout Boy Scouts of America, Troop 22 Cleveland, Ohio 2004
Crooner & Fiddler in Crooked River Blues Band 2009-2013
Publications


Murray, Myles P., *Cleveland Needs Solar Power* Cleveland Plain Dealer 8/7/12


Talks & Posters

**Talks:**
- SPIE Solar 2012 San Diego, California
- IEEE Energy Tech 2012 Cleveland, Ohio
- MRS Fall Meeting 2011 Boston, Massachusetts

**Posters:**
- Workshop on Interfaces 2011 Seville, Spain *Introduction to the Sun Farm: Outdoor Exposure Capabilities at Case Western Reserve University*
- IEEE Energy Tech 2011 Cleveland, Ohio *Durability of Acrylic Back-Surface Mirrors for Low Concentration Photovoltaics*
- PVSC conference 2011 Seattle, Washington *Radiation Durability of Materials Components and Systems for Concentration Photovoltaics*
- PVIC general meeting 2010 Columbus, Ohio *Introduction to the Sun Farm: Outdoor Exposure Capabilities at Case Western Reserve University*
- ACS Yearly Meeting, New Orleans, 2008 *Measurement of dissolved metals in Brewer Lake, Arkansas*
- Midwest Enzyme Chemistry Conference, November 2005 *Class A Beta-Lactamase Production is Influenced by Beta-lactam Concentrations for Inhibitor Resistant Enzymes but not for ESBLs or Wild Type*
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