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Effects of Cerium Removal from Glass on Photovoltaic Module Performance and Stability

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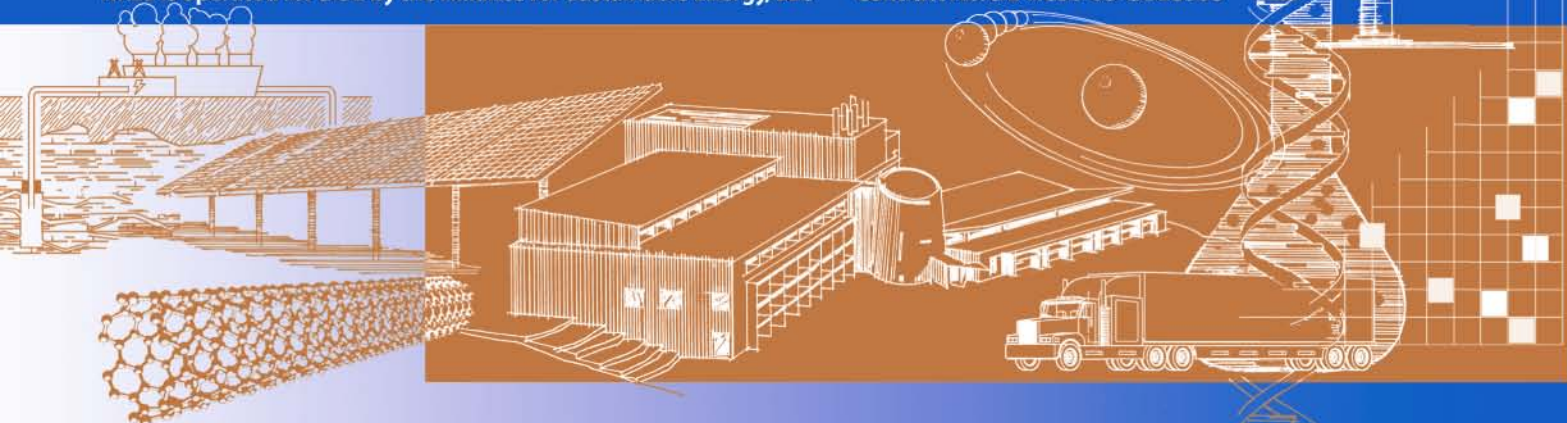M.D. Kempe and T. Moricone
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Effects of Cerium Removal From Glass on Photovoltaic Module Performance and Stability

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ABSTRACT

Photovoltaic modules are exposed to extremely harsh conditions of heat, humidity, high voltage, mechanical stress, thermal cycling and ultraviolet (UV) radiation. The current qualification tests (*e.g.* IEC 61215) do not require sufficient UV exposure to evaluate lifespans of 30 years. Recently, photovoltaic panel manufacturers have been using glass that does not contain Cerium. This has the advantage of providing about 1.3% to 1.8% more photon transmission but potentially at the expense of long term stability. The additional transmission of light in the 300 nm to 340 nm range can cause delamination to occur about 3.8 times faster. Similarly, UV radiation will cause polymeric encapsulants, such as ethylene vinyl-acetate (EVA), to turn yellow faster losing photon transmission. Silicones do not suffer from light induced degradation as hydrocarbon based polymers do, therefore if silicone encapsulants are used, a 1.6% to 1.9% increase in photon transmission can be obtained from removal of Ce from glass, with no tradeoff in long term stability. Additionally antimony can be added to non-Ce containing glass to further improve photon transmission (principally in the IR range) by an additional 0.4% to 0.7%; however, this does not significantly affect UV transmission so the same UV induced reliability concerns will still exist with common hydrocarbon-based encapsulants.

Keywords: Cerium, Photovoltaic, Glass, EVA, Stability

1. INTRODUCTION

In photovoltaic (PV) modules, polymeric materials provide electrical insulation and protect modules from mechanical damage and environmentally induced corrosion. When used in front of a PV cell the principal attributes of interest are that the encapsulant transmits photons and maintains adhesion to all surfaces. The PV module qualification tests (such as IEC 61215 [1]) are designed to provide minimum standards for module durability and to demonstrate a degree of safety in the production of electricity [2, 3, 4]. Because of the extreme difficulty of exposing production modules to concentrated light sources, the UV exposure required by these tests corresponds to an equivalent field exposure of several months to 1.5 years rather than the desired service life of 30 years.

During the early development of PV technology in the 1950s and 1960s, polydimethyl silicone (PDMS) based encapsulants dominated designs because of their exceptional UV resistance and thermal stability [5]. Because of the higher price of PDMS relative to other materials, effort was focused on development of lower cost alternatives. This effort resulted in the Flat Solar Array PV block purchase program in the 1970s and 1980s [6]. With this program, laminant encapsulation films composed of copolymers of ethylene vinyl acetate (EVA) were developed as a compromise between superior performance of PDMS and the lower cost of EVA. In the mid 1980s there were a number of notable problems with EVA yellowing [7, 8, 9]. The block V purchase installed 6 MWp of solar PV between 1983 and 1985 at Carissa Plains California. However, by 1988 the system performance was down more than 30%. This degradation was partially the result of polymer embrittlement, delamination and/or discoloration ("yellowing") [10, 11]. One component of the solution to these issues [12, 13] was to use glass containing Ce, which selectively blocks UV-B radiation [14, 15, 16, 17]. In recent years better stabilizer formulations for EVA have been developed, giving more confidence in the long-term stability of PV packaging materials [18,19,20]. Because of this, some manufacturers have stopped using Ce-doped glass.

The use of glass doped with Ce can be problematic. In typical low-Fe Ce-doped glass, the Ce is present in both the Ce^{3+} and Ce^{4+} states. It is the Ce^{3+} state that absorbs ultraviolet light (UV) with peak absorption around 314 nm [17]. Upon exposure to UV radiation, the Ce^{3+} will be oxidized to the Ce^{4+} state. Because diffusion of oxygen into soda lime glass

occurs on geologic time scales, Ce cannot be oxidized without a corresponding reduction reaction. In glass containing iron, Fe^{3+} will be reduced to Fe^{2+} as,

$$Ce^{3+} + Fe^{3+} \xrightarrow{\text{UV Light}} Ce^{4+} + Fe^{2+}$$

Less light transmission
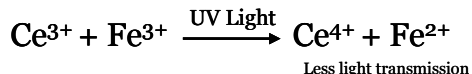
Unfortunately, Fe^{2+} has a broad absorption peak around 1050 nm [21,22,23], which is within the useful range of typical silicon based PV technologies. Fe^{3+} has a relatively weak absorption peak in the UV range around 370 nm [24]. Thus, "solarization" results in a small decrease in PV performance upon field exposure of Ce-containing glass. This work is an effort to help quantify the magnitude of this effect and to give insight into the potential benefits and problems associated with Ce containing glass.

2. EXPERIMENTAL METHOD

Ultraviolet exposure was obtained using an Atlas Ci4000 Weather-Ometer® with a light intensity of 114 W/m^2 between 300 nm and 400 nm at the typical sample plane located at 32.4 cm from the Xenon Arc lamp (Fig. 1). The light was filtered using a type "S" borosilicate inner and outer filter. This light intensity equals about 2.5 AM 1.5 global UV Suns. To further increase the UV dose, test samples were also placed at a radius of $r = 7$ cm from the lamp. The UV dose at this radius was estimated to be about 42 UV suns by approximating the lamp as a 12.5-cm long line source with the light intensity decreasing as $1/r^2$ from each point along the length. At this close distance, additional ducting was required in the Weather-Ometer™ to maintain reasonable specimen temperatures. Chamber conditions were maintained at 30°C and 30% relative humidity (RH) resulting in a sample temperature of 80°C to 95°C for new, transparent samples [25].

Atlas Ci4000 setup for high UV flux irradiance
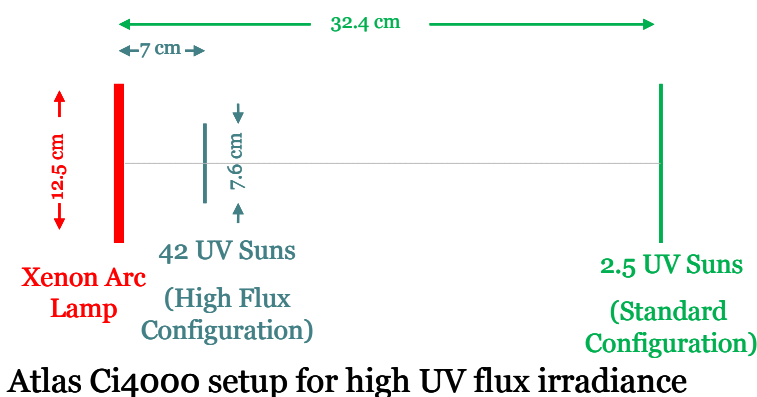
**Figure 1**. Test geometry in cross-section for ultraviolet exposure.

The percent transmission through samples was measured using a Lambda 9 UV-Vis spectrophotometer (Perkin-Elmer, Inc.) equipped with an integrating sphere or with a Cary spectroradiometer (Varian Co.). Some samples began to degrade at their perimeter as a photo-oxidation process dominated the degradation. Other specimens (including all EVA samples tested), turned yellow in the center first as oxygen ingress serves to remove chromophores at the perimeter [26]. In both cases, these perimeter oxidative effects were limited to the outer few mm of the samples. Because flat-plate PV modules typically use a glass front sheet located in front of a relatively large cell, it was thought that the anaerobic center of our samples would be more representative of the majority of the encapsulant used in a PV application. Therefore, all transmission measurements were made in the center of the samples.

Lap shear tests were conducted as described previously by Kempe *et. al*. [27] using an Instron Test Unit (model 1122/5500R) for samples as shown schematically in Fig. 2. Two 5.61 mm thick 7.6 cm × 7.6 cm glass pieces were used to construct the test specimens. The polymer under test was applied to an approximately 19-mm-square area at a thickness of about 0.5 mm, similar to the thickness typically used in a PV module. The black panel standard temperature was maintained at 100ºC ± 7ºC resulting in a temperature of 70ºC to 80ºC for the transparent glass lap shear samples.
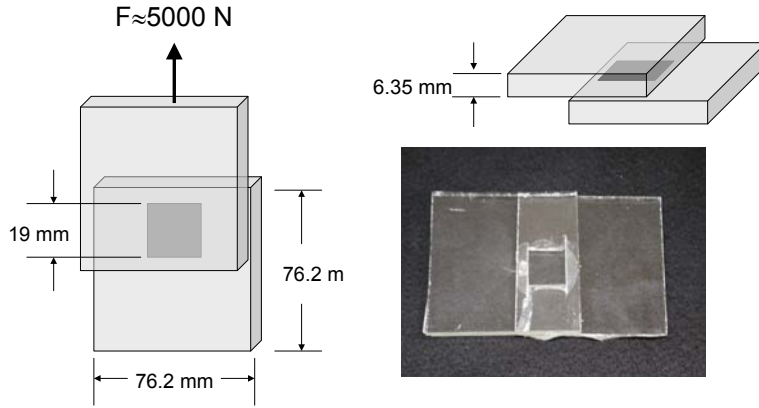
Figure. 2. Schematics and photo of the lap shear design.

All the polymeric samples used in this study were obtained from commercial sources and were used in accordance with the manufacturer's specifications (Fig. 3). In the figure, PDMS is unique, since it does not bear a carbon backbone.
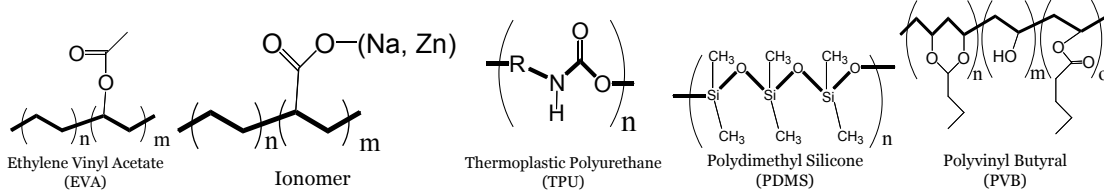

Figure 3. Generalized Polymer Structures

3. RESULTS

3.1 Light Transmission for Different Glass Samples

The light transmission through a variety of glass samples is plotted in Fig. 4 to show how the UV portion of the spectrum is attenuated. This is useful to illustrate how a small portion of the solar spectrum can have a profound effect on polymer stability. Also included in Fig. 4 is an internal quantum efficiency curve obtained as an average of 8 different crystalline silicon cells. A curve for the light transmission through stabilized UV-absorbing EVA is shown indicating that even if the UV light makes it through the glass most of it won't transmit through the EVA., Furthermore, the quantum efficiency in the region is also very small for Si. From Fig. 4, one can see that blocking light in the UV region will have a negligible effect on PV module efficiency.
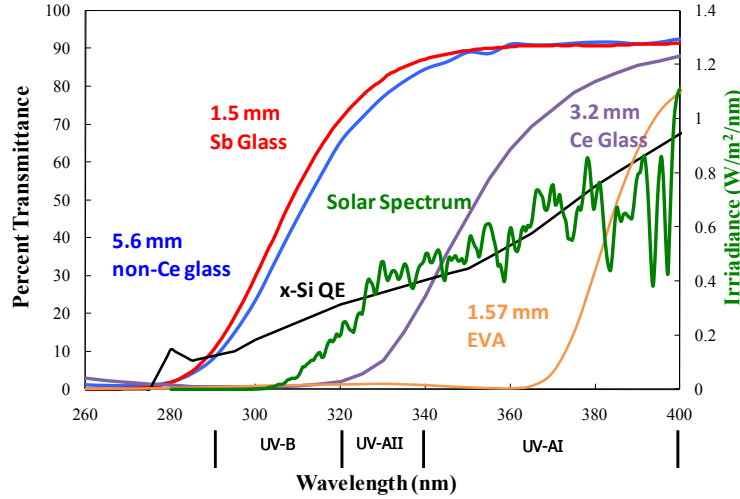
Figure 4. Glass transmission after solarization by exposure to 114 W/m^2 (300 - 400 nm) in a Ci4000 Weather-Ometer® at 60ºC and 60% RH. Solarization only takes a few hundred hours under these conditions. The AM 1.5 global solar spectrum is also shown [28]. The measurement for EVA was obtained from a laminate construction consisting of 3.2 mm Ce Glass/1.57 mm EVA/3.2 mm Ce Glass.

Because of the absorption of Fe^{2+} in the near-IR region, the choice of glass composition will affect the overall module performance. To quantify this, the absorptivity as a function of wavelength [$\alpha(\lambda)$] was estimated for a variety of glass and polymer samples. The normal-incident transmittance, T, through a thick plate can be analyzed according to Eqs. 1 and 2, where the refractive index of the surrounding medium is assumed to be 1, t is the plate thickness, k is the extinction coefficient, n is the real component of the refractive index for the glass, and $\alpha=4\pi k/\lambda$,

$$T = \frac{(1-r_i)^2 e^{-t\alpha}}{1-r_i^2 e^{-2t\alpha}} \tag{1}$$

$$r_i = \frac{(1-n)^2 + k^2}{(1+n)^2 + k^2} \tag{2}$$

Using values of n obtained from Rubin [29], Eqs. 1 and 2 can be solved numerically to estimate $\alpha(\lambda)$ [30]. The absorptance of polymers was obtained from materials laminated between two glass plates [Fig. 5(B)]. Assuming, the reflection at the interface between polymer and glass is at most approximately 0.17% (n~1.4 for silicone and n~1.52 for glass), and that the absorption in the polymer layer upon subsequent reflections is relatively small, the total transmission of these samples (T_{tot}) can be estimated by Eq. (3)

$$T_{tot} = T_{glass} e^{-(t_p \alpha_p)}. \tag{3}$$

Here T_{glass} is the transmission through a single plate of glass with a thickness equal to the sum of the thicknesses of the two plates of glass, t_p is the polymer layer thickness and α_p is the polymer absorptivity.
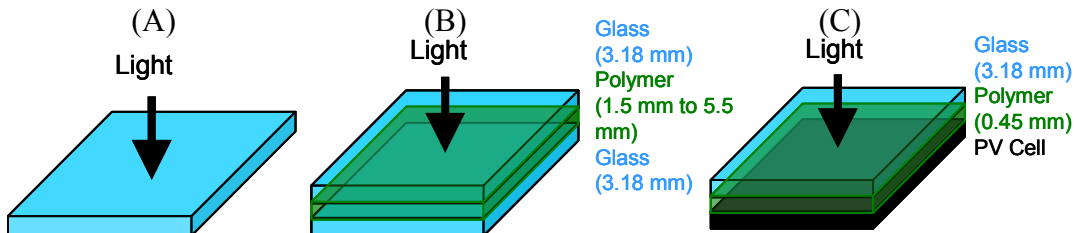
Figure 5. Schematic diagrams of samples used to estimate the useful photon flux for a PV device.

From the absorptivity of the different materials, the transmittance to a cell (located beneath the encapsulant) can be estimated using

$$T_{cell} = \left[1 - \frac{(1 - n_g)^2 + k_g^2}{(1 + n_g)^2 + k_g^2} \right] e^{-(t_g \alpha_g + t_p \alpha_p)}. \tag{4}$$

Here the transmission losses are represented by reflection at the glass/air interface, absorption in the glass, and finally absorption in the polymer neglecting higher order reflections [Fig. 5(C)]. Thus, this represents the photon flux available to a cell. To obtain an estimate of the effect of light on the J_{sc} produced by a cell, the transmittance estimated from Eq. (4) was weighted against the photon density of the AM 1.5 solar spectrum and also according to the quantum efficiency of a x-Si cell, obtained as an average of 8 different cell technologies. The results of these measurements are shown in Tables 1 and 2.

Table 1. Estimated photon transmission to a x-Si cell behind a 3.18-mm thick glass and 0.45-mm thick polymer layer. Transmittance is weighted against the AM 1.5 global solar spectrum and the internal quantum efficiency of a hypothetical typical x-Si solar cell obtained from the average of cells from 8 different manufacturers. A poly ethylene-tetrafluoroethylene (PETFE) superstrate is listed for reference..

Percent Transmission to Interface:	Global Solar Photon and Internal Quantum Efficiency Weighted (Glass is 3.18 mm thick, Polymer is 0.45 mm thick)								
	Ce Glass				Non-Ce Glass		Sb Glass		PETFE (0.0381 mm thick)
	Unexposed		Solarized		Unexposed	Solarized	#3	#4	
	#1	#2	#1	#2					
Glass Sheet Transmission	90.3	90.9	89.5	89.3	90.2	90.7	90.8	91.5	94.2
Glass/Polymer	94.1	94.8	93.6	93.1	94.8	95.1	94.6	95.4	95.5
EVA/Cell	93.4	94.1	93.0	92.5	94.0	94.3	93.9	94.7	94.7
DC 527/Cell	94.0	94.7	93.6	93.1	94.7	95.0	94.6	95.4	95.4
GE RTV615/Cell	94.1	94.7	93.6	93.1	94.7	95.0	94.6	95.4	95.4

Table 1 shows transmission results for glass with and without cerium and also for glass containing Sb. During the manufacturing of float glass, small additions of Sb along with careful control of the thermal and redox profiles can result in virtually complete oxidation of Fe^{2+} to Fe^{3+} [31].

$$Sb^{5+} + 2Fe^{2+} \longrightarrow Sb^{3+} + 2Fe^{3+}$$
High Photon Transmission

This has the result of creating a very highly transmissive glass in the near-IR that has a slight yellowish appearance rather than a typical green color. However, this process would also result in the oxidation of the UV absorbing Ce^{3+} ion resulting in high UV transmission.

$$Sb^{5+} + 2Ce^{3+} \longrightarrow Sb^{3+} + 2Ce^{4+}$$
Low UV absorbtion
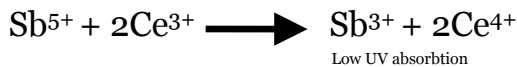
Because of this, the use of Sb to promote high optical transmission is not compatible with the use of Ce to block UV transmission.

Table 2 summarizes of the effects of glass substitution on photon transmission for different glasses in combination with different polymers. Here it can be seen that the removal of Ce can result in up to a 2.3% increase in J_{sc}.

Table 2. Effect of glass substitution on photon transmission. These differences are weighted by the AM 1.5 solar spectrum and a typical x-Si internal quantum efficiency; therefore, they should correlate well with J_{sc} changes.

Replace	EVA	Silicone
Ce with non-Ce	1.3% to 1.8%	1.6% to 1.9%
Ce with Sb	0.9% to 2.2%	1.0% to 2.3%
non-Ce with Sb	0.4% to 0.7%	0.5% to 0.6%

3.2 Effect of Glass Composition on Adhesion

The Ce in low-Fe glasses only absorbs a small fraction of light in the UV-B and UV-AII regions below about 340 nm (Fig. 4); however, this small number of high-energy photons has a dramatic affect on adhesion. In Fig. 6 the results of the loss of lap shear strength for EVA after exposure to 2.5 UV suns, 60°C, and 60% RH are shown for samples constructed using Ce or non-Ce containing glasses. Failure was always on the side of the specimen facing the UV lamp, indicating a dominant UV-induced degradation mechanism. Because, the UV light below 370 nm is significantly attenuated by the EVA, the effective UV dose is expected to be about 100 times smaller at the polymer/cell inteface [32]. Thus, it is not expected that UV light will accelerate delamination at the cell-to-EVA interface in a PV module.

The fit lines for the Ce and non-Ce EVA lap-shear data series are offset from each other by a factor of 8. However, because the glass used in these lap-shear samples is 6.35-mm thick rather than the 3.18-mm more typical of a PV glass this over-estimates the expected difference in adhesion retention for a PV module. An analysis of the activation spectrum performed in prior work by Kempe [30] estimated that the use of thinner glass would result in loss of adhesion about 3.4 to 3.8 time more quickly.
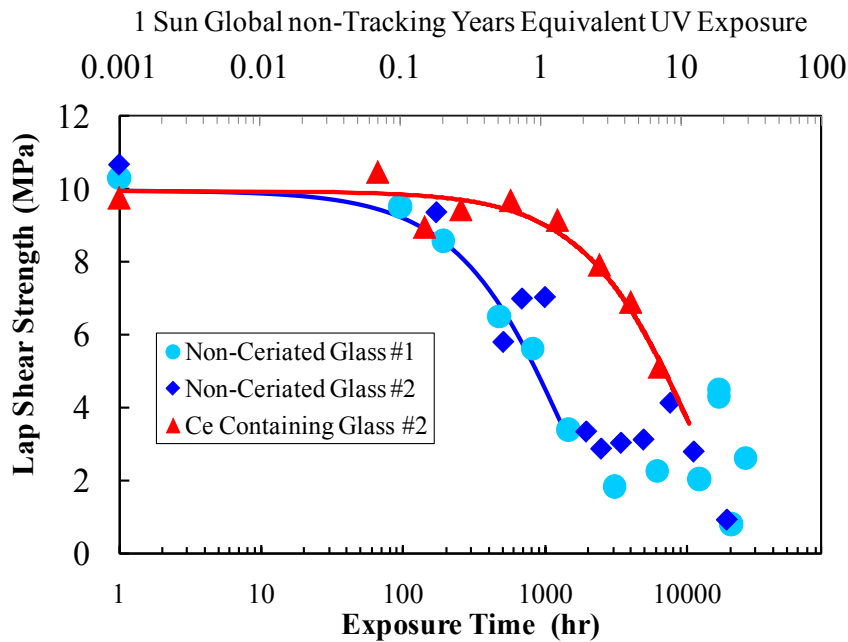
Figure 6. Lap-shear strength after exposure to 60ºC/60% RH at 2.5 UV suns. Samples #1 and #2 refer to slightly different formulations from the same EVA manufacturer [30].

Lap-shear measurements were also conducted for PDMS materials subjected the same exposure conditions as the EVA specimens (Fig. 7). The PDMS lap-shear samples proved more durable over time than EVA. After almost 3 years of exposure to 2.5 UV suns at elevated temperatures, the lap-shear strength is sustained, even being possibly increased. The failure of the silicone materials was in a cohesive manner within the polymer matrix, indicating that the interfacial adhesion strength is higher. Therefore, a slight increase in lap-shear strength is due to increased cross-link density caused by thermal and/or UV induced processes. It should also be noted that the silicone materials showed no signs of yellowing, whereas the EVA samples were discolored [30].
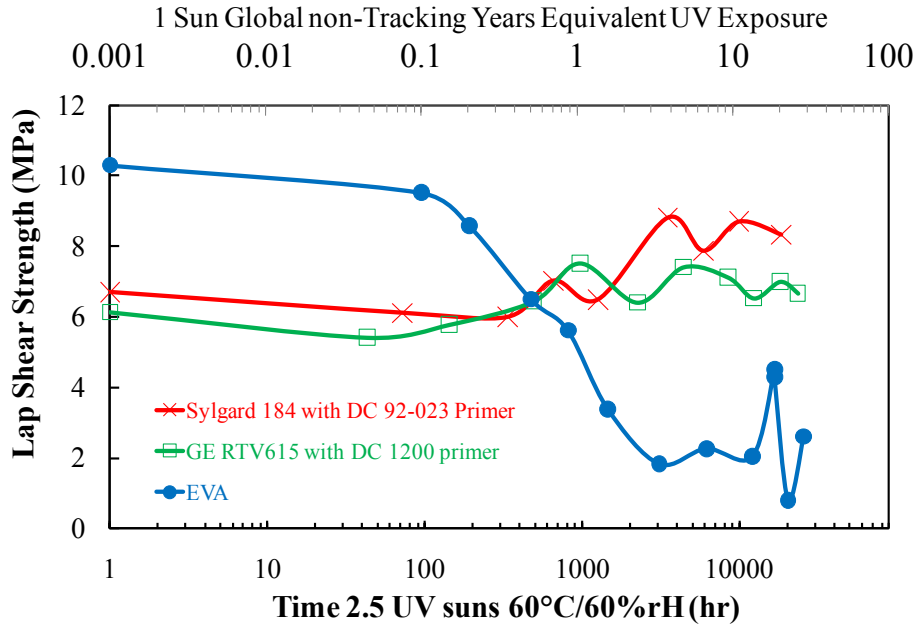
Figure 7. Lap-shear strength after exposure to 60ºC/60% RH/2.5 UV suns. Sylgard 184 and GE RTV615 (now Momentive) are both addition cure PDMS.

For Figs. 6 and 7, the top horizontal axis indicates an equivalent field exposure in years. Because the UV exposure here is only 2.5 times higher than one would see in the application, the assumption of a linear relationship between dose and acceleration is not unreasonable. This equivalent exposure was calculated assuming a time-averaged dose of 250 W/m^2 as compared to 1000 W/m^2 of the AM 1.5 spectrum. This results in a total estimated acceleration factor of 10X. If this estimate is accurate, the silicone samples should not experience loss of adhesion strength for far more than the life of a PV module; however, EVA would be expected to experience significant adhesive strength loss midway through its expected life.

3.3 Effect of Glass Choice on Light Transmittance

In a separate set of experiments the effect of glass choice on light transmittance was investigated. Here a UV dose of about 42 UV suns was used to obtain an acceleration relevant for concentrating PV systems. When evaluating these results for 1-sun applications, the dose is so high that it is very unlikely that a linear relationship between UV dose and acceleration is reasonable; therefore, this analysis serves more as a qualitative demonstration. Typically the acceleration is related to dose by a power-law relation bearing an exponent that is most often less than one, making it likely that the acceleration is less than the nominal concentration [33].

Figure 8 shows the effect of 42 UV suns at a temperature of between 80°C and 95°C on four different EVA samples behind Ce (solid lines) and non-Ce containing glass (dashed lines). Samples were constructed by laminating EVA between two pieces of 3.18-mm-thick glass. The samples with Ce glass initially show some losses attributable to the solarization effect, but with stable performance thereafter. The non-Ce samples however begin to show degradation after 700 h. Of the four different EVA samples, two retained good transmittance throughout the experiment. This demonstrates the importance of selection of proper EVA formulation if one uses a non-Ce glass in a PV module.
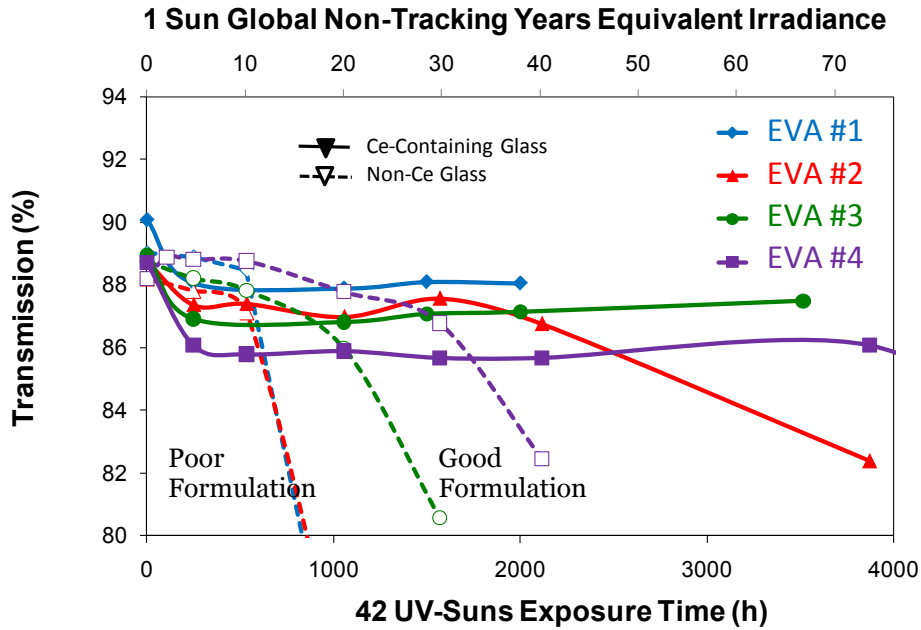
Figure 8. Transmittance and yellowness index for different EVA materials as a function of exposure to 42 UV suns of radiation behind different glass materials.

The top axis of Fig. 8 relates the equivalent UV dose as compared to a 1-sun non-tracking system. Using the same factor of 4 for temporal acceleration and 42 for dose acceleration, the total estimated acceleration is about 168×. From this estimate, the increased transmittance obtained by using non-Ce glass are likely to be lost by the end of the useful life of a PV module, resulting in only a marginal at best. This highlights the high potential of UV-B (λ=285 nm to 320 nm) to cause degradation.

Figure 9 shows that samples behind non-Ce glass initially demonstrate browning in the center but good transmission around the edges. This is presumably an oxidative bleaching effect promoted by oxygen ingress from the edges of the samples [26, 34]. Diffusion times are expected to vary with the square of the characteristic length scale. Taking a cell dimension of 15 cm as compared to the samples' size of 2 mm, the oxygen ingress rate should be about $15^2/2^2$=56 times slower. From this estimate, one would expect to still see discoloration (yellowing) in the interior of a module constructed using EVA.
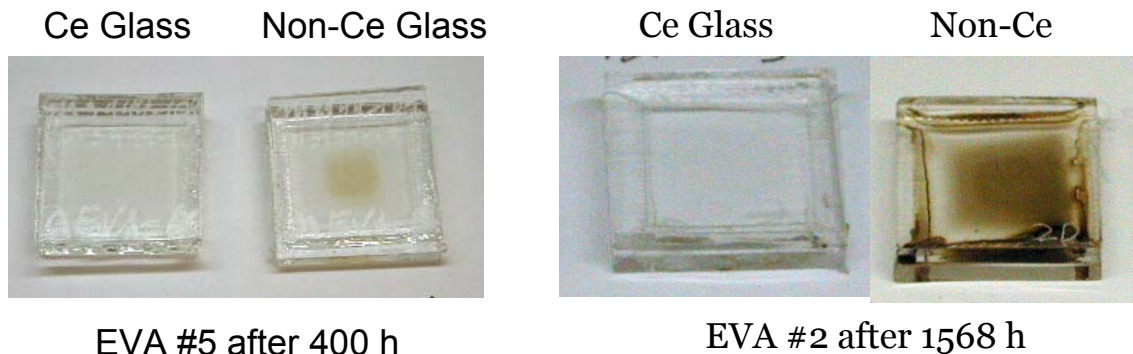

Figure 9. 1"x1" laminated composite EVAspecimens after exposure to 42 UV suns.

After 4000 h of exposure, even the samples behind Ce glass began to turn yellow (Fig. 10). However, the degraded area changed from the center to the perimeter indicating that oxygen ingress is now producing chromophores rather than consuming them. It would appear that one of the stabilization components is UV degraded to produce a chromophore

that is then degraded by reaction with oxygen. At long times, after (the stabilizer loses effectiveness), oxygen is able to react with another compound to produce a chromophore.

Ce Glass
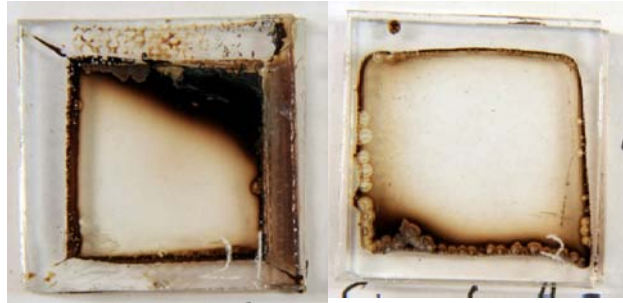
Figure 10. EVA #2 (Left) EVA #4 (Right) after 4125 h of 42 UV suns.

Many other materials are currently being considered for use in PV applications, some of which were also tested at 42 UV suns (Fig. 11). The PVB samples lost transmission very quickly even when UV blocking Ce glass was used. The TPU sample gave results similar to EVA. However, the ionomer gave exceptionally good results with virtually no loss in transmission for either glass type after 4000 h of exposure. The perimeter area of the ionomer specimens did show some photo-oxidative yellowing (Fig. 12). Because the extent of the yellowing is greater for the UV transmitting non-Ce glass, it is clear that the UV light is enhancing discoloration at the edge, suggesting the influence of oxygen or possibly ozone in the chamber.
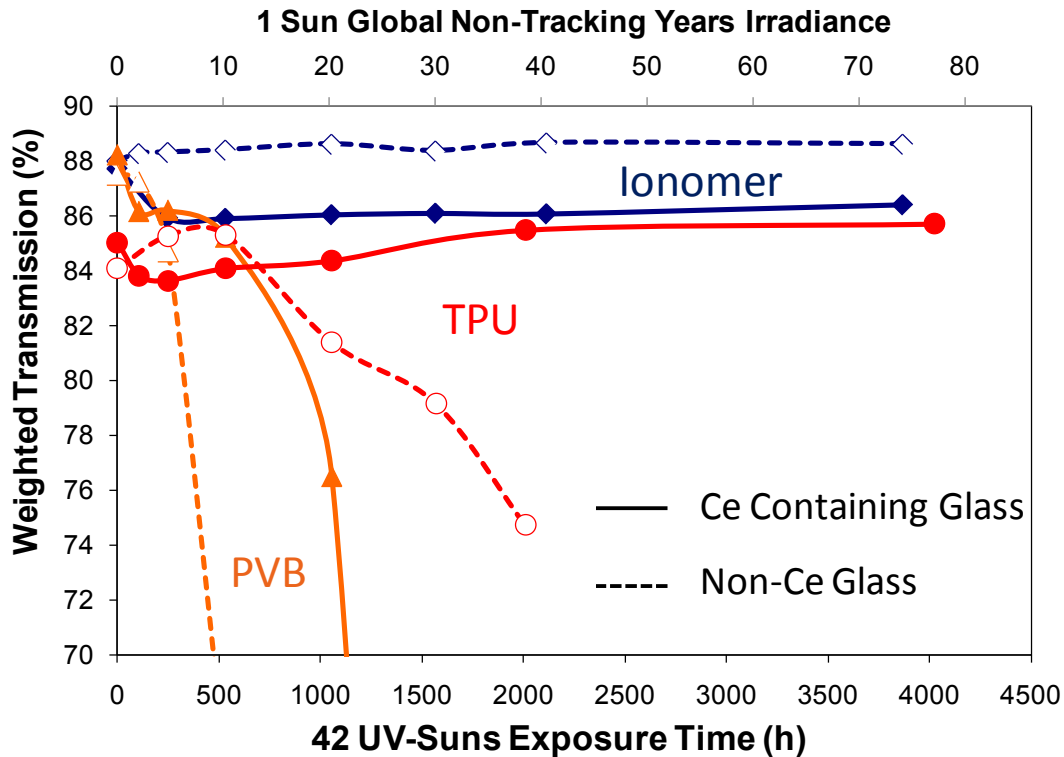
Figure 11. QE- and solar-weighted transmission for a thermoplastic polyurethane (TPU), an ionomer, and polyvinyl butyral (PVB) as a function of exposure to 42 UV suns.

Non-Ce Glass   Ce Containing Glass

Ionomer After 3900 h exposure

Figure 12. Photograph of ionomer sample after exposure to 42 UV suns.

Even under the increased flux of about 42 UV suns, the PDMS samples did not show any signs of degradation after 4000 h of exposure (Fig. 13). Visual inspection did not reveal any yellowing in the center of the specimens or around their perimeter. This, in combination with the lap-shear results, demonstrates the excellent UV and thermal stability of PDMS.
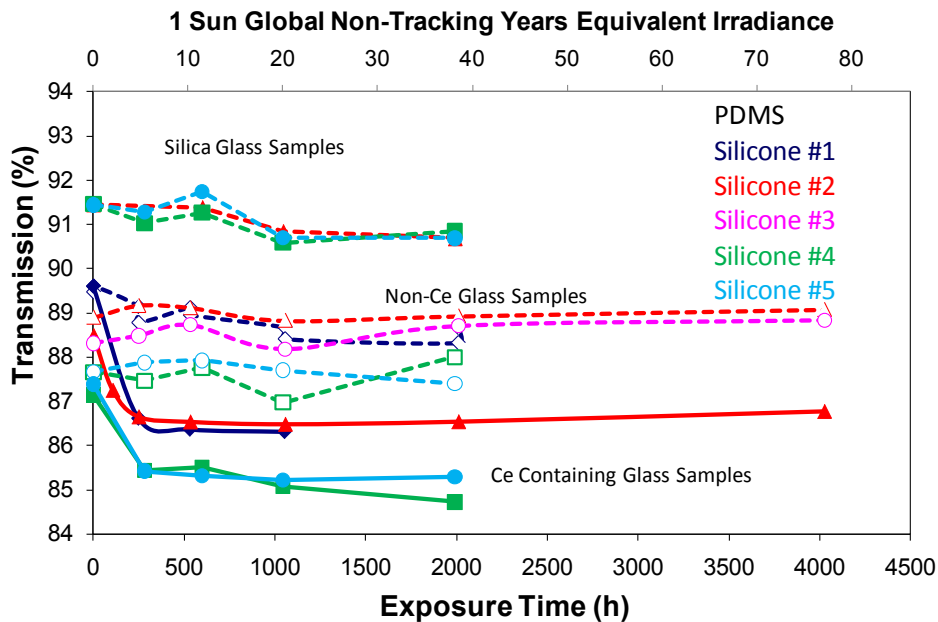
Figure 13. Measured optical transmission for PDMS specimens.

4. CONCLUSIONS

Experiments here have focused on the benefits and problems associated with the use of Ce in low-Fe glasses used in the PV industry. We have shown that the removal of Ce from PV glass provides improvements to J_{sc} of about 0.9% to 2.3% and that the addition of Sb can also improve light transmission. These benefits arise from improved transmission in the near-IR region because of a decreased concentration of Fe^{2+} ions in the glass. The improved transmittance in the UV range does not significantly improve the PV performance because of low intensity and low quantum efficiency in the UV, and because the encapsulants absorb much of the UV light .

Unfortunately the removal of Ce from glass can accelerate the delamination of EVA from the front glass by about 3.4 to 3.8x. Because most encapsulant materials strongly absorb UV light, Ce removal should not affect the adhesion of the

10

encapsulant to the PV cells. With regard to loss in transmittance through the encapsulants over time, significant problems were seen in the PVB sample tested. The EVA and TPU samples gave equivalent results at 42 UV suns, indicating that with regard to light transmittance, the use of non-Ce glass may be acceptable if a well-formulated material is used. The ionomer tested retained excellent transmission even without UV-blocking Ce. Lastly, the PDMS encapsulants showed no signs of UV-induced degradation with respect to either lap-shear strength or optical transmittance. If not for the typically higher prices for using PDMS materials, they would be the clear choice for PV modules.

Removal of Ce and addition of Sb to glass has the potential to improve PV module performance, but these improvements may be short lived because of enhanced UV degradation. One solution would be to use an antireflective coating that also blocks UV light below 350 nm. If one could also develop it to reflect the far-IR, the module temperature could also be reduced creating further improvements in module performance,

ACKNOWLEDGMENTS

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14. ABSTRACT *(Maximum 200 Words)*
Photovoltaic modules are exposed to extremely harsh conditions of heat, humidity, high voltage, mechanical stress, thermal cycling, and ultraviolet (UV) radiation. The current qualification tests (e.g., IEC 61215) do not require sufficient UV exposure to evaluate lifespans of 30 years. Recently, photovoltaic panel manufacturers have been using glass that does not contain cerium. This has the advantage of providing about 1.3% to 1.8% more photon transmission, but potentially at the expense of long-term stability. The additional transmission of light in the 300 nm to 340 nm range can cause delamination to occur about 3.8 times faster. Similarly, UV radiation will cause polymeric encapsulants, such as ethylene vinyl-acetate (EVA), to turn yellow faster, losing photon transmission. Silicones do not suffer from light-induced degradation as hydrocarbon-based polymers do; therefore, if silicone encapsulants are used, a 1.6% to 1.9% increase in photon transmission can be obtained from removal of Ce from glass, with no tradeoff in long-term stability. Additionally, antimony can be added to non-Ce-containing glass to further improve photon transmission (principally in the IR range) by an additional 0.4% to 0.7%; however, this does not significantly affect UV transmission, so the same UV-induced reliability concerns will still exist with common hydrocarbon-based encapsulants.

15. SUBJECT TERMS
PV; cerium; glass; ethylene vinyl-acetate (EVA); stability; module; photon transmission; silicone; antimony; ultra violet transmission;

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