

PROJECT #26: NEW MATERIALS FOR WINDOWS OF THE FUTURE

FINAL REPORT









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1 Introduction

Double alazed windows are a norm in many countries across Europe, Asia and North America due to their effective reductions of heat loss (~30%) compared to single -glazed windows. However, in Australia approximately 10% of current window installations are double glazed [i]. This arises in part due to the temperate climate across many parts of Australia, but equally their high costs. This high cost arises in part because of the high relative cost of the individual glass windows within the conventional window-gap-window double glazed configurations, as well as the enhanced challenges in their installation due to the sheer weight of these structures. An alternative approach towards achieving double glazed configurations is to utilise polymer-based sheets, such as perspex (i.e. poly(methyl methacrylate), that replace the glass panels. Polymers tend to be inherently cheaper and lighter in weight than glass; however, can be less environmentally stable and more porous and lack the optical clarity of glass. While a detailed assessment of the general properties of specific polymers is presented in later sections of this report, we start by outlining some of the more general challenges that need to be engineered around and relevant Australian standards that need to be complied in order to fully harness polymers for glazing applications.

1.1 Adverse aspects of plastics

Oxidation and degradation

Being organic-based, plastics are therefore subject to degradation and breakdown through natural oxidation. Degradation may be initiated or accelerated by numerous factors, including ultraviolet light, visible light, ozone, pollutants, manufacturing impurities, oxygen, and heat. Figure 1 shows the mechanism for a general polymer auto-oxidation process. Auto-oxidation refers to oxidation brought about by reactions with oxygen at normal temperatures without intervention, such as a flame or electric spark. Free radicals are initially formed from photo and thermal degradation. They react in the presence of oxygen to form peroxy radicals, which further react with organic material leading to hydroperoxide formation (ROOH) [ii]

During degradation, oxidation can result in the breaking and rearranging of chemical bonds along the polymer chain, making it susceptible to embrittlement and surface erosion. The oxidation also results in the loss of optical properties, including reduced light transmission, loss of gloss, increased haze and discoloration [iii]. The oxidation process can begin even during the manufacturing process, due to the conditions of extreme heat or pressure that may be used to form the polymer.



Figure 1. General polymer auto-oxidation process.

To delay the onset of degradation as long as possible, both process stabilisers and long-term stabilisers are added to the polymer. The most important primary anti-oxidants used for long-term stabilisation are the sterically hindered phenols. Unfortunately, these exhibit an undesirable side effect when exposed to ultraviolet radiation. They develop deeply coloured molecular structures resulting in irreversible yellowing of the plastic. To minimise this effect, polymers are also provided with an ultra-violet stabilisation package. This package is often made up of ultraviolet absorbers (UVA) and hindered amine light stabilisers (HALS).

Low scratch resistance

Plastics are generally much softer than glass and thus vulnerable to scratching. Pencil hardness measurements have been used by the coatings industry for many years to determine the hardness of clear and pigmented organic coating films; it is also often used to determine the scratch resistance of plastics. During a pencil hardness test, the pencil lead is maintained at an angle of 45° and pushed with uniform pressure on to the sample. Pencil hardness of plastics generally ranges from 2B to 4H, whereas glass will not be scratched by pencil. Glass is rated 5.5 at Mohs scale of mineral hardness. A pencil is made of Graphite which only rates at 1-2H on the Mohs scale.

1.2 Australian Standards

Any selected plastics need to meet the Australian standards (AS 1288—2006) if they are to be used as glazing materials.

Compressive strength

When tested in accordance with ASTM C1279, heat-strengthened glass shall have a surface compression strength of 24–52 MPa. Toughened glass shall have a surface compression strength of not less than 69 MPa.

Maximum deflection

The maximum deflection for all glass under serviceability shall be limited to -

(a) span/60 for two-, three- or four- edge supported panels; or

(b) height/30 (or cantilever length/30) for cantilevered panels such as cantilevered structural glass balustrade.

• Service life

In general, a window is expected to have at least 25-year service life . Table 1 summarize the lifespan of windows with various frame types [iv]. Table 1. Lifespan of window units, various frame types

Frame material	Survey results (years)	Literature review range (years)
Aluminum	43.6	35 - 60
UPVC	24.1	25 - 30
Timber	39.6	35 – over 60
Aluminum-clad timber	46.7	45

1.3 Objectives

The adverse aspects of plastics can be resolved through appropriate coatings that serve to enhance their specific function, whether it be infrared reflection, hardness, moisture or oxygen ingress, and colour. This project is focussed on providing a critical assessment of where such plastic-based double-glazed systems are currently at compared to glass-based solutions. Furthermore, it aims to identify the key technical challenges and opportunities that are evident for such plastic based solutions to reach competitiveness with glassbased systems.

The overall objectives of this report are to:

- 1. Review the state-of-the-art in double glazed windows
- 2. Review plastic-based double-glazed windows
- 3. Identify near-term research and development gaps and opportunities for plastic double-glazed windows

2 Review of Existing Glass Double Glazing

In this section, the basic physics of glazing is presented. Conventional glass double glazing, including their materials and coatings will be reviewed.

2.1 Basics of glazing

When the sun ray reaches glazing material, it is either transmitted, reflected, or absorbed by the glazing depending on its optical properties. Sun ray consists of visible, and invisible radiation that covers a broad spectrum of wavelengths. Solar energy is distributed differently among these spectra. As it can be seen in Figure 2 [v], the solar energy distribution is different for various wavelength. Solar energy from invisible solar spectrum comprises more than 50% of the solar energy. Controlling the invisible solar spectrum using different glazing materials, and coatings can play a significant role in building energy savings.



Figure 2. Solar energy distribution.

UV radiation is divided into three classifications called UV-A, UV-B, and UV-C. Of these three classes, UV-A and UV-B are associated with sunburns and tanning, and UV-A is thought to suppress the immune system. UV-A and UV-B are also associated with tanning beds, and there are hazards associated with the overuse of these beds. UV-C is produced only by specialized lights and the sun. When produced by the sun, UV-C does not penetrate the earth's atmosphere.

Conventional glass glazing will allow UV-A to pass through while almost 100% of the UV-B and UV-C light is blocked. Therefore, using standard glass glazing, some UV radiation can enter home and potentially affect the users' skin. Some of these effects could include increased freckles and increased sensitivity to sunlight that could result in rashes such as photodermatitis.

Three key parameters are used to evaluate the performance and characteristics of glazing and windows: overall heat transfer coefficient (U), visible transmittance (T_v) , and solar heat gain coefficient (SHGC).

 U indicates heat loss through the window assuming indoor and outdoor environment temperature differences. Factors that have substantive impact on the U values include the window types (single-glazed, doubleglazed, triple-glazed), frame types and the frame structures (length and area of the frames). It is noted that, for double glazing and triple glazing, the thickness of glass panes have little contribution to U values.

- SHGC is the measure of solar thermal energy transmitted directly or indirectly (absorbed and then transmitted inward) and ranges from 0 to 1 for which higher value means higher solar heat transmitted.
- T_v is a factor for visibility of glazing material and is the portion of visible light that passes through the window. Table 2 captures the window properties suggested for three climates [v].

climate	Tv	SHGC	U(Wm ⁻² K ⁻¹)
Cool climate (prevailing heating loads)	> 0.70	> 0.60	< 2
Temperate climate (prevailing heating loads)	> 0.70	> 0.50	< 2.5
Hot climate (prevailing cooling loads)	> 0.60	< 0.40	< 4

Table 2. Suggested window properties for different climates.

Windows should have different properties for various climates. The ideal window for hot climate is a window that reflects all the infrared radiation (including from sun and environment), and UV, while allowing all visible light to enter and being completely transparent to the infrared radiation from the inside. For cold climates, the ideal window allows all radiation wavelength from outside except UV and reflects all radiations from the inside.

2.2 Glass double pane glazing

In many countries, the use of double glass glazing, a typical example of which is shown in Figure 3, is virtually standard practice, due to their good thermal and acoustic properties [vi]. A typical glass pane is 4mm thick. Using thicker glass can achieve slightly better thermal insulation benefits and significantly improved acoustic benefits. A typical single glazed window has a U value of 5 or 6 Wm^{-2K-1}. Double glazed windows usually have U values in a range between about 1.5 and 3.5 Wm^{-2K-1}[vii]. The lower the U value the better for reducing winter heat loss and summer heat gain. More than three sheets of glass can be used, but the window system then begins to become impractical due to the increased size, weight and cost. Air, argon, krypton, and xenon gas is used to fill between the glass panes. There is an optimum distance between each glass pane to minimize heat transfer through filled gas. This optimum space is less for xenon relative to air argon and krypton. The three key parameters (U value, SHGC and T_v) for some double glazing with low-E coatings are listed in Table 3 [v]. Low-E coatings give significant benefits of reducing the U value and more details will be presented in section 2.3.3.



Figure 3. Schematic of the conventional double glazed window unit.

TUDIE 5. 0, 17, UTU STOC TO SOTTE CONTINEICIUI GIUSS UUDDIE PUTTE GIUZITIG WITH 10WEL COUTINGS	Table 3. U, Tv,	, and SHGC for some	commercial glass	double pane	glazing with	low-E coatings.
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Layer Structure	U(Wm ⁻² K ⁻¹)	Shgc	Tv
DGU [Clear(4 mm)a/Air(16 mm)/ low-E(4 mm)]	1.4	0.73	0.81
DGU [Clear(4 mm)/Argon gas(16 mm)/ low-E(4 mm)]	1.2	0.73	0.81
DGU [Clear(4 mm)/Argon gas(16 mm)/ low-E(4 mm)]	1.3	0.55	0.70
DGU [Clear(6 mm)/Argon gas(16 mm)/ low-E(6 mm)]	1.3	0.54	0.69

2.3 Conventional glazing materials and coatings

In this part, conventional glazing related materials and coatings are investigated. Clear glass, tinted glass, low-emissivity coatings, and multiplepane glazing are discussed in the following sections. Detailed processing schemes, materials and the state-of-the-art research regarding low-emissivity coatings are discussed as well.

2.3.1 Soda-lime glass

Soda-Lime type float glass is the standard glass utilized in windows, doors, mirrors and most architectural applications. The glass is an inorganic material with an amorphous and non-crystalline structure which results from the fusion of silica with a basic oxide. Soda-lime glass is produced through the floating process. This glass is mainly composed of 71–75% by weight silica SiO₂, 12–16% sodium oxide Na₂O from Na₂CO₃ known as soda, and 10–15% lime CaO, and a small amount of other materials that are employed for adding special properties such as colouring. This glass can be enhanced with functional coatings as well.

The transmittance of clear glass for visible and near-infrared (NIR) spectrum is about 0.75–0.92 at 4 mm thickness. Table 4 shows visible transmittance and SHGC for a commercial low iron content float glass. Note that low iron content increases the visible transmittance of float glass.

Table 4. Visible transmittance	and solar heat gain	coefficient of clear	glass for various thicknesses.
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Thickness (mm)	T _v	SHGC
4	0.91	0.88
6	0.90	0.87
8	0.89	0.85

2.3.2 Tinted glass

Tinted glass is a clear glass with some metal components added during the floating process. The ratio of these metallic colorants affect the colour and optical properties of the glass. Tinted glass reduces solar transmittance while decreasing visibility and colour. In addition, it warms up and reradiates the heat inside. Several tinted glasses are shown in Figure 4.



Figure 4. Tinted glass.

The visible transmittance depends on the glass colour and thickness. Tinted glass specifications for various colours and thicknesses are summarized in Table 5.

Colour	Thickness (mm)	U(Wm ⁻² K ⁻¹)	SHGC	Tv
Bronze	4	5.8	0.68	0.60
	6	5.7	0.6	0.49
Green	4	5.8	0.63	0.40
	6	5.7	0.55	0.33
Gray	4	5.8	0.66	0.55
	6	5.7	0.57	0.43

Table 5.	Tinted	glass	specifications.

2.3.3 Low-emissivity and solar control coatings

Emissivity is the heat radiated by a material relative to a blackbody (blackbody emissivity is 1). Emissivity value ranges from 0 to 1. Upon solar energy absorption by the glass, its temperature increases consequently and the glass will exchange heat with the environment. Glass can reradiate the heat inside the

building and affect building temperature. This unwanted heat exchange can be avoided using low-emissivity (low-E) coatings. In addition, low-E coatings are spectrally selective. It means that their optical properties are tailored to have high reflectance in the infrared region (wavelength 3–50 μ m) and high visible light transmittance while maintaining very low emissivity. Figure 5 [v] schematically illustrates a window with a spectrally selective coating. Low-E coatings minimize heat loss by reflecting back thermal energy which is in the form of long wavelength infrared radiation back into a building and trap the heat inside. This is favourable for cold conditions where a high solar gain is required while internal heat loss should be minimized. Glazing with low-E with various SHGC is depicted in Figure 6 [v].



Figure 5. Transmission and reflectance of (a) Soda-lime glass (b) Glass with a spectrally selective coating.



Figure 6. Spectral transmittance curves for windows with low-E coatings.

Materials used in low-E and solar control coatings are typically doped metal oxides and metal films sandwiched between dielectric layers. Doped metal oxides are mostly used for low-E coatings fabrication while metallic film stacks are more versatile and can be designed to achieve low-E or solar control coatings.

Deposition technique	Layer structure (nm)	Transmittance
PVD (WO3, ZnS), Electron beam evaporation (TiO2)	TiO2(40)/Ag(20)/TiO2(40)/glass	0.75(500 nm), 0.16 (1200 nm)
	ZnS(31)/Ag(20)/ZnS(34)/glass	0.8(500 nm), 0.1 (1200 nm)
	WO3(37)/Ag(20)/WO3(37)/glass	0.825(550 nm), 0.15 (1200 nm)
DC reactive magnetron sputtering	TiO2(38)/TiN(56)/TiO2(38)/glass	0.48(600 nm)
RF plasma enhanced chemical vapor deposition (DLC), RF magnetron sputtering	DLC(52)/Ag(11)/DLC(50)/glass	0.7(500 nm)
Atmospheric pressure chemical vapor deposition	[(SnO2:F+void)(42.1)/ SnO2:F (334.4)/ SiCxOy(63.5)/ SiCxOy: Na+(16.4)/glass]	0.80(625 nm) 0.60(1250 nm) 0.16(2000 nm)
RF magnetron sputtering (TiNx), DC magnetron sputtering (Ag (Mo))	[TiNx(10) /Ag(Cr)(15)/ TiNx(10)/ glass]	0.52(500 nm) 0.13(1250 nm)
Magnetron sputtering	[Si3N4(22)/SnO2(13)/NiCrOx(1.1)/ Ag(12.2)/ZnO(4.6)/NiCrOx(1.1)/ TiO2(16.5)/Si3N4(12)/glass]	0.81(550 nm)
Electron beam evaporation	NiO(30)/Ag(20)/fused silica and tantalum	0.69(554 nm)
Thermal evaporation	MoO3(40)/Ag(22)/fused silica and tantalum	0.78(492 nm)
RF magnetron sputtering (TiNx, Si, TiO2), DC magnetron sput- tering (Ag(Cr))	TiO2(30)/Si(3)/Ag(Cr)(20)/TiNx(10)/ soda-lime glass	0.88(550 nm)
RF magnetron sputtering (TiO2), DC magnetron sputtering (Cu)	TiO2(25)/Cu(20)/TiO2/borosilicate float glass	0.82(600 nm)

Table 6. Literature data on solar control and low-E coatings materials, deposition techniques, and performance.



Figure 7. Position of low-E coating .

Low-E coatings are sensitive to weather and pollutants making it difficult to clean without damaging the surface. For double glazing, low-E coatings are placed at on one or two of the interior surfaces of an insulating glass unit (#2 and/or #3 Surface, counted from the exterior surface on the outer glass pane), as shown in Figure 7. The u value of a Low-E insulating unit is the same whether the Low-E coating is on the #2 or #3 surface. However, orienting the Low-E coating to the #2 surface reduces solar heat gain and cooling costs. In double glazed windows low-e glass can be specified on a particular side of the glass depending on what you want to achieve. In cold climate, the low e coating shall be placed on surface #3 to get higher solar coefficient and sunlight, whereas in hot climate, low-E coating shall be on surface #2 to avoid sunlight.

3 Review of Existing and Emerging Plastics and Their Application in Double-Glazed Windows

In this section, six plastics (PMMA, polycarbonate, ETFE film, PET, polypropylene and Tritan) are be reviewed. Their advantages and limitations for the application of glazing are analysed.

3.1 PMMA

PMMA (also called acrylic or perspex) is a non-toxic, inexpensive, strong and lightweight material. PMMA with different molecular weights (chain length and entanglement) could be obtained and processed easily. It is a clear, colorless polymer with a glass transition temperature range of 100°C to 130°C [viii]. This polymer melts at 130°C, with a moisture absorption of 0.3%. It has very good thermal stability, and is known to withstand temperatures as high as 100°C and as low as -70°C. The ignition temperature ranges between 250°C and 400°C [ix]. The combustion productions consists of carbon dioxide, water, carbon monoxide and low-molecular-weight compounds, including formaldehyde [x]. The density of acrylic ranged between 1.17-1.20 a/cm³ which is half than that of alass[xi]. PMMA can transmit up to 92% of visible light at 3 mm of thickness. It has refractive index of 1.49 at 589-nm wavelength and filters ultraviolet (UV) light at wavelengths below about 300 nm, similar to ordinary window glass. The influence of the near-UV from solar radiation on PMMA is minor, because PMMA is only sensitive to high energy UV radiation with wavelengths shorter than 300 nm [xii]. The impact strength of PMMA is greater than of glass: it does not shatter upon rupture, and is one of the hardest thermoplastics with high scratch resistance [viii]. Its compressive strength is greater than 120 MPa, which is significantly beyond the minimum compression strength (69 MPa for tempered glass).

Due to its good transparency, mechanical properties and durability, PMMA sheets have been used to replace glass as structural material for construction of transparent walls and tunnels in land-based aquaria and shore-based underwater walkways, hotels, and laboratories where the occupants can, in comfort, observe sea life beyond the acrylic walls of the structure. Much of the literature examining the durability of PMMA is more than 20 years old [xiii].

However, due to its flammability and poor fire resistance, it is less often used in other building types due to fire incidents, such as the Summerland disaster, where 50 people were killed [xiv]. The Summerland was a large leisure center with 4600 m². Its street frontage and part of the roof were clad in transparent acrylic glass sheeting. As the fire spread quickly, the acrylic material melted, allowing more oxygen to enter, and dropping burning molten material, both starting other fires and injuring those people trying to escape. The fire rating of PMMA is B2 according to the German standard (DIN4102), meaning flammable; correspondingly, it is rated E in the European Fire classification, meaning high contribution to fire spread.

3.1.1 Plastic secondary glazing

Smith et al. from Victoria University of Wellington, New Zealand, have investigated the thermal performance of plastic secondary glazing as a retrofit alternative for single-glazed windows [xv]. Installation of secondary glazing (or storm windows) can involve the internal addition of a second pane of glass or plastic to existing single-glazed windows to improve the thermal performance of windows during the heating season, without the need to remove the existing glazing.

Four secondary products available on the local market were tested in laboratory by retrofitting each successively to the single-glazed, which was an aluminum-framed (non-thermally broken) and fixed-light window, as shown in Figure 8. The details of the product configuration and the measured U and R values are presented in Figure 7. The R value represented the thermal resistance of the glazing system. It was found the three systems (thin plastic film, magnetically attached acrylic sheet and aluminum-framed clear glass) to all have similar thermal performance (0.35 \pm 0.05 m²K/W), with the low-E aluminum-framed glass system providing additional benefit, due to the effect of the low-E surface (0.57 \pm 0.08 m²K/W). The lowest-cost solution, the thin plastic film secondary glazing, provided comparable R-values to the more expensive solutions based on plastic or glass sheets, but is a temporary solution.

Although the analysis focused on the thermal performance improvement over a base case single glazed window system with an R-value of $0.15 \pm 0.05 \text{ m}^2\text{K/W}$, it is possible that home occupants will receive other benefits from the installation of secondary-glazing systems such as reduced air infiltration and condensation. Where large areas of single glazing are found in homes, these systems offer a realistic, low cost method to reduce space heating energy use by improving the thermal performance of the window system.



Figure 8. The tested glazing systems. Dark blue is the aluminium frame, light blue is the glazing, green is the air cavity, and orange is the timber sill/reveal. The different secondary glazing systems are added as a layer to the right hand side. (a) Base single glazed aluminium framed window with 4 mm thick clear glass, (b) base system plus magnetically attached acrylic, (c) base system plus plastic film and (d) base system plus clear glass in an additional aluminium frame.

Glazing system	Thickness (mm))	U(Wm ⁻² K ⁻¹)	R(m ² k/W)	
	External glass	Airgap	Internal glazing		
Base window	4.0	-	-	6.9 ± 2	0.15 ± 0.05
Base + magnetic acrylic	4.0	48.5	3.0	2.8 ± 0.4	0.36 ± 0.05
Base + thin plastic film	4.0	88.1	0.2	2.9 ± 0.4	0.35 ± 0.05
Base + clear glass	4.0	61.4	6.0	3.0 ± 0.5	0.34 ± 0.05
Base + low-E glass	4.0	61.4	6.0	1.8 ± 0.3	0.57 ± 0.08

Table 7. Results from measurements in the guarded hot box (GHB)

3.2 Polycarbonate

Polycarbonate is a high-performance tough, amorphous and transparent thermoplastic polymer with organic functional groups linked together by carbonate groups (-O-(C=O)-O-). When considering glazing solutions, polycarbonate glazing materials have sometimes become a preferred choice for architects and building owners. This is mainly because polycarbonate has good compressive strength, and has over 250 times stronger impact strength. It maintains approximately equal optical transparency. In addition, lightweight polycarbonate glazing solutions are easier to handle and install. Moreover, polycarbonate can be curved, formed, and fabricated into nearly any geometric configuration that a designer might imagine. Lastly, it offers fair resistance to UV light and weatherability.

Glazing solutions using polycarbonates have been widely applied in various areas, such as acoustic barriers along highways, bulletproof dividers in banks and retail outlets, skylights in commercial/industrial/residential buildings, aircraft windows, race car/boat windows and shatterproof shed windows, etc., as shown in Figure 9.

Polycarbonate has a classification of s1, which means that the speed or quantity of smoke is absent or very low, per European fire classifications (EN 13501-1). This is significant as Stanford Medicine reports that smoke accounts for approximately 70 percent of fire victim deaths. In addition, polycarbonate produces very little flaming droplets, which contribute to the spread of fire by igniting other surfaces, and therefore receives a Bd0 rating. Overall, polycarbonate is a flame retardant materials with good fire resistance behaviors.



Figure 9. Polycarbonate glazing.

Polycarbonate, even though being resistant to high impact, is susceptible to scratching; it is rated 2B on Pencil Hardness Scale, meaning that a scratch can be made using a 2B grade pencil. Moreover, polycarbonate is susceptible to photodegradation under ultraviolet (UV) radiation, which causes breakdown of the material, leading to yellowing, loss of toughness and embrittlement [xvi]. Therefore, a hard coat with UV protection is often applied on polycarbonate.

Both PC and PMMA are clear and shatter-resistance alternatives to glass. PMMA is often used as a reasonable substitute to polycarbonate (PC) thanks to its cost-effectiveness and when extreme strength or flame retardancy is not essential. Also, PMMA is less likely to scratch and does not yellow over a period of time. The other benefits of PMMA includes higher transmissivity and better optical clarity which can be also be restored by polishing.

3.3 ETFE film

ETFE is a fluoropolymer, or a polymer (a macromolecule consisting of a chain of the same molecules) that contains fluorine atoms. It is density is around 1.7 g/cm³, and has the following distinguished advantages compared with other polymers:

- Withstands temperatures over 200 °C
- Light transmission is higher than 90%
- Chemical and soil resistant
- Easily cleaned simply by wiping surface
- Superior tear strength
- Absorbs noise

- Non-sticking
- Long-term weatherability
- Nonflammable

ETFE films are used around the world because they exhibit exceptional transparency, antifouling properties and durability. It is usually used in buildings in the form of roofing and architectural façade (e.g., air pillows). The superior transmission of UV and long wave radiation also leads to it being used extensively in outdoor constructions, for example, green houses. There are several well-known buildings that have used ETFE in their construction, including:



The outer shell of the Water Cube (the Beijing Olympic Swimming Stadium) consists of 3065 ETFE air pillows covering an area of 100,000 m², and the membrane material has an area of 260,000 m².

ETFE has excellent thermal performance and is universally recognized as an excellent building material from the point of view of fire behavior. The material is combustible but not very flammable. Thanks to the melting temperature of around 275 °C, the film is destined to break and shrink before the high temperatures can lead to the collapse of the main structure of the structure. In the event of a fire ETFE does not promote the spread of flames because it self-extinguishes and does not generate any flaming droplets or particles. Therefore, it is rated Bs1d0 in European fire classification, meaning ETFE is a combustible material with a very limited contribution to fire with a weak quantity/speed of smoke emission and no dripping of flaming droplets.

While no ETFE structures have been in place for long enough to gain a true understanding of the life cycle of the foil, the material has been extensively researched and tested in a laboratory environment and out in the field. These tests have concluded that no degradation or loss of strength has occurred and there is no sign that the material will become brittle or discolour over time. As a result, it is anticipated that the material has a life expectancy in excess of 50 years.

Despite of its extraordinary durability and reasonable flammability, there are two major issues for ETFE to be applied for glazing which serves as the visual connection between indoor and outdoor environment. First, the intrinsic haze level is around 5% for 0.3 mm thick film; the haze level is expected to be much higher when the thickness increases to 4 mm thick, which is the thickness of glass panes of conventional double glazing, resulting in the milky appearance, as shown in the top story of the KCPL Facades, Kansas City, US, in Figure 10 [xvii]. The second issue is its high transmission (80% - 88%) across the ultraviolet range (300 - 380nm), as shown in Figure 11 [xviii]. Though this characteristic allows plants and vegetation underneath to thrive in ETFE green houses, it is not desirable for glazing solutions in office and residential buildings. Particularly, ETFE glazing allows UVB (280 – 315 nm) enter interior spaces and constant UVB exposure can enhances skin ageing and significantly promotes the development of skin cancer.



Figure 10. ETFE glazing for the KCPL Facades, Kansas City, US.



Figure 11. Transmission spectrum of ETFE compared with glass and polycarbonate.

3.4 PET

Polyethylene terephthalate (PET) is a chemically stable polyester and its use has risen dramatically in the last few decades with a multitude of applications, ranging from food and drink containers to the manufacture of electronic components. PET has a glass transition temperature of 67–81°C and melting point of 260°C. It is commonly used in industrial applications due to excellent moisture and fair oxygen barrier characteristics [xix]. It is a naturally translucent and semi-crystalline plastic. Some of its most important characteristics include its resistance to water, its high strength to weight ratio, the fact that it is virtually shatterproof (it won't break like glass packaging), and its wide availability as an economic and recyclable plastic (resin identification code "1").

PET exists both as an amorphous (transparent) and as a semicrystalline (opaque and white) thermoplastic material. The crystallinity of PET is an important property to measure, as it influences its mechanical and optical properties. The semicrystalline PET has good strength, ductility, stiffness, and hardness. The amorphous PET has better ductility but less stiffness and hardness [xx]. Semi-crystalline or crystalline polymers are translucent or opaque as there are differences in refractive indices between the crystalline regions and amorphous regions

There are three concerns for PET to be applied in windows. First, there might be haze because we need to optimize the crystallinity which is a compromise between the optical transparency and mechanical properties. Second, it has low heat deflection temperature which is around 70 °C, which is easily reached on an object's surface in a hot summer. Third, it has rather poor UV resistance (as shown in Figure 12), making it not very suitable for outdoor application.

3.5 Polypropylene (PP)

Polypropylene (PP) is regarded as one of the lightest and most versatile polymers due to its multiple advantages: low cost, easy processing, great recyclability, good resistance to many acids, alkalis and solvents, low density, high stiffness, good balance of impact strength vs. rigidity, good heat resistance [xxi]. It exists in three different tactic forms: isotactic polypropylene has a 30 – 60% degree of crystallinity, syndiotactic polypropylene which is less crystalline, and atactic PP which is amorphous. Commercial PP is typically a mixture of 75% isotactic and 25% atactic. Atactic polypropylene is a soft, rubbery polymer, while isotactic polypropylene is strong and hard with excellent resistance to stress, cracking, and chemical reaction. Syndiotactic polypropylene has only recently been made on a large scale [xii]. Crystallinity affects optical transparency because of the scattering taking place when light passes from amorphous to crystalline regions: spherulites in i-PP are much larger than the wavelength of visible light (0.4– 0.7μ m), and the refractive index of crystalline regions is higher than that of amorphous regions; as light rays pass from amorphous to crystalline regions, they encounter the large spherulites, resulting in light scattering; as a result, optical transparency is lower, and haze is produced. Due to their noncrystalline structure, amorphous materials have better optical transparency than semicrystalline materials, and a decrease in crystallinity of a semicrystalline polymer enhances the clarity. However, excessive reductions in crystallinity can result in unacceptable reductions in strength, stiffness, and resistance to softening, so a compromise must be reached that is appropriate for the application [xxiii].

There are two main issues for PP to be applied for windows. First, it develops brittleness and therefore has poor resistance to bending, breaking, crushing when the temperature is as low as 0°C. Second, during exposure to

weathering, such as UV, oxygen, pollution, humidity and heat, polypropylene undergoes changes in its main chain that lead to the start of the PP's oxidative degradation and later to the crazing and cracking that deepen and become more severe over time [xxiv].

3.6 Tritan

Copolyesters [xxv] are produced when more than one glycol and/or more than one dibasic acid are used during the synthesis of polyesters. Due to the presence of these comonomers, the chains of copolyesters tend to be less regular than those of the homopolyesters and thus have a lower tendency to crystallize. As a result, copolyesters can be amorphous or crystalline depending on the types and concentration of comonomers used.

Developed by Eastman Chemical Company and launched in 2007, Eastman Tritan is a new-generation copolyester that retains the versatility of a copolyester but also offers additional advantages including easier processing, higher temperature resistance, and significantly higher impact resistance [xxvi]. Tritan copolyesters are prepared by the melt phase copolymerization of 2,2,4,4-tetramethyl-1,3-cyclobutanediol (TMCD) and 1.4 cyclohexanedimethanol (CHDM) with dimethyl terephthalate (DMT). Important material variables include the mol% of TMCD present in the copolyester and the inherent viscosity (IV) of the copolyester. Due to their similarity in toughness with additional dishwasher durability and freedom from consumer concerns related to BPA, Tritan copolyesters allow Eastman compete in lucrative polycarbonate plastic markets.

The T_g of most traditional copolyesters is limited to around 80 °C while the T_g of Tritan TX2001, a typical grade designed for houseware application, is about 120 °C. Tritan copolyesters crystallize very slowly relative to polyesters such as PET, and are easy to mold into amorphous parts that are transparent even in thick sections and have mold shrinkage of only 0.5–0.7%. Compared with many commercial transparent TP, Tritan copolyesters have excellent clarity and transparency with water-like, neutral color.

Dishwasher durability is a very important attribute for housewares applications. The hot and caustic environment of the dishwasher renders it one of the most demanding tests for plastics. Temperatures in residential dishwashers can reach up to 75 °C, while those in commercial dishwashers can reach up to 90 °C. A pH level of 11.8 has been recorded in residential dishwashers, and detergents used in commercial dishwashers may be more aggressive. While the commercial dishwasher cycle usually lasts only a few minutes, residential dishwashing cycles can last 1.5–2 h.

Currently, as listed in its website [xxvii], Eastman has developed several series of Tristan copolyesters to serve the markets in appliance, bulk water, cosmetics, electronics, housewares, infant cares, in-mold decoration, medical, ophthalmic, signs, smart cards, visual merchandising, water bottles, and water filtration.

Despite of its attractive properties and versatility, unprotected Tritan is not immune to the effects of damaging UV rays. Figure 12 shows the effect of UV

sunlight on PET, polycarbonate, and Eastman TritanTM copolyester. Virgin Tritan has poor resistance to UV radiation: Delta E colour difference is 2 times higher than that of PC at 4000 KJ/m² (roughly equivalent to 40 – 240 weeks). The impact resistance of Tritan suffers dramatic loss when the accumulated UV expose is around 500 KJ/m² (equivalent to 5 – 30 weeks). These data were obtained on specimens cut from extruded sheet and tested on Atlas Ci65A xenon arc accelerated weathering machines according to ASTM Practice G155 cycle 1 and ASTM D2565 using borosilicate inner and outer filters. Samples were removed periodically and tested for Delta E color difference (ASTM D2244) and impact strength (ASTM D6395). The "% retention of toughness" was calculated as the impact strength of the exposed sample divided by the impact strength of the unexposed sample. Each 500 kJ/m²/nm@340nm of machine exposure might be roughly equivalent to 5–30 weeks of actual outdoor exposure, depending on many factors, including time of year, degree of sunshine, global location, and conditions of use.







Figure 13. Weathering test results of Tritan with a UV cap layer.

Eastman has various UV absorbers that can help stabilize Spectar and Tritan in certain conditions. Customers may request UV-protected Spectar and Tritan sheet from the suppliers. As seen in Figure 13, Tritan with a UV cap layer exhibits low color shift and its impact resistance is reduced by 20% - 30% when the UV exposure is equivalent to 6 - 8 years in the sun (the general rule of thumb is that

1500 kJ of exposure is usually equated to about one year in exterior environments).

In addition to its UV resistance, another concern of Tritan for glazing solutions is its mechanical properties. The compressive strength should be equal to or higher than that of tempered glass (69 MPa); so far, Tritan's compressive strength is not disclosed on Eastman's website. Moreover, its Young's modulus, which is the measure of stiffness, is 1.55 GPa, and it is half the value of PMMA's Young's modulus. Whether Tritan's compressive strength and Young's modulus are good enough or not, requires more experimental tests to decide.

4 R&D Gaps and Opportunities for Plastic Double Glazing

In this section, first a simple format for plastics assessment and selection will be presented. The R&D gaps and general solutions for PMMA applied to building glazing are briefly discussed, followed by an assessment of the R&D gaps and opportunities of polycarbonate glazing, mainly including the development of hard coatings with UV protection and low-E coatings. At the end, the general principles, steps and methods for reliable service life prediction on the polycarbonate glazing will be introduced.

4.1 Simple format for plastics assessment and selection

Table 8 presents a simple format for the assessment and selection of plastics for building glazing. The critical parameters are listed along with the corresponding benchmarks. For polycarbonate and Tritan, the service life is for UV-protected protected, whereas the service life of PMMA is for virgin products. It should be noted that, the service life depends on where the plastics are applied; for example, even if identical polycarbonate sheets may be applied, the service life of polycarbonate automotive glazing would be shorter than polycarbonate greenhouse glazing, due to the different failure standards: automotive glazing has more stringent requirements than greenhouse glazing. Generally speaking, UV-protected or hard-coated polycarbonate sheets have a 10-year warranty, whereas virgin PMMA sheets have a 30-year warranty.

Virgin polycarbonate has low scratch resistance and suffers UV degradation, which are the critical factors on its service life. Virgin PMMA has good UV resistance and its service life satisfies the requirement of 25-year payback period; however, similar to polycarbonate, PMMA's scratch resistance needs to be improved; more critically, it has high contribution to fire spread, and this would significantly limit its application in buildings. Regarding Tritan, the information on its compressive strength and scratch resistance were not disclosed. In any case, due to its rather short service life, Tritan is not suitable for building glazing, even if with UV protection.

	Polycarbonate	РММА	Tritan	Tempered glass
Clarity	\checkmark	\checkmark	\checkmark	\checkmark
Stiffness (Young's modulus)	3.17 GPa	3 GPa	1.55 GPa	48 GPa
Compressive strength (Benchmark: 69 Mpa)	\checkmark	\checkmark	unknown	> 69 MPa
UV resistance (Virgin)	Fair	Good	Poor	Good
Fire resistance (Benchmark: European fire classifications - B)	\checkmark	×	1	\checkmark
Scratch resistance (Benchmark: Pencil Hardness Scale - 4H)	×	×	unknown	\checkmark
Heat deflection temperature at 0.46 MPa (benchmark: 80 °C)	\checkmark	\checkmark	1	\checkmark
Life time (benchmark: 25-years payback period)*	X 10 – 20 years	\checkmark	X 5 – 10 years	\checkmark

Table 8. Table with critical parameters and benchmarks for plastics assessment and selection.

The estimated lifetime is for UV-protected PC and Tritan, Virgin PMMA

PC (UV-protected or hard coated) warranty: 10 years, PC automotive glazing: ~ 10 years, PC green house: 15-20 years

More details regarding the mechanical, optical, thermal and other properties of the plastics mentioned in the previous sessions have been in summarized in the appendix 6.1.

4.2 PMMA: improving flame retardant properties

Because of its poor thermal stability, flammability, serious tendency to drip upon melting, and production of a large amount of smoke, PMMA's application prospects have been limited in many fields. The methyl methacrylate (MMA) monomer, which is formed when PMMA burns, will decompose into many small molecules consisting of combustible materials such as methane, methanol, propylene, and acetone and release a large amount of heat, which promotes the combustion of the material.

There are two types of approaches have been quite well accepted and commonly to improve PMMA's flame retardant (FR) properties [xxviii, xxix, xxx]. The first common approach involves mechanical incorporation of flame retardant additives into the bulk polymeric matrix, which is mostly low cost and fast blending technique. However, the concentration of FR needed to be effective is usually too high, which can lead to a significant influence on the mechanical and optical properties of the materials. The second way to reduce the flammability of the matrix is to bind units chemically to it by using FR segments that contain functional groups. Through this approach the FR element becomes an integral part of the polymer chain and usually results in higher efficiency and longer durability of FR effect. Such incorporation could change the morphology and physical properties of the bulk polymer, such as melting point, density and glass-transition temperature and presents relatively higher difficulties in industrial manufacturing for certain materials. Moreover, similar to the problem of first approach, the chemical modification can have significant impact on the transparency and colour of a polymer, as shown in Figure 14 published in 2019 [xxx]. In the work for Figure 14, an acrylate derivative was synthesized and introduced into the main chain of MMA by free radical copolymerization to form a PMMA copolymer (PMMA-co-BDPA). The sample of PMMA-co-BDPA-15 passed the UL-94 (the Standard for Safety of Flammability of Plastic Materials for Parts in Devices and Appliances testing) V-1 rating [xxxi], i.e., showed meaningful improvement of flame retardant properties for building glazing. However, it also showed the unwanted yellowness which is not acceptable for building glazing for visual connection.

Tremendous research is required to improve the flame retardant properties of PMMA and meanwhile retain its mechanical as well as optical properties. An alternative is to define the scenarios where flame retardant glazing is not essential. The definitions involve multiple variables and uncertainties, such as location, functions of buildings, numbers of floors, mobility of people, etc. Fire safety has become a priority for the building construction industry. Rigorous definition of scenarios is not in the scope of the current project and should be done by experts in fire risk assessments. Therefore, in the following sections, we will focus on the development of polycarbonate glazing, which do not suffer from these adverse risk factors.



Figure 14. Ultraviolet-visible spectroscopy (UV-vis) spectra and digital photos of poly(methyl methacrylate) (PMMA) and copolymers.

4.3 Development of polycarbonate glazing

Polycarbonate (PC) has been chosen as good substitute for heavy glass in glazing, optical lenses, and electronic devices due to its excellent transparency. Compared to other transparent plastics, such as polyethylene terephthalate (PET) and poly(methyl methacrylate) (PMMA), PC possesses excellent toughness and heat capability. However, polycarbonate is susceptible to mechanical damages by external forces (mainly scratches) and degradation upon long-term exposure to UV light, transforming into opaque or yellowish appearances. Therefore, assurance of surface hardness and durability is a prerequisite for building glazing. Although intrinsic properties of PC can be improved by modifying the chemical constituents in a monomeric unit, the structural design is rather complicated and is not economical for mass production [xxxii]. Another approach is based on the use of protective coatings. The coatings not only compensate for the soft surface of the underlying PC but also absorb UV light to prohibit photodegradation, resulting in an extended lifetime.

4.3.1 Hard coating

Surface hardness is one of the fundamental requirements for polycarbonate to maintain the long-term stability of optical clarity. Upon expanding the

application fields, harder PCs with excellent durability are in demand. There are two types of hard coatings: an ultraviolet curing type or a thermal curing type. Each type of hard coating can be used in accordance to the required specifications. For outdoor use, thermal curing type hard coating is commonly used because it has better weathering resistance than the ultraviolet curing type

Selection of appropriate coating techniques is of crucial importance to satisfy the desirable properties. There are several techniques for the modification of the PC surface, including liquid-driven chemical coating, physical vapor deposition (PVD), and chemical vapor deposition (CVD). Chemical coating is suitable for the versatile substrates covering metals, ceramics, and polymers. When PC is coated with liquid precursors, uniform coating on the PC substrates with complicated geometry can be achieved over a large area with dipping or spraying methods. Vacuum deposition, on the other hand, is a simple and environmentally clean process that involves inorganic substances being directly deposited on the PC substrates under vacuum.

A problem frequently encountered with polymer substrates is poor interfacial adhesion of deposited films due to the lack of chemical bonds and different thermal expansion coefficients [xxxii]. Polycarbonate features a limited chemical affinity and poor wettability with most coating materials. Weakly bonded films on the PC substrate tend to be delaminated when the flexible PC is deformed. Therefore, numerous studies of coatings have concentrated on identifying the optimal sequence of surface pre-treatment steps and deposition conditions.

In general, hard coatings consist of organic-inorganic hybrid or pure inorganic materials. The hybrid sol-gel approach is one of the most prevalent coatings for PC, where curable organic resins are embedded with inorganic particles by chemical reactions. Upon increasing the inorganic content and the miscibility of dissimilar constituents, the mechanical properties of coatings increase. Nano-sized silica particles have been preferably chosen in the coating industries because they reveal better levels of optical, thermal, and mechanical properties [xxxii]. The Si-O-Si inorganic backbone in combination with the level of crosslinking of acrylate groups is responsible for higher scratch resistance. Pencil hardness of PC increases up to 5H by loading 40 wt% of nanosilica without deteriorating adhesion strength between the coating and substrate. However, it is difficult to embed pure silicas into organic substances because of their hydrophilic nature and the strong adhesion between them. The introduction of functional groups on the silica surface is effective to generate covalent linkages on the interface. Strong interactions between two phases lead to compacted networks, resulting in greater resistances against external forces. The urethane acrylate films reinforced with modified nanosilica particles show higher hardness and elastic moduli at the same filler loading in comparison to the coatings with untreated nanosilica. Hardnesses of silicabased coatings obtained by pencil tests are represented in Table 9.

Hardness of coated layers is easily evaluated by conducting pencil or indentation tests. In the pencil test, as described by ASTM D3363¹, the sharp tip of a pencil with hardness from 9B to 9H is moved over the coating layer at an angle of 45° under a constant load. The pencil scratched coating off from the surface is taken as a measured hardness. In the case of the indentation test, a hard indenter is pressed on substrates and then moved to evaluate hardness and elastic modulus from the load and displacement behaviours. The scratch property of coatings mainly depends on layer thickness, elastic modulus, fracture toughness, and hardness.

Table 9. Pencil and nanoindentation hardness of silica-based hybrid and inorganic coatings on polycarbonate substrates.

Test methods	Coating m	aterials	Thickness (µm)	Hardness	Ref.
Pencil test	hybrid	UA-TMSPM/SiO ₂	15 - 20	3H	xxxiii
		UA/TMPTA/SiO ₂	20 - 30	5H	xxxiv
		GLYMO/TEOS/SiO ₂	25	5H	XXXV
	Inorganic	SiOx	1	4H	xxxvi
		SiOxCy	1.8	6 – 7H	xxxvii
		SiOxCyNz	~ 0.3	7H	xxxviii

UA-TMSPM urethane acrylate 3-(trimethoxy silyl) propyl-methacrylate (TMSPM), TMPTA trimethylol propane triacrylate, GLYMO, 3-glycidoxypropyl trimethoxysilane, TEOS tetraethylorthosilicate, TPGDA tripropylene glycol diacrylate, DPHA Dipentaerythritol hexa-acrylate, HDDA 1,6-hexanediol diacrylate

4.3.2 UV degradation and protection

Degradation from sunlight is the primary concern when plastics are applied outdoor. The photochemical effect of sunlight on a plastic depends on the absorption properties and the bond energies of the plastic. Approximately 5– 6% of the total radiation that reaches the surface of the earth is in the UV region of the spectrum and varies with daily weather conditions. The wavelengths that have the most effect on plastics range from 290 to 400 nm. The wavelength of UV radiation whose photon energy corresponds to a particular bond energy in the polymer chain (as shown in Table 10) can break the chemical bonds (chain scission), changing the properties of the polymer [xxxix].

¹ ASTM is an international standards organization, and ASTM D3363 is the Standard Test Method For Film Hardness By Pencil Test.

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Bond Type	Bond Energy (kJ/mol)	UV Absorption Wavelength (nm)						
C–H	380-420	290						
C–C	340-350	300						
C–O	320-380	320						
C–Cl	300–340	350						
C–N	320-330	400						

Table 10. Bond energy and UV absorption wavelengths for the bond types in plastics.

The UV radiation degradation of the plastic materials used in outdoor applications can be somehow inhibited or delayed by additives. Their concentration range usually employed is 0.05–2%. The concentration is optimized depending on many variables, such as optical clarity, mechanical properties, refractive index, etc. Based on the chemical reactions, there are four types of the additives:

• Absorber

Additives that absorb UV light before it can contact the degradation-initiating centre, and the energy is dissipated as heat. Benzophenones are good general-purpose UV absorbers for clear polyolefin systems, and can also be used in pigmented compounds. Benzotriazoles are used mainly in polystyrene and PVC, but can also be used in acrylics and polycarbonates, and in polyurethanes and unsaturated polyesters. They also improve the light stability of polyacetals, urea, melamines and epoxies.

• Stabilizer

Additives that deactivate the excited state of the initiating centre.

• Quencher

Additives that transform the hydroxyperoxides into other stable forms that do not generate free radicals. Metal complexes of sulfurcontaining compounds are very efficient in this regard and can be effectively used at low concentrations.

• Scavengers

Additives (free radical scavengers) whose purpose is to stop further reaction of the formed free radicals, by simply reacting to them and forming a stable compound. The hindered amine light stabilizers (HALS) most likely operate by this chemical mechanism. The addition of UV absorbers alone may not always give adequate protection to all type of plastics and generally they are therefore used in combination with HALS.

The service time of a UV-protective coating is limited by the initial concentration of UV absorber and the rate at which the absorbance is lost. Moreover, degradation of the coating matrix could always cause earlier

failure. The rate of loss must be determined experimentally for each coating or laminate material by use of free-standing films, coatings on UV-transparent substrates, or cross-sectional analysis methods [xl]. It is important that the data be treated in a way to give a reliable rate constant that does not depend on the initial absorbance of the sample. The practice of plotting A/A_0 as a function of exposure gives particularly misleading rates. Plotting $log_{10}(10^A - 1)$ versus exposure gives consistent rates independent of the initial absorption. The service can be calculated using this rate constant, the initial transmission of the dose of UV (or time of exposure) required to cause a non–UV-blocking coating to delaminate.

4.3.3 A hard coating with UV absorber from TEIJIN LIMITED

The automobile industry has a strong need for weight savings in order to meet the regulations regarding the issue of the global environment. TEIJIN LIMITED started the development of plastic glazing in 1998, not only the functional materials (polycarbonate resin and hard-coating (HC) liquid) but also the processing technology, and succeeded in the industrialization of the plastic glazing production of limited parts in 2006. Since then, mass-produced glazing products are being supplied to automotive manufacturers. To promote business expansion in plastic glazing, TEIJIN LIMITED clarified the deterioration mechanism of plastic glazing used in the outdoors and established the plasma chemical vapor deposition (CVD) coating method. It design and provide "the new driving experience" with our plastic glazing.

A hard coating (HC) for automotive glazing requires superior heat resistance, water resistance and toughness against elastic stress, an HC that could persist throughout the service life of automobiles has not been established until recently (around 2019)[xli]. To be able to blend the maximum amount of a triazine-based UV absorber with superior durability and heat resistance into the HC, the copolymerization composition ratio of the multicomponent copolymer thermoset acrylic resin was uniquely adjusted to optimize their compatibility. By adding inorganic metal particles into a thermoset silicone HC on top of the primer coat, the DMT series that satisfies the above characteristics was developed [xlii]. This line up possesses particularly strong heat resistance compared to that of conventional HC and can even withstand use at 130 °C



Figure 15. Layer structure of Teijin's silicone hard coat DMT series.

The DMT series consists of two layers of HC. The top layer (hard coat layer) is a hard coat formed via the sol-gel method using colloidal silica and alkyl alkoxysilane that add abrasion resistance. The primer layer is formed between the PC and hard coat layer (top layer), and in addition to the function of making these two layers adhere together, UV resistance is added by blending triazine-based UV absorber. In addition, to further increase the weather resistance of the DMT200 series, UVA is used in the top layer to a degree that does not affect the abrasion resistance. By using this kind of proprietary technology, characteristics that can be useful in automotive glazing applications are added. Not only is the DMT series used in HC applications for plastic glazing, but it is also being used in sheet film applications that require weather resistance and abrasion resistance. The characteristics of the DMT series is summarized in Table 11.

The DMT200a series, developed using chemical vapor deposition (CVD) technology for automotive glazing applications, is garnering attention from automotive manufacturers, as it possesses surface hardness on par with glass that can comply with the abrasion resistance required by new safety standards (UNECE regulation 43 on glazing materials) applied to new vehicles from July 2017. In addition, the weather resistance is approximately doubled by creating a dense CVD layer with increased gas barrier performance, which prevents the deterioration of the hard coat layer and PC substrate by oxygen and water.

		Condition/method		DMT200α (CVD)	DMT200 (WET)	European Union Standard (ECE R43, rigid plastic)	American Standard (FMVSS, ANSI AS2)
Optical property - Transparent -PC substrate (5mmt)	Light Transmission	Light source; A	%	>88	>88	>70	>70
	Haze	Light source; A		<0.6	<0.6	-	No requirement (<1% only for AS4a)
Scratch Resistance	Taber abrasion ∆Haze	ASTM D1044; For Exterior CS- 10F, 1000cycle		0.5~1.5	5~7	<2	<2 (both sides)
		ASTM D1044; For Interior CS- 10F, 100 cycle		0.4	0.4	<4	-
	Pencil hardness	JIS K5600		F~H	F~H	-	-
	Boiling water resistance	Dipping 3 h		Excellent	Excellent	-	-
Durability -Appearance - Adhesion	Heat resistance	110 °C, 1000 h		Excellent	Excellent	-	-
	Heat cycle	–30 °C, 1h and 100 °C, 1h interval 15 min each		Excellent	Excellent	-	-
	Weathering (Super Xenon Weather Meter)	JIS K7350-2, 180 W/m ² (Exposure >10 years equivalent)		6500 MJ/m ²	3800 MJ/m ²	500 MJ/m ²	306 MJ/m ²

Panlite[®] is the trademark of Teijin's polycarbonate sheet/film, which has various line-ups from thin films to thick sheets, including improved pencil hardness and hard coating. The main grades of Panlite[®] film/sheet are shown Table 12.

Table 12. Main grades of Panlite[®].



A hard coat loses its viability because of cracking and delamination that occurs due to ultraviolet, oxygen and moisture exposure in actual use environments as well as in accelerated weathering tests. Of these factors, the first focus is on t_{fail}, the time when the HC delaminates, and D_{fail}, the amount of ultraviolet radiation. Using the following formula [xliii], the UVA decrease coefficient k is obtained (T₀ is initial light transmittance).

$$t_{\text{fail}} = \frac{1}{k} \log_{10} \left[\frac{10kD_{\text{fail}} + T_0 - 1}{T_0} \right]$$

Using the UVA decrease coefficient in the primer layer of DMT250 aged in a xenon weather meter until delamination and of DMT250 after three years of outdoor exposure testing in Arizona, a predicted lifespan of DMT250 in Arizona of ~5 years was derived. The predicted lifespan of DMT200, with its improved

weather resistance compared to that of DMT250, was approximately 10 years using the same evaluation method. The validity of these predicted values is currently being examined.

In 2017, Teijin announced that it has developed the world's first PC pillar-less automotive front window for use in the Tommykaira ZZ, a sporty electric vehicle (EV) produced by GLM Co., Ltd., an EV manufacturer launched by Kyoto University. The newly developed PC pillar-less front window has high abrasion resistance and excellent weather resistance and meets new vehicle safety standards applied from July 2017. A weight savings of 36% compared to the combination of a conventional glass window and an A-pillar was realized by making it a pillar-less front window. In addition to contributing to improvements in the running performance of the vehicle, the transparent PC glazingintegrated front window and pillar provide unobstructed sight lines for safer driving and contribute to more enjoyable sightseeing.



Figure 16. Optical property changes of DMT200a during a weathering test. (Xenon weather meter, irradiance of 180 W/m2@300-400 nm, black panel temperature of 63°C, light for 102min, Light+spray for 18 min)



Figure 17. Tommykaira ZZ with Polycarbonate-resin pillar-less front window; conventional glass front window with an A-pillar (left) and a newly developed pillar-less front window (right).

4.3.4 Low-e coating varieties for polycarbonate glazing

There are two types of transparent low-E coatings that dominate in today's market: (1) semiconductive coatings, e.g., ITO (indium tin oxide) and FTO (fluorine-doped tin oxide) and (2) metallic coating. Some common low-E materials are listed in Table 13 for comparison [xliv].

Technical specifications of the transparent conductive coating	Indium oxide (ITO) coatings	Tin oxide (FTO) coatings	Gold layer systems	Silver layer systems
Layer thickness (nm)	>20	>20	>6	>6
Sheet resistance R (Ω)	>8	>8	>5	>1
Light transmittance (%)	>80	>80	>25	>85
Abrasion resistance	Very good	Very good	Good	Good
Chemical resistance	Good	Very good	Adequate	Adequate
Thermal stability to technical parameters	Adequate	Adequate to good	Adequate	Good
Adherence to the glass surface	Very good	Very good	Good	Good
Preferred coating technique for deposition onto flat glass	Sputter process	APCVD Pyrolytic process	Sputter process	Sputter process
Pane thickness for coating (mm)	>0.3	>2	>0.3	>0.3
Planes of coated pane	As uncoated flat glass	Poorer than uncoated flat glass	As uncoated flat glass	As uncoated flat glass

Table 13. The common low-E materials comparison.

Although there are many thin film coating methods available for glass coatings, such as sol-gel, PECVD, ALD, and E-beam evaporation, there are only two major low-E window coating technologies in today's market: sputtered coating and pyrolytic coating, which provide cost-effective, high durability, excellent uniformity on jumbo glass (3 m or more wide glass).

• ITO coatings on polycarbonate

Indium tin oxide (ITO) films, as one of the transparent conducting oxide (TCO) thin films, have been widely investigated and discussed because of their unique optical and electrical properties, such as high electrical conductivity and optical transmittance in the visible region. This has made them suitable for use in optoelectronic devices, light-emitting devices, solar cells, transparent electrodes for flat-panel displays [xlv]. The increasing demand for lightweight and flexible devices has strongly stimulated the investigation of ITO depositions on a large variety of polycarbonate substrates, using different synthesis techniques such as sputtering, evaporation, and sol-gel. Among them, sputtering is one of the most investigated, mainly because of the possibility to get uniform and homogeneous materials on wide-area substrates, hence representing a promising synthetic route for the large-scale production of ITO electrodes [xlv].

In order to achieve the desired electro-optical properties of sputtered ITO thin films, heat treatment higher than 200 °C is usually conducted on the substrates. However, such heat treatment is not applicable when the substrate is a thermoplastic material. One of the effective ways to realize improved optical transmission and electrical conductivity is to use sandwich type structures such as dielectric/metal/dielectric (D/M/D) multilayer films. In this structure, a very thin metal film is embedded as an interlayer to improve the electrical conductivity without deteriorating the transparency of the dielectric material. Metals such as Au, Ag, Cu, Al and their alloys have been extensively studied for interlayer of D/M/D films [xlvii]. Another critical point of ITO on polymers is the poor mechanical adhesion of the resulting electrodes, due to the low surface energy nature of polymer materials together with their pronounced surface roughness. Generally, this drawback is faced by pre-treating the polymer with plasma surface treatments, with the final aim of activating the polymer surface and promoting the adhesion as well. Another solution to such problems is represented by the use of either inorganic and organic buffer layers, which could increment the mechanical adhesion of ITO and prevent, at the same time, oxygen diffusion phenomena coming from the polymer toward the ITO layer, which could be detrimental for its electrical and optical properties.

All the solutions discussed above, even if promising, strongly affect the competitiveness of the deposition process, increasing the overall costs and time production and thus reducing the possibility to easily transfer the ITO deposition process on wide-area substrates for large scale production.

The development of a completely room-temperature deposition process to prepare ITO-coated polycarbonate structures has been investigated for use as lightweight plastic smart windows for buildings and vehicles [xlvi]. Different deposition conditions were used for growing ITO thin films by the radiofrequency magnetron sputtering technique, without using any surface activation pre-treatment of the bare polymer supports nor heating the substrates during the depositions. According to the characterization results, the optimal deposition conditions were selected for growing high-quality ITO thin films with a 90% optical transmittance in the visible range together with a sheet resistance of 8 ohm/sq. The optimized ITO/PC structures were then successfully tested as conductive supports for the fabrication of smart windows based on plastic substrates.

• FTO and other doped metal oxide coatings

Fluorine-doped tin oxide (FTO) is another commercially available semiconductive coating in today's market. The commercial FTO films on glass substrates are usually prepared by pyrolytic method (also called chemical vapor deposition). Their extinction coefficient k is very small (0.01 at 550 nm), so that the typical thickness is of µm scale, whose transmittance of >80% is acceptable. The refractive index is typically around 2. Compared with ITO, FTO shows better environmental and chemical resistance, and therefore it is called hard-low-E coating. Moreover, FTO films are more cost-effective because it does not because they do not contain expensive indium (In) element.

For FTO deposited on polycarbonate substrates, there are far less publications. Low-E coating on plastic substrates usually serve as transparent conductive film for electronics. However, FTO films have a higher surface roughness than that of ITO films. A rough surface complicates electrical contact with the adjacent layer, and can result in poor or erratic electrical transport at the interface, increases the chance of forming electrical shorts, and increases the scattering of light and therefore haze, which lowers optical specular transmission. For these reasons, any lowering of the FTO surface roughness (to make it smoother), without significantly harming its other properties is a research trend to expand the use of FTO to additional optoelectronic applications.

ZnO doped with group-III elements, such as AI and Ga, similar to FTO, is also regarded as an candidate for replacing ITO owing to its promising optical and electrical properties as well as its low cost, nontoxicity, and long-term environmental stability[xlviii]. Several groups have reported AI- or Ga-doped ZnO (AZO) thin films on polycarbonates [xlviii, xlix, I, Ii]

• Metallic coatings on plastic substrates

A multilayer low-emissivity (low-E) coating with oxide-metal-oxide structure on polyethylene terephthalate (PET) and polyethylene (PE) films by magnetron sputtering has been developed by Solovyev, et al. [lii]. The TiO2/ZnO:Ga/Ag/ZnO:Ga/TiO2 coating on the PET film with high waterresistance and capability to be used outside of sealed double-glazed panes was proposed. It was shown that TiO2(10 nm)/ZnO:Ga(20 nm)/Ag(9 nm)/ZnO:Ga(28 nm)/TiO2(20 nm) () is an optimal structure of a multilayer low-E coating on a PET substrate from the point of view of transmittance and reflectance, which were 85% and 91%, respectively. The multilayer low-E coating on a PE substrate with the structure ZnO:Ga(12 nm)/Ag(8 nm)/ZnO:Ga(16 nm) has transmittance over the visible spectrum T550=77% and 91–92% reflectance in the IR wavelength range. The window with a PET film and low-E coating was investigated in terms of heat engineering. It was revealed that heat transfer resistance increased up to 0.73 m2 °C W-1 for the windows with a PET film and low-E coating. Heat transfer resistance of the windows without a polymer film was 0.38 m² °C W⁻¹. The water-resistant ZnO:Ga/Ag/ZnO:Ga/SiO2 coating on a PE film with 77% transmittance and 91– 92% reflection in the IR range was proposed to be used as greenhouse covering material. The possibility of using the PE film with a low-E coating to reduce heat loss in greenhouses and enhance yielding capacity was demonstrated.



Figure 18. Scheme of low-E coatings structure on PET (a) and PE (b) substrates.

• Flexible Low-e coating film

Flexible low-E film is a coated plastic film, available in rolls. The existing low-E films in markets, e.g., 3M[™] Thinsulate[™] Window Film Climate Control Series, can significantly block the amount of harmful UV rays, in addition to seasonal thermal regulation. They also offer high visible light transmission with a neutral appearance. The 3M Company is an American multinational conglomerate corporation with revenue over 30 billion US dollars. Application of this film is less expensive than replacing all that glass. 3M[™] Thinsulate[™] Window Film carries a 15 year warranty when professionally installed on a commercial building. The payback period for the film on single-pane glass is as short as 1.4 years and for double-pane glass is as short as 2.1 years.

Another company that provides reliable solar-control films (e.g., low-E coating films) is Hanita Coatings. Hanita Coatings has been developing solar control and security films for 30 years, and has earned a reputation of being an innovative independent manufacturer of window film products, with a range of energy-efficient interior and exterior solar control films. Two typical series of their exterior films are presented below.

XTRM

XTRM is a new generation of extended-life exterior window films. Thanks to its specially engineered construction and robust polymeric and chemical components, XTRM sets new standards on the long-term durability of outdoor window films. It's been tested and proven in the lab and the field under extreme weathering conditions. Hanita warrants XTRM exterior films for up to 15 years.

SolarZone Xtra

SolarZone Xtra range of window films are special versions of many of their standard SolarZone window films, developed for installation on the exterior of the window, instead of on the interior (room side) of the glass. Exterior films reject solar energy on the outer pane, keeping the inner pane cool and reducing the energy that penetrates inside even more effectively than a similar interior film. For locations in warm climates, solarZone Xtra offers one of the best solutions for keeping interiors cooler in the summer, maximizing savings on energy costs, and upgrading energy efficiency.

4.3.5 Positions of coatings

There might be multiple coatings applied for polycarbonate double glazing and the possible configurations are shown in Figure 19. The #1 surface represents the surface which faces to the exterior side of the outer plastic sheet. In the configurations, low-E coatings are considered on either #2 or #3 surface, depending on the climate. The configurations are meant to be durable, energy-efficient and cost-effective.



Figure 19. Configurations of multiple coatings on double glazing

From a practical perspective, to achieve these requirements, hard we selectively propose the use of hard coat technology on surfaces #1 and #4, and potentially on one of the interior surfaces also (#2 or #3). Of course, one may think of applying low-E coating film on the #1 surface (and likely #4 also) of a double glazing to as hard coat, because low-E coating is hard and provides UV-protection. However, there are four critical factors which would significantly reduce the interest in this regard, and therefore, it is till essential to develop appropriate hard coats for polycarbonate glazing in buildings.

• Narrowing the markets

Solely relying on low-E coating as hard coats instead of developing the conventional hard coats will narrow down the markets. As described in session

2.3.3, Low-E coating can reject large amount of IR radiation into a room, meaning a lower SHGC, reducing energy consumption in warm climates. In cool climate, low-E coating are sitting on surface 3 or 4, to maximize heat coming into a room while minimize heat loss from a room. Solely relying on low-E coatings as hard coats means a low-E coating has to be on surface 1, and this is not desired in cool climates.

• Difficulty in manufacturing

The two main coating categories are Magnetron Sputtering and Pyrolytic Coating. Low-E Magnetron Sputtered Coating (also known as a Soft Coat or Sputter Coat) is designed to go inside the unit and therefore double-glazed in an Insulated Glass Unit (IGU) [liii]. The low-E coatings which are prepared by magnetron sputtering are usually ITO-based or Metallic-based. Pyrolytic Coatings (also known as Hard Coat or Room Side Coat) are a single layer vapour deposit that is fired on at high temperatures during the float glass process and is tough enough to be exposed internally and used on surface 2 of a single glazing or surface 4 of a double glazing. The typical low-E coating prepared by pyrolytic is FTO-based. The high-temperature manufacturing process of relatively cheaper and durable FTO-based coating is not suitable for polycarbonate substrates which require less than 80 °C processing.

Lamination of low-E coating films is an option and requires much less manufacturing effort for glazing suppliers. However, this is a good option for sitting on the surface 2/3/4 but not surface 1, and the reason is stated in the section "Lack of enough durability" below.

• Lack of enough durability

ITO-based coatings are sensitive to weather and pollutants making it difficult to clean without damaging the surface [liv] and are not used on the exterior side of a glazing, as mentioned above. The warranty periods of existing exterior low-E coating films are less than 15 years, which doesn't satisfy the expected service; considering that hard coat with UV protection is a perquisite for the application of polycarbonate glazing, it may significantly reduce customers' interest if the service life of the coating is not long enough and they have to change the film every 15 years.

• More research and potential higher cost required

ITO-based coating and metallic-based low-E coating contains expensive elements. Some of the other commercial low-E coatings are also not colourneutral. Furthermore, the adhesion and the mechanical properties of low-E coatings deposited on polycarbonate remain a challenge due to the low surface energy of polymer materials and low-temperature processing. All these factors suggest that further research is required to achieve durable and lowcost low-E coatings for plastics that fulfill the durability requirements and are more suitable than tailored hard coats with UV absorbers.

4.3.6 Major manufacturers for high-quality polycarbonate sheets

In this section, two major manufacturers of polycarbonate sheets with hard coats are presented.

Bayer AG

Bayer AG is a German multinational pharmaceutical company and one of the largest pharmaceutical companies in the world. Makrolon® is its registered trademark. The Makrolon® trademark stands for a comprehensive range of first-class polycarbonate sheets of constant high quality, based on the use of superior raw materials, an extensive quality management system and production processes certified to EN ISO 9001. Makrolon® solid sheets are a product range of polycarbonate sheets (Table 14) with high transparency and extremely high impact strength that are suitable for many different industrial applications.

• Makrolon® Polycarbonate GP

General purpose polycarbonate sheet, rod and tube for high performance engineering applications that require clarity and high impact resistance.

• Makrolon® Polycarbonate UV2

Manufactured with two sided UV protective coatings, Polycarbonate UV2 offers excellent weathering properties, whilst maintaining high clarity and impact resistance.

• Makrolon® Polycarbonate AR

Polycarbonate Abrasion Resistant (AR) is an impact resistant polycarbonate sheet manufactured with a unique hard coat surface which is applied to both sides, Polycarbonate AR Sheets provide excellent scratch, chemical and graffiti resistant properties.

Makrolon ® Series	Sheet Width	Thickness - mm	Colour	Applications	
Makralan [®] CP	1220 x 2440	1 - 12mm	Clear	General purpose, interior	
Waki Olone GP	1830 x 2440	2 - 12mm	Clear	applications, machine guards	
	1220 x 2440	1 - 12mm		Two sided UV coat, signage.	
Makrolon [®] UV2	1830 x 2440	2 - 12mm	Clear	covered walkways, bus shelters,	
	2050 x 3050	2 - 9.5mm]	sky lights & exterior applications	
	1220 x 2440	4.5, 6, 9.5 & 12mm		Two sided abrasion resistant	
Makrolon® AR	1525 x 2440	6mm	Clear	- High traffic areas, security	
	1830 x 3050	4.5, 6, 9.5 &12mm	1	applications and mass transport	

Table 14. Makronlon polycarbonate sheet product range.

Sabic

Sabic is a Saudi Arabian multinational chemical manufacturing company. Based on high-performance Lexan[™] resin which is a polycarbonate (PC) material, SABIC's engineering plastics solid sheet portfolio helps customers develop lightweight, durable parts with tailor-made performance across a wide variety of industries.

• Lexan[™] MR Polycarbonate

In addition to the inherent characteristics of a polycarbonate material, LEXAN[™] MARGARD[™] has a proprietary hard surface coating, on one or two sides, which provides a high level of resistance to abrasion, chemical resistance and weathering. Depending on the product, they are backed by a

10 Year Limited Warranty against loss of light transmission as well as coating failure and breakage. SABIC has a very diverse range of MARGARD[™] products depending on the application which can add features such as Antifog; for different processing methods offer grades for flat or formability and finally, offer flame-retardant grades.

• Lexan™ UV Resistant Sheet

This virtually unbreakable, transparent Lexan polycarbonate sheet comes with UV protection on one or both sides of the surface and provides remarkable clarity – even after many years of intense sunlight and weather extremes.

4.4 Weathering model and service life prediction

The degree of certainty required for service life prediction (SLP) depends on the consequences of a failure. Risk at one end of the spectrum can be from cosmetic failure resulting in damage to a company's reputation and costs from warranty claims. This type of failure can sometimes cost the company a billion dollars or more, so even this risk is consequential. At the other end of the spectrum is failure resulting in death, serious injury, extensive property loss, or environmental damage. We want to be very certain to avoid these consequences.

4.4.1 General procedures and principles

There are many ways to describe the process for service life prediction (SLP) or the development of predictive test conditions, and most use the same general steps summarized in Figure 20 [Iv]. Excellent predictions may require several person-years of work and a great deal of resources. In any case, this will be a research project and will require some time and effort. In addition, "failure" must be defined and quantified in some reasonable way so that the amount of degradation that can be tolerated is known.

Pre-steps

Define failure Assess risk associated with failure Determine time and resources available Retrieve and analyze aged and failed parts from the field

Step 1: Define the use conditions

Benchmark to a defined environment, e.g., Miami, FL Define environmental variables and their magnitude Define the duty cycle, not just the extremes

Step 2. Find material responses to the environmental stresses

Apply stresses at several levels alone and in combination, if possible Look for underlying processes that can be monitored Develop a rational model that fits the data—ANOVA is not enough

Step 3. Establish reasonable test conditions

Use results from Step 2 to set maximum acceleration conditions Account for all of the acceleration observed Validate with materials that have field data Apply test conditions to variations on materials of the same class

Figure 20. General steps for service life prediction.

Step 1. Identification and Quantification of the Use Conditions

The first step is to define the use environment. For weathering, this might be a benchmark environment, such as south Florida or the Arizona desert, where many samples are exposed at commercial test sites and for which there are abundant environmental data. In any case, the nature, magnitude, and duration of the environmental stresses must be quantified. This can include ultraviolet (UV) radiant exposure, temperature, humidity, dew, rain, chemical pollutants, mechanical stresses, radiation, electrical loads, and biological growth. Not all variables are important for all materials or use conditions, and judgement is required to decide what needs to be included and what does not. In addition, the selection of conditions can depend on the degree of risk. For example, one might choose the 95th percentile of severity when setting a warranty for a coating where the failure is only cosmetic, while one might choose absolute worst case plus a large margin for a safety-critical application.

Step 2. Determining the Response of Materials to the Environmental Variables

Some degradation modes, such as changes in appearance properties (e.g., colour shift, gloss loss or surface roughness) give continuous data that can be used to determine rates of degradation. Other modes, such as cracking and delamination are binary and take a long time to give a single data point. Mechanical property changes often require destructive testing and many

replicates, so very large numbers of specimens are required, making testing under multiple conditions tedious or impracticable. A powerful technique is to find a way to monitor the chemical or physical changes that underlie the property changes of interest. If these can be measured, especially by nondestructive techniques or on very small samples, then the rate of change can be determined long before actual failure occurs. For example, in some cases photochemical destruction of a UV absorber (UVA) in a coating is a prime cause of delamination. The rate of UVA loss can sometimes be determined in a few hundred hours as opposed to thousands of hours before delamination failure [lvi], and the relevant theory will be introduced in section

4.4.2 Prediction of UV absorber permanence and coating lifetime

The maximum lifetime of a coating on a non-weatherable substrate is limited by the initial concentration of UV absorber and the rate at which the absorbance is lost. Degradation of the coating matrix could always cause earlier failure. The rate of loss must be determined experimentally for each coating or laminate material by use of free-standing films, coatings on UVtransparent substrates, or cross-sectional analysis methods. It is important that the data be treated in a way to give a reliable rate constant that does not depend on the initial absorbance of the sample. The practice of plotting A (absorbance)/A₀(initial absorbance) as a function of exposure gives particularly misleading rates. Plotting log₁₀(10^A-1) versus exposure gives consistent rates independent of the initial absorption. The maximum lifetime can be calculated using this rate constant, the initial transmission of the coating at a wavelength where the substrate exhibits high sensitivity, and the dose of UV (or time of exposure) required to cause a non–UV-blocking coating to delaminate. Since these values can be determined long before the coated material exhibits failure, product development cycles can be greatly accelerated. We have applied this methodology to the development and commercialization of several highly weatherable coating systems where accelerated weathering times to failure were 16 months or more and outdoor lifetimes were expected to be >10 years. Customers such as automotive manufactures also are demanding this information to ensure that their coatings will have the desired lifetimes.

4.4.3 Lifetime predictions tests for hardcoated polycarbonate

Service life prediction has been performed by accelerated aging tests for hardcoated (with UV-protection) polycarbonate for the application of automotive glazing, by General Electric [Ivii]. One goal of this work was to determine whether accelerated weathering exposure conditions were likely to be predictive for plasma-deposited abrasion layers on weather- able silicone hardcoats. Secondly, since it was expected that at least one year of accelerated exposure would be required to cause failure, it sought methods for further accelerating the weathering process to determine more quickly if future compositional or process changes might affect the weathering performance of the product.

Analysis of the spectral power distributions of Florida sunlight and xenon arc sources suggests that approximately 3,700 kJ/m² of borosilicate/borosilicate

filtered xenon arc exposure should be the equivalent of one year of Miami, FL sunshine. However, the higher temperature of xenon arc protocols reduces this by about 25% for each 10°C that the test temperature differs from the effective exposure temperature for polycarbonate materials. Empirical data on a wide range of hardcoated polycarbonate show good agreement of xenon arc and Florida results, assuming a correlation of approximately 3000 kJ/m² per year in Florida on clear polycarbonate for the test conditions described in this work.

The amount of exposure required to cause failure is very high, and even at the high irradiance level of 0.75 W/m^2 , test times are >1 year for samples within the specification range. Samples can also be exposed using offsets to move them closer to the lamp, and an acceleration of $1.9 \times$ can be achieved using a 4 in. offset, thereby reducing test times to seven to eight months. Exposure times to microcrack failure can be reduced by applying strain to the samples.

Results from offset and normal position exposure indicate that the delamination of the 2nd generation SHC with or without plasma will occur after approximately 24,000 to 30,000 kJ/m² of exposure at a SHC thickness of 7 to 8 μ m. The conservative correlation factor of approximately 3000 kJ/m² of xenon arc exposure equating to a Florida year gives a predicted lifetime of at least 10 years of direct Florida exposure for coatings with sufficient SHC thickness.

5 Summary and proposal for future development

In this report, we critically assessed a variety of polymer candidates for use as glazing. Of these, PMMA was found to be an excellent materials, with superior longevity than all other polymers considered. However, it's flammability and high smoke-rating make it a questionable candidate for use in many glazing situations. In contrast, polycarbonates do not suffer from these adverse fire risks. In general, the density of polycarbonate is half that of glass, while also exhibiting higher transparency, optical clarity and being cheaper. Moreover, its superior impact resistance (250 times higher than that of glass) can bring distinctive safety benefits, e.g., avoiding vandalization, and can be a very attractive selling point.

However, polycarbonate has a number of pitfalls that prevent it from being used in glazing applications currently. We have identified that there are four aspects requiring research and development in order to achieve high performance and cost-competitive polycarbonate double glazing in buildings.

• Development of durable hard coats with UV protection

Glazing in buildings in standards is expected to have at least 25-year service life. Researchers believe insulating glass units should last in situ for between 20 and 60 years. Based on a survey conducted in the UK, the actual average service life for uPVC glass double glazing is slightly less than the standard value, around 24 years, as shown in

Table 1. At the moment, the service life for polycarbonate automotive glazing is around 10 years, whereas for a polycarbonate greenhouse it is 15 – 20 years. We estimate that the service life of current polycarbonate in glazing applications would be around 15 years, because this application is less stringent than for automotive glazing but more stringent than that for a greenhouse. The short service life of polycarbonate glazing has significantly negative impact on its market competence. Therefore, the development of hard-coated & UV-protected polycarbonate that can last for at least 20 years is essential and a prerequisite for building glazing.

• Development of cost-effective Low-E Coatings for Polycarbonates

Low-emissivity (low-E) technology is an effective solution to save energy in buildings for different climates. While there are many different low-E coating varieties, including on flexible polycarbonate substrates, their application to glazing has been well documented. As such, there is a need to consider which low-E coating variety is most suitable for double-glazed configurations and determine their optical, mechanical and thermal properties.

• Reliable service life prediction

As noted above, service life of any glazing should be at least 25 years. Analysing any polycarbonate double-glazed configuration which includes any accompanying hard coat and low-E coat requires a detailed analysis of the service life prediction. These environmental analyses would predicate deployment of the outlined solutions in the field.

• Energy performance evaluation

Evaluation of the energy performance of polycarbonate double glazing includes the measurement U value and g value, which are essential to present in the data sheet of the relevant products.

5.1 Proposed project and planning

We have formulated a structured research program focussed on addressing the four key aspects outlined above. The tentative actions, schedules and potentially needed equipment are summarized in Figure 21. A more detailed synopsis across each activity area is included below.

Year 1 - planning



Year 2 - planning

	1	2	3	4	5	6	7	8	9	10	11	12
Act.5: Refinement of the formulation of the novel hard coats												
Refine of formulationCharacterization: optical, structural, hardness												
Act.6: Determination of the scalability of the poyel hard coats												
 Improve and adapt the laboratory fabrication process to industrialization. 												
Act.7: Service life prediction • Step 1 - Preliminary service life prediction: prediction of UV absorber permanence • Step 2 - Thorough service life prediction of												
hardcoated polycarbonate												

Needed equipment

- Chemical vapor deposition chamber for the preparation of abrasion-resistant top coat \checkmark
- Spray/dip/spin coater for the preparation of primer layer ✓
- Oven for thermal curing
- Nano indentation device for hardness characterization \checkmark
- pencil test device for hardness characterization (to purchase)
- Xenon Arc Weather-Ometers for accelerated aging tests (to purchase or outsource for testing)
- Test bench for experimental determination of g value (available at EPFL, Switzerland)

Figure 21. Tentative actions, schedules and needed devices for the proposed project.

Year 1 – planning

Activity 1: Structural and compositional designs for novel hard coats

The commercial samples from Teijin, Ibiden, Bayer and Sabic will be requested. Particularly, the structural and compositional characterization will be conducted on Teijin's and Ibiden's samples, as the their hard coats will be taken as the base for the development of our novel hard coats. Then structural and compositional design will be made, and the recipes will be developed for the laboratory fabrication of novel hard coats. Meanwhile, for service life prediction on the developed hard coats, the failures, experimental variables and magnitudes will be defined. The needed experimental will be set up and calibrated.

Activity 2: Development of novel hard coats

The aim of this activity is to develop the novel hard coats that has the desired mechanical and optical properties, and meanwhile has better durability than Teijin's hard coats. For the development of novel hard coats blended with UV absorber, Teijin's hard coats would be taken as the base structure, and then the structures of the multilayer stacks, the thickness, the concentration of UV absorbers, and the technical parameters for coating can be tuned to improve the service life of the novel hard coats. A potential solution is to increase the thickness of the primer layer in Teijin's hard coat is also an alternative.

Activity 3: Development of cost-effective Low-E Coatings for Polycarbonates

For the development of the low-cost low-E coating, commercial low-e coating films will be considered. Since the surface energy of polycarbonate is lower than glass (which may reduce the adhesion of low-e coating films), proper lamination techniques need to be investigated. Low-e coating will also be prepared by magnetron sputtering technique at low temperature, without providing any substrate heating or performing surface-activating pretreatment; the simplified coating procedures can also contribute to cost reduction.

Activity 4: Energy performance evaluation

Polycarbonate double glazed prototypes with different configurations (with and without the novel hard coats, with and without low-E coatings) will be constructed. U value and g value which describe glazing energy performance will be measured and calculated.

Year 2 – planning

Activity 5: Refinement of the formulation of the novel hard coats

The refinement of the formulation of the novel hard coats aim at further improving the durability of the hard coats developed during the first year to a reasonable service life for commercialization. The formulation may also be refined to better adapt the processing procedures at the industrial scale.

Activity 6: Determination of the scalability of the novel hard coats

The scalability of the novel hard coats will be evaluated. The laboratory fabrication process will be adjusted to adapt to the industrial fabrication process.

Activity 7: Service life prediction

The service life prediction includes two steps. The first step is the preliminary prediction on the UV absorber permanence, based on the discussion in section 4.4.2. The second step is the thorough service life prediction of the hardcoated polycarbonate, based on the work done by General Electric (GE) [Ivii]. The test would be done on the commercial hardcoated polycarbonates (as references) and the polycarbonates with the novel hard coats developed in our laboratory. In the work of General Electric, the time taken was 7-8 months for the accelerated aging tests for the prediction of 10-year service life. Therefore, the test time is assumed to be 14 - 16 months for the prediction of 20-year service life. However, the test time may be reduced to 7 - 8 months. The test done by GE was in 2009 and the used power of the arc xenon lamp was 0.75 w/m^2 . However, nowadays the power seemed to be improved by 2 times, meaning the accelerated factor is potential improved by 2 times.

Key Deliverables:

- 1. Durable novel hard coats that can support the commercialisation of polycarbonate double glazing
- 2. Cost-effective low-e coating solutions on polycarbonate
- 3. U value and g value assessments of polycarbonate double glazed prototypes
- 4. Prototype double glazed window
- 5. Estimation of service life

Stage-Gates:

Year 1 Stage-Gate Deliverable: Functional prototype polycarbonate double glazed device with preliminary thermal and mechanical property evaluations.

5.2 Resources and devices

Most material processing and characterisation equipment exists at Monash for the materials development and characterisation. However, there are a few pieces of equipment that are not available. There is no Pencil Hardness Tester found at Monash university; however, it is a cheap device (around 250 AUD). In addition we do not have a Xenon Arc Weather-Ometers for accelerated aging tests. Since any UV absorption study will need to be carried out continuously for a year of time, it is unlikely to find an academic to collaborate with. Alternatively, we can outsource the testing to a company like Intertek which provides reliable services in testing, inspecting and certifying products.

For the experimental determination of g value, we need to measure the angular-dependent (i.e., as function of various incident angles) energetic transmittance and reflectance over the UV-Visible-Near infrared radiation, based on the method described in the appendix 6.4. We can collaborate with an academic partner (e.g., Solar Energy and Building Physics Laboratory, EPFL, Switzerland, which is my former lab) who has the relevant test devices.

Alternatively, we can do the measurement to obtain the transmittance/reflectance spectrums at normal incidence, and perform the simple measurement of the temperature differences between inner and outer panes of a double glazing; based on these data, we can estimate the g value by optical modelling and simulation. A simple thermal conductivity system would be required for this (<\$1000), but all optical equipment are available at Monash.

For the preliminary determination of U value, we can already get a rather precise estimation by calculation with the relevant parameters described in standards (e.g., EN 673). This would be done using an Engineering software in the form of Ansys or equivalent.

5.3 Likelihood of success

There are several risk factors in this project, which are explored below.

Development of hard-coating solutions.

• What is the likelihood of success of the development of the novel hard coats that has an acceptable service life

Commercially, a hard coat that has an acceptable service life (i.e., 20 years) is almost there. As mentioned above, the service life of common hardcoated polycarbonate (with UV protection) for building glazing is around 15 years, and this estimation is based on the service life of Teijin, Sabic, and Bayer's polycarbonate products. Based on Teijin's product, Ibiden (a Japanese electronics company) has introduced an interlayer between the primer coat and the top coat, prolonging the service life by 3 years [Iviii]. Therefore, the state-of-the-art technology is not far from having a hard coat that has 20-year service life. However, whether this level of service life could be achieve in a hardcoat developed in-house is to be determined. Working with existing hard coat manufactures would be a starting point, but if these pathways fail, then development of a formulation will be required. There is risk in this given the short time scale of this project.

• Can the hard coats be manufactured at industrial scale?

The potential for scale-up of hard coats is also promising, The manufacturing of durable Teijin's hard coats are by both sol-gel method (one of the wet chemical coating methods) and chemical vapor deposition (CVD). The industrialization of wet chemical coating have been very well established. In December 2016, a pilot plant that enables CVD processing on 3-dimensional substrates of actual product size (1300 mm × 800 mm) was installed. Moving forward, Teijin aims to establish mass production technology for CVD plastic glazing to bring this technology to the market [xli]. As above, this demonstrates scalability, but exploring any potential assessment of Teijin's coatings would

need to be negotiated. If in-house coatings would need to be formulated, then scaling would need to be considered, which isn't a trivial task in itself. All solutions developed would harness scalable deposition processes, but these would need to be translated.

Low-E Coatings on Polycarbonates

Several companies offer low-E coatings on plastic substrates. In this project we would evaluate these coatings through lamination directly onto polycarbonate sheets. The only risk here is that the lamination process will require optimisation.

Energy Performance and Service Life

The energy performance evaluation and the service life prediction are about assessments and time, i.e., there are no essential R&D obstacles in these activities. Therefore these are low risk activities. However, achieving commercially viable performance characteristics across both of these parameters inherently depends on the progress around Low-E coatings and the hard-coatings, which does present a risk that depends on accessible commercial materials and/or developing proprietary solutions.

6 Appendix

6.1 Mechanical, optical, thermal properties of the selected plastics

	PMMA	ETFE	Polycarbonate	PET	PP	Tempered glass
Mechanical properties						
Tensile strength	60 - 70 MPa	38 – 48 MPa	55–75 MPa	5.52 - 90.0 MPa	4.00 - 369 MPa	120 – 200 MPa
Compressive strength	124 MPa	10 - 20 MPa (film)	>80 MPa	2.30 - 103 MPa	34.5 - 55.2 MPa	Surface compression strength > 69 MPa
Young's modulus	3 GPa	0.8 GPa	3.17 GPa	0.140 - 5.20 GPa	0.00800 - 8.25 GPa	70 GPa
Pencil Hardness Evaluation (Scratch resistance)	2H	/	2B-3B	1H-6 H	/	Not scratchable by pencil
Optical properties						
Total transmission	92%	> 92%	> 90%	70.00 - 90.00%	< 85%	92%
UV-transmission	0 at 320 nm Similar to glass	83 – 88% at 200 – 380 nm	0 at 380 Good protection from UV	Similar to polycarbonate	85 - 90% at 250 – 380 nm	Allow UV-A to pass through while almost 100% of the UV-B and UV-C light is blocked.
Haze (2 mm thick)	0.2 - 1%	> 5% (0.3 mm)	Less than 1%	0.2 - 40 %	8 - 91%	Crystal clear
Refractive index	1.49	1.40	1.60	1.57	1.49	1.52
Thermal properties						
Glass transition temperature	100-115°C	100°C	147°C	60 - 87°C	-2010 °C	564°C
Melting point	130 °C	225 – 270 °C	155°C	250 - 270°C	210 - 290°C	1400 °C to 1600 °C
Thermal conductivity	0.19 W/mk	0.238 W/mk	0.2 W/mK	0.15 - 0.24 W/mK	0.11 - 0.44 W/mK	1.1 W/mK
Thermal expansion coefficient (µm/K)	70 -77	90 - 170	65 – 70	20-80	100-180	9.0 μm/K
Heat distortion temp.	99 °C	105°C	140 °C	70 °C	100-105 °C	/
Service T. (continuous)	- 70 – 84 °C	-100 – 165 °C	-30 - 140 °C	Minimum: -40°C	Mimimum: -4010°C	-190 - 250° C
				Maximum 80.0 - 140.0°C	Maximum: 100 - 140°C	
UL Flammability	Burning rate 30.5 mm/min at 3mm thickness, molten drips EU fire classification E	Low flammability, self- extinguishing, no molten drips Flame retardant	EU fire classification B UL94 V-0 Flame retardant	UL94 V-0 Flame retardant	UL94 V-0 Flame retardant	/
Ignition temperature	250 – 400 °C	540°C	580°C	349°C	570°C	/
Combustion products	carbon dioxide, water, carbon monoxide and low-molecular- weight compounds	Corrosive and toxic gas (HF)	Similar to PMMA	Similar to PMMA	Similar to PMMA	1
Other properties						
Density	1.12 -1.2 g/cm ³	1.7 g/cm ³	1.2 g/cm ³	1.3-1.4 g/cm ³	0.9 g/cm ³	2.5 g/cm3
Life time (virgin, outdoor)	> 30 years	> 50 years	< 5 years	< 1 year	< 1 year	Hundres of years

6.2 Calculation of U value

The thermal performances of a double glazing can be estimated using the standards developed for the buildings such as the EN 673 for the calculation of the U value. It is noted that the overall U value for a window depends on window types (single, double, or triple glazing), the frame types, frame patterns, etc. Below shows the calculation the U value of the glazing part of a clear glass double glazed window.



$$U_g = \frac{1}{R_{tot}} \qquad R_{tot} = \frac{1}{h_i} + \frac{d_{glass}}{\lambda_{glass}} + \frac{1}{\Lambda} + \frac{d_{glass}}{\lambda_{glass}} + \frac{1}{h_e}$$

Empirical values, $h_i = 8 \text{ w/m}^2 \text{k}$, $h_e = 18 \text{ w/m}^2 \text{k}$

 λ – thermal conductivity of external glass pane / the air gap / internal glass pane h_i - thermal conductivity of the thin transition layer between internal glass pane and the air h_e - thermal conductivity of the thin transition layer between external glass pane and the air h_i and h_e can be checked in the standards, e.g., European standard EN673

6.3 Calculation of solar heat gain coefficient

1. Determination of the equation of the value g of the single clear glass glazing shown schematically below



 $I_{inc}: \text{ incident irradiance (W/m^2)}$ $I_r: \text{ reflected irradiance (W/m^2)}$ $r_e: \text{ reflection coefficient (-)}$ $I_i: \text{ transmitted irradiance (W/m^2)}$ $t_e: \text{ transmission coefficient (-)}$ $Q_{e/i}: \text{ specific heat flux in direction from outside / inside (W/m^2)}$ $R_{e/i}: \text{ thermal resistance outdoor / indoor ((m^2K)/W)}$ $Q_{abs}: \text{ specific heat flux absorbed by glazing (W/m^2)}$

We consider the following equations with r_e and t_e as constants:

$$Q_{abs} = I_{inc} \cdot (1 - r_e - t_e) \tag{1}$$

$$Q_{abs} = Q_i + Q_e \tag{2}$$

$$Q_e \cdot R_e = Q_i \cdot R_i \tag{3}$$

Based on equation (3), we get:

$$Q_e = Q_i \cdot \frac{R_i}{R_e}$$

We replace Q_e in the equation (2) and therefore we get:

$$Q_{abs} = Q_i + Q_i \cdot \frac{R_i}{R_e}$$

Finally we replace Q_{abs} in the equation (1):

$$I_{inc} \cdot (1 - r_e - t_e) = Q_i + Q_i \cdot \frac{R_i}{R_e}$$

Then we get Q_i :

$$Q_i = \frac{I_{inc} \cdot (1 - r_e - t_e)}{1 + \frac{R_i}{R_e}}$$

Then we get the expression for the g value:

$$g = \frac{I_t + Q_i}{I_{inc}} = \frac{I_{inc} \cdot t_e + Q_i}{I_{inc}} = \frac{I_{inc} \cdot t_e + \left(\frac{I_{inc} \cdot (1 - r_e - t_e)}{1 + \frac{R_i}{R_e}}\right)}{I_{inc}}$$
$$g = t_e + \frac{(1 - r_e - t_e)}{1 + \frac{R_i}{R_e}}$$

2. For double glazing with two clear glass panes, the solar heat transfer process is shown schematically below



Where

$$R_{i1} = \frac{1}{\Lambda} + \frac{d_{glass}}{\lambda_{glass}} + \frac{1}{h_i}$$
$$R_{e1} = \frac{1}{h_e}$$
$$R_{i2} = \frac{1}{h_i}$$
$$R_{e2} = \frac{1}{h_e} + \frac{d_{glass}}{\lambda_{glass}} + \frac{1}{\Lambda}$$

6.4 Experimental determination of g value

The experimental determination of g value is based on the equation below:

$$g(\varphi) = \tau_e(\varphi) + q_i$$

where

 $g(\varphi)$ – g value at incident angle φ

$au_e(arphi)$ - energetic transmission coefficient at incident angle arphi

 q_i - coefficient of energy absorbed by the glass and reemitted as heat on the interior side

Considering an isotropic diffuse sky, an effective g value was calculated by integrated the measured $g(\phi)$ on a half sphere.

The angular dependent solar gain can be experimentally determined using the method developed by Reber et al. [lix]. This method is based on the measurement of the external and internal surface temperature of the double glazing while being under a beam from a solar simulator. The light source of the solar simulator is a water-cooled xenon arc lamp (1000 W). A spectral distribution similar to that of solar radiation is achieved by the means of filters. The example set up can be found at the Solar Energy and Building Physics Laboratory, EPFL, Switzerland [lx]. The set up can measure the transmittance and reflectance at angles of incidence from 0° to 75°. Spectral intensities are measured for wavelengths ranging from 350 to 2150 nm (UV-Vis-NIR).

6.5 Service life prediction: correlation between accelerated aging test and exposure in actual weathers

A great deal of confusion arises from the simple question, "What is the correlation of this test to Melbourne?" The problem is that the acceleration from particular exposure conditions is a function of the response of a particular material to those conditions. That is, the equipment and settings accelerate the degradation of materials, and how that translates to service life depends on the degree to which the various processes are accelerated for that material. Therefore, the question should be, "What is the correlation of this test to Florida for my material?" The same test conditions can have different correlations for materials X, Y, and Z.

A larger question to be addressed is what is meant by the term "correlation." An empirical correlation might show that an article reaches failure after 500 h of exposure under some test conditions and after 2 years of exposure at a given test site. But this cannot be applied to other materials without knowing (1) what is causing the acceleration of the benchmark material, and (2) how other materials respond to the conditions. Indeed, some conditions might have no correlation at all if critical failure modes are not accelerated properly.

Analysis of solar spectral power distributions indicates that radiant exposure in the 1-nm interval at 340 nm, where weathering instruments commonly measure their irradiance, is 1.37% of the exposure received in the range 295–385 nm, where outdoor UV is commonly measured. Average annual exposure at a Miami (Florida) test site for samples exposed south-facing at a 45 degree angle is 286 MJ/m² in the range 295–385 nm, so the annual dose is 3.9 MJ/m2/nm at 340 nm. This should be the radiant exposure required by a properly filtered xenon arc (e.g., using Atlas Right Light or Q-Lab Daylight-F filters) to simulate 1 year of Florida exposure. The simulation of the solar spectrum in the UV by these filters eliminates the need to make corrections for acceleration by unnaturally short wavelength UV. However, to achieve acceleration, more photons have to be applied per day than outdoors. This acceleration can be done by using "summer noon" or higher irradiance all the time and eliminating or reducing the dark period. In any case, it must be shown how much acceleration is expected from the higher irradiance.

This is not the full story. Samples in a weathering device usually are hotter than the effective temperature outdoors. This can be corrected by finding the activation energy, the effective temperature outdoors (T_{eff}), and measuring the temperature of the specimen in the test chamber (Ttest). The ratio of the $\frac{k_{\rm eff}}{k_{\rm test}} = \exp\left[\frac{E_{\rm a}}{R}\left(\frac{1}{T_{\rm test}} - \frac{1}{T_{\rm eff}}\right)\right] = \exp\left[\frac{E_{\rm a}(T_{\rm eff} - T_{\rm test})}{RT_{\rm eff}T_{\rm test}}\right]$ temperatures rates at the two is Equation 1, where the temperatures are in kelvins. As an example, several plastics have Ea = 4 kcal/mol (17 kJ/mol), a Teff in Florida of ~32°C (305 K), and a Ttest of 45°C (318 K). Using R = 1.987 cal/K and Ea = 4000 cal/mol, the rate ratio is 0.76. This reduces the filtered xenon arc dose expected to cause one Miami-year's-worth of degradation to 3.0 MJ/m²/nm at 340 nm.

$$\frac{k_{\text{eff}}}{k_{\text{test}}} = \exp\left[\frac{E_{\text{a}}}{R}\left(\frac{1}{T_{\text{test}}} - \frac{1}{T_{\text{eff}}}\right)\right] = \exp\left[\frac{E_{\text{a}}(T_{\text{eff}} - T_{\text{test}})}{RT_{\text{eff}}T_{\text{test}}}\right]$$
Equation 1

Finally, the effects of humidity, condensation, and rain must be accounted. In many cases, humidity and condensation are not particularly important variables, with transportation coatings being a notable exception [12]. Simulating the washing effects of rain is important for materials that undergo erosion, and this has proved more difficult.

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