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Polymer Interlayers for Glass Lamination-A Review

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Abstract: ***This review presents the current status of research on the development and applications of different polymeric materials used in the preparation of laminated glasses. Safety glasses used for construction are annealed, heat strengthened, toughened, and laminated. Under normal serviceability temperatures, glass behaves like a linear elastic material, which will break suddenly when tensile stresses exceed a critical value. The main safety concept is to create an overall structural plasticity for the glass. This is usually done by building laminates, individual glass panes alternated with soft transparent interlayers are composed as one coherent structural element. Different types of polymeric interlayers have been used in laminated glasses depending upon their applications for example polyvinylbutyral, polycarbonates, polyurethanes and other polymers or copolymers etc. A wide range of applications of different types of interlayers of polymer composites have been compared. The review includes physical properties of different types of glasses, fabrication procedures such as heat and pressure method and UV Curing method. In the present work it was observed that the extensive efforts are being made to replace polyvinyl butyral film by alternate films made up of polycarbonate, polyurethanes, vinyl resins etc.***

Keywords: Safety glass, Laminated, polycarbonate, polyvinyl butyral, polyurethane

1. Introduction

This review deals specifically with different types of polymeric materials used as interlayers in laminated glasses. The choice of the interlayer materials stems from the type of application of laminates. In the present world, crime is growing rapidly which has led to increased use of safety glass in constructions because it works as a guard of the building. These glasses offer protection from natural and man made disasters. Safety glasses used for construction are annealed, heat strengthened, toughened, and laminated. Their demand is increasing day by day as safety and security concerns are gaining importance because of environmental threats and security.

1.1 Annealed glass

Annealing is a process of slowly cooling glass to relieve internal stresses after it was formed allowing the glass to be cut by scoring and snapping(Collins E.F., 1921). The glass is heated until the temperature reaches a stress relief point (annealing temperature) at a viscosity, η, of 1013 Poise, at which the glass is still too hard to deform, but is soft enough for the stresses to relax. It is then allowed to heat soak until its temperature is even throughout. The glass is then slowly cooled at a predetermined rate until its temperature is below the strain point (η = 1014.5 Poise). Now the temperature can safely be dropped to room temperature at a rate limited by the heat capacity, thickness, thermal conductivity, and thermal expansion coefficient of the glass. After this process the material can be cut to size, drilled or polished. On breakage, the glass tends to form sharp-edged, pointed shards (Haldimann et al., 2008).

1.2 Heat-strengthened glass

In the production of the glass plates there are processes which increase the strength of annealed glass. Heat treated glass possesses additional strength, impact resistance and resistance to thermal stresses. Annealed glass which has been heated to a temperature near its softening point and forced to cool rapidly under carefully controlled conditions is described as heat treated glass.

1.3 Toughened Glass

The term toughened glass is generally used to describe fully tempered glass but is sometimes used to describe heat strengthened glass as both types undergo a thermal toughening process. The basic principle employed in the heat treating process is to create an initial condition of surface and edge compression. This condition is achieved by first heating the glass, then cooling the surfaces rapidly. This leaves the center glass thickness relatively hot compared to the surfaces. As the center thickness then cools, it forces the surfaces and edges into compression. Wind pressure, missile impact, thermal stresses or other applied loads first overcome this compression before there is any possibility of fracture.

Table 1: Summarized data of different safety glasses used as structural materials

Type of glass	*Bending strength*	*Required Characteristic strength*	*Residual Compressive surface stress*	*Quality Standards*	*Breakage characteristics*
Annealed glass	Low	45 N/mm^2 (EN 572-1)	-	ASTM C 1036, EN 572-1 & EN 572-2	Forms sharp edged, pointed shards
Heat strengthened glass	Higher than annealed glass	75 N/mm^2 (EN 1863-1)	24-69 N/mm^2	ASTM C 1048 & EN 1863-1	Similar to annealed glass
Toughened glass	Higher than heat strengthened glass	120 N/mm^2 (EN 12150-1)	Over 69 N/mm^2	ASTM C 1048 & EN 12150-1	Breaks into small, relatively blunt glass fragments(dice)

1.4 Laminated Glasses

Laminated glass consists of two or more glass panes bonded by one or more plastic or resin interlayer (Figure 1). Laminated glass units can be prepared by using soda lime glass, annealed glass or tempered glass (heat strengthened glass). Laminated glass offers the potential for a vast range of options in regard to the energy performance and aesthetics of building glazing, while also furnishing enhanced safety, structural integrity, security and sound attenuation. In the event of breaking, laminated glass is held in place by the interlayer and its high strength prevents the glass from breaking up into large sharp pieces.

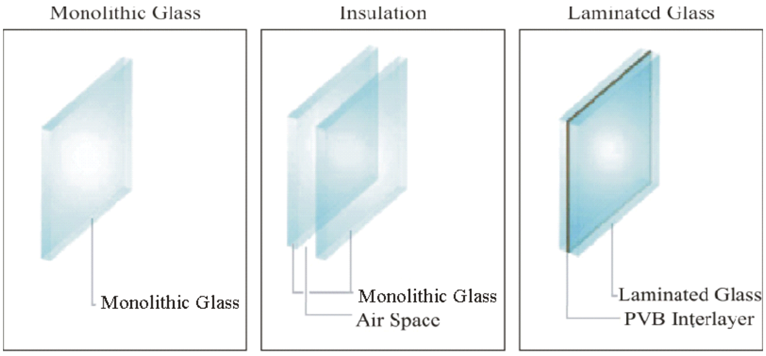

Figure 1: Shows the basic structures of a monolithic and a laminated glass with a PVB interlayer

It has gained popularity as an architectural glazing product and its safety and versatility are being used to satisfy many design situations. It is useful in a variety of innovative applications, and can provide benefits such as physical barrier properties for security applications, impact resistance against low and high energy airborne objects and projectiles, blast resistance, acoustical dampening and other architectural capabilities (Ando et al., 1981). Laminated glass exhibiting these features is increasingly in demand because of environmental threats and security. In order to set the stage around this review and underline its peculiar message, it is important to provide a background of fabrication procedures and polymeric materials used in the preparation of laminated glasses.

2. Methods used in preparation of Laminated glasses

Laminated glasses are produced by either of the following two methods i.e. heat and pressure method and UV curing method or sometimes both processes are combined in their preparation. Figure 2 shows the different types of laminated glass assemblies that can be used as safety glasses in various applications depending on their impact resistance and other desirable properties.

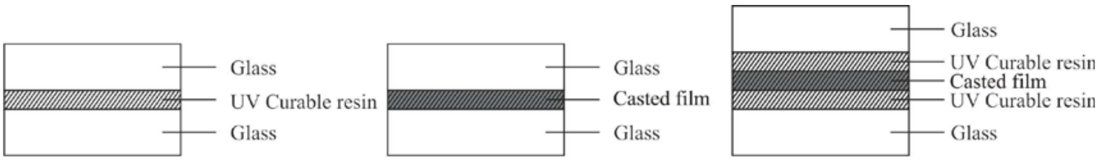

Figure 2: Different types of Laminated Glasses

2.1 Heat and Pressure method

This method is practiced since 1930s which involves the use of a film interlayer that is bonded to the glass by application of heat and pressure. In this process, typically a polymer film is placed between two glass pieces and air is removed in a clean room by specialized nip rollers or vacuum. Final lamination is completed in autoclaves, hydraulic presses and similar devices (Snedeker et al., 1972) where high temperature and pressure are applied to effect bonding of the film to the glass. The bonding is conducted usually at temperature ranging from about 110 ^{0}C – 140 ^{0}C and at pressures of from about 2 to about 300 psi. This method generally requires a huge capital investment and labor as the size of the autoclave required depends upon the laminate dimensions which results in added cost to the finished laminate. Such laminates suffer from residual stresses from heat bonding (Audi et al., 1988).

2.2 UV curing method

Laminated glasses are readily obtained by UV curing at ambient temperature through the use of a liquid based interlayer system, wherein the interlayer space is filled by pumping a low viscosity liquid. Several reports have already demonstrated the feasibility of the UV curing method to produce safety glasses (Herman F.M., 2003). In this process, the two glass panes are cleaned thoroughly and a cavity is created by applying double sided tape. The resin is then pumped into the cavity using an automated metering pump system. The final step is to cure the resin by exposure to UV radiation. UV based laminating systems are cost effective as compared with conventional autoclaved systems. They offer a number of advantages such as rapid filling and curing process, making production of made to size, high quality glass laminates in a very short time. UV curable interlayers are single component systems that do not require mixing prior to use ensuring a more consistent laminated resin product. They have a virtually limitless pot life until exposed to UV light.

3. Interlayers for Laminated Glasses

Although different polymeric materials have been used as interlayers in the fabrication of laminated glasses, it is important to emphasize the development and applications of polyvinylbutyral polycarbonates, and polyurethanes interlayers in laminated glasses.

3.1 Polyvinylbutyral based Interlayers

PVB has been the dominant interlayer material since the late 1930's. It is a 70 year old product. It is currently manufactured and marketed by a number of companies worldwide, including Du Pont (Welmington. Del.) (Butacite- brand PVB, introduced in 1938), Solutia (St. Louis, Mo.) (Saflex- brand PVB, introduced in 1940), Kururay specialities Europe (Frankfort, Germany) (Trosifol-brand PVB), and Sekisui (Kyoto, Japan).

Synthesis: It is synthesized by reaction of polyvinylalcohol with butyraldehyde. PVB resin can be produced by either a solvent process or an aqueous process. In the solvent process, polyvinylacetate is saponified by transesterification in the presence of ethanol and a mineral acid catalyst to produce polyvinyl alcohol. The ethanol and ethyl acetate are separated from the precipitated polyvinyl alcohol by centrifugation. In a separate unit operation, polyvinylalcohol is acetalized after being reslurried with ethanol and heated with butyraldehyde and the acid catalyst. Upon completion of the acetalization reaction in both solvent and aqueous process, the acid catalyst is neutralized. In both aqueous and solvent processes, resin is then separated, washed and dried.

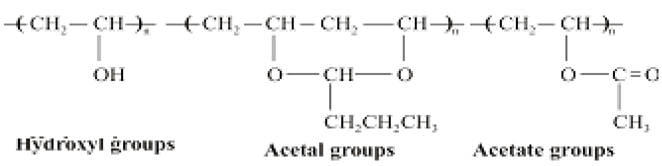

Figure 3: Functional groups present in polyvinyl butyral backbone

Properties: Laminated to glass, polyvinylbutyral exhibits high adhesion, optical clarity, stability to sunlight, and high tear resistance and impact absorbing characteristics (Faries R.H., 1993). Its main function is to absorb energy, such as that caused by a blow from an object, without allowing penetration through the opening or the dispersion of shards of glass, thus minimizing damage or injury to the objects or persons within an enclosed area. It adheres to glass fragments after a glass breaking impact, thereby helping to reduce injury form flying glass. The interlayers high tear strength and resiliency acts like safety net by absorbing enough energy to resist penetration by a projectile or a vehicle occupant's head Johnson et al., 1999). It also provides other beneficial effects, such as to attenuate acoustic noise, reduce UV and/or IR light transmission, and/or enhance the appearance and aesthetic appeal of window openings. PVB is superior over the vinyl acetal polymers from formaldehyde, acetaldehyde or propionaldehyde for laminated safety glass owing to its better cold temperature characteristics.

Applications: It is commonly used in the manufacture of polymer layers that can be used as interlayers in light transmitting laminates such as safety glass or polymeric laminates. For laminated safety glass, the strength of adhesive bond between the glass and the interlayer is carefully controlled with additives usually salts, to achieve the desired balance (D'Errico J.J. et al., 1995). PVB composites layered with a film of poly (ethyleneterephthalate), acrylic or polycarbonate, or metal fabric in addition to glass are designed for some specialty uses. Most laminated glazing are a trilayer composite of glass-PVB-glass, but some specialty applications can include a polymer film interlayer in the PVB to achieve decorative designs or solar reflective properties, or even a metal fabric to provide radiofrequency shielding from external sources. For security applications, such as bullet and projectile resistant laminates, multiply PVB interlayer and glass composites are designed.

Endo et al., (1990) has reported an improved interlayer for laminated glass that can be used in various transportation facilities such as automobiles and aircraft and windowpanes of buildings. Schimmelpenningh et al. (1998) have developed safety glass structure resistant to extreme wind and impact. PVB (plasticized) interlayer was used to construct the laminated glass units. If an interlayer is too soft or adhesion with glass is too low, then the interlayer will not have the strength to resist high wind loading after an impact. On the other hand, if the interlayer is too stiff and adhesion to glass is too high, then the interlayer will not have a high level of impact resistance. Therefore improved interlayer compositions are needed to provide safety glass that has high impact resistance and high cyclical wind loading resistance for hurricane protection. Chen Wenjie et al., (2008) have developed PVB interlayers with high level of impact protection that can be used in hurricane protection applications or in bullet proof glass applications. The developed PVB interlayer comprises a relatively stiff polyvinyl butyral inner layer disposed between two relatively soft outer PVB layers.

Polyvinyl butyral interlayer in laminated glasses has also been used for printing an image by an inkjet printing process. Eric T. Pray (2006) has devised a process for the production of decorative laminated safety glass with good adhesive qualities. The UV curable ink was used on the embossed surface of the PVB film, which adhered evenly to the surface without undue thickening or thinning in the low and high areas of the embossed surface. Kiyofumi Toyama et al., (2007) describe a PVB interlayer for laminated glass with outstanding sound insulation performance over a broad temperature range on a long term basis, and owing to satisfactory physical properties further protected against panel shear and foaming, without sacrificing the basic functional characteristics required of laminated glasses. Some modeling studies have also been performed on PVB glass laminates to investigate the mechanical behavior of windshield glass (Yong et al., 2013).

3.2 Polycarbonates

The vast majority of the polycarbonates are based on bisphenol A (BPA), and sold under the trade names Lexan (GE), Makrolon (Bayer), Calibre (Dow), Panlite (Teijin) and Iupilon (Mitsubishi). Polycarbonate is the object of both industrial and academic research because of its widespread utility and unusual properties.

Synthesis: These are produced either by an interfacial polymerization process utilizing phosgene or through trans-esterification or melt process. Most BPA polycarbonates are produced by an interfacial polymerization process utilizing phosgene. This process involves stirring a slurry or solution of BPA and 1-5% of a chain stopper such as phenol, p-t-butylphenol or p-cumylphenol in mixture of methylene chloride and water, while adding phosgene in the presence of a small amount (0.1-3%) of a tertiary amine catalyst. Preparation of polycarbonates through melt process is advantageous as it eliminates the use of solvents and phosgene.

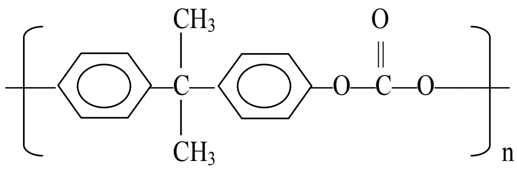

Figure 4: Repeating chemical structure unit of Polycarbonate made from BPA

Properties: Polycarbonates are unusual and extremely useful class of high heat polymers known for their toughness and clarity. PC has the advantage over other transparent polymers such as polymethylmethacrylate (PMMA) of being very tough (Chiou et al., 1997, Grellmann et al., 1997, Wu et al., 2001). Extreme toughness, transparency, low color, resistance to burning and maintenance of engineering properties over a wide thermal range are the outstanding properties of polycarbonates that make it useful for a variety of applications. Polycarbonate is a transparent nearly colorless polymer making it attractive glass replacement and is also useful in glass laminates. The clarity and impact resistance of polycarbonate is 250 times greater than glass and 30 times greater than acrylic sheet. It has a high refractive index (1.588) and a lower density than glass (1.52 g/ml). Its visible light transmission is about 90% and haze is only 1-2% in typical commercial products. Majority of the polycarbonates are based on bisphenol A (BPA) having glass transition temperature in the range of 140-155 ^{0}C.

Applications: Polycarbonate is used for security applications with glass or other materials and has projectile stopping capability, as the material softens upon impact with a bullet, absorbing the projectile's energy. If a bullet is fired against such a construction, although the glass shatters, it is still able to deform and slow the projectile enough so that the tough PC is able to prevent penetration of the projectile (or other fragments) (Kajon et al., 2000). One difficulty in polycarbonate laminates arises from the fact that glass and polycarbonate have different coefficients of thermal expansion and therefore expands at different rates. Gasper Mucaria (1983) prepared glass units which include one or more polycarbonate layers which avoids the problems inherent in the differential heat expansion rates of glass and plastic material. In this safety glass unit a central polycarbonate sheet is surrounded by and spaced apart from glass sheets and one or more of the peripheral edges of the polycarbonate sheet lies within a channel which in turn provides room for lateral expansion of polycarbonate sheet.

Polycarbonate film has also been used as thermoplastic substrate to impart improved abrasion resistance as well as improved weathering resistance. It has also been used in hurricane resistant windows and doors with a unique framing system that will not only prevent wind and flying debris from entering the enclosed structure but assists in keeping the glass structure in the frame (Bahya Charles E., 1998). A considerable amount of research has been devoted to PC laminates. In the same series, Cook and L. Richard (2009) also published a patent claiming optically transparent resilent laminate materials.

3.3 Polyurethane based Interlayers

Polyurethanes are in the class of compounds called reaction polymers, which include epoxies, unsaturated polyesters and phenolics (Woods G., 1990 and Ulrich H., 1996). Since the discovery of polyurethanes in 1937 there has been an ever increasing and widespread use of urethanes in coatings industry. Pus are sold under the trade names Lycra, Estane, Biomer and Pellethane.

Synthesis: Polyurethane synthesis contains a diverse range of synthetic options. Generally polyurethanes are synthesized either through addition polymerization or condensation polymerization. Addition polymerization occurs when unsaturated monomers react through stages of initiation, propagation and termination. Condensation or step growth polymerization occurs when bifunctional molecules react in a stepwise fashion to produce linear chain of monomer residues. Although no small molecule is eliminated during polymerization the reaction between the diol and diisocyanate can be classified as a condensation reaction. The primary reaction for polyurethane synthesis is the reaction of isocyanate group with a hydroxyl terminated molecule (Figure 5) in the presence of a catalyst or other additives. Both aliphatic and aromatic isocyanates can be used to synthesize polyurethanes. Polyols available for elastomer synthesis include polyesters, polyethers, polycarbonates, hydrocarbons and polydimethyl siloxanes. Catalysts used are sodium hydroxide, sodium acetate and triamines. DABCO (tri-ethylene diamine 1,4 diazo (2, 2, 2) bicycle-octane is commonly used triamine.

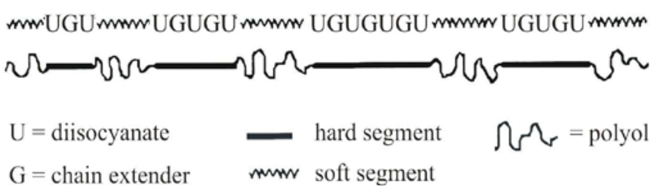

Figure 5: Segmented polyurethane structure

Properties: The chemistry of polyurethane elastomers can be varied through the chemistry of the diisocyanate, the polyol, the chain extender and the ratios in which they are reacted. The chemistry ultimately affects the mechanical and physical properties of Pus and their application. Polyurethanes are a family of heterogeneous polymers, they contain the urethane linkage, analogous to the carbamate group in organic chemistry, within the polymer chains. Urethane groups usually do not constitute the majority of the functional groups within polyurethane. It is the ability to incorporate other functional groups into the polymer network that contributes to the range of properties exhibited by polyurethane materials. Consequently, the properties of polyurethanes range from rigid hard thermosetting materials to those of much softer elastomers. Generally thermoplastic polyurethanes have very high tensile strength, toughness, abrasion resistance and resistance to degradation, in addition to biocompatibility that has sustained their use as biomaterials (Lamba et al., 1998). Polyurethanes coatings are used wherever applications require abrasion resistance, skin flexibility, fast curing, good adhesion and chemical resistance. Hydrogen bonds from the urethane groups of the hard segments give rise to a physical network which account in part for many desired properties.

Applications: The domain structure of polyurethanes and their scope for structural diversity creates a wide variety of uses and applications for these materials. PU interlayers exhibit interesting properties but because of their specific manner of application, they have limited applications. Kraemling et al. (1990) have developed an insert layer that can be used in laminated glasses with very diverse applications. The plastic adhesive layer for use as an insert layer in laminated glasses was formed from a castable mixture of a polyfunctional polyol component and a polyfunctional isocyanate component. The laminated glasses developed using these formulations along with polycarbonate sheets and PVB films were used as shatter proof glass and bullet proof glass respectively. The soundproofing qualities of these laminated glasses were found to be much better than those of a similar glass having a polyvinylbutyral insert layer.

Two properties of the bond which are important are the adhesive or bond strength and performance of the bond under impact. To improve the bond, without significant loss of water resistance in particular however presents a problem because these properties conflict. Adhesion to glass requires a degree of hydrophilicity whereas hydrophobicity is required for water resistance. Improvement was found in one or both of these properties by using the proposed adhesive. This adhesive enables the replacement of PVB conventionally employed as an interlayer in safety glass laminates by alternative plastic materials which perform better at elevated temperatures especially at temperature of 40 ^{O}C or more at which PVB tends to lose much of its strength. Mark G. Tilley et al. (1992) had discussed UV curable coating compositions, which are abrasion resistant and UV stabilized containing aliphatic acrylated urethane. Bruce R.S. and Hall, Christopher W.G. (1995) has also used polyurethane as a thermoplastic material in the preparation of laminated glazings. A polyurethane based on an aliphatic polyether or polyester has been used to protect the chemically strengthened glass surface. Polyurethanes are highly effective in laminated glass films, since these materials provide the films with a high tear and cohesive resistance (Haverkamp U., 1998). Such laminated glass units has been used as burglar proof glass windows and glass doors.

Polyurethane interlayers also find diverse applications in laminated glasses used as automotive windows. A sufficiently thin laminated glazing comprising a rigid thermoplastic polyurethane sheet was developed for intrusion resistant automotive applications (Wang et al., 2005). Rukavina Thomas G. (2008) has invented a novel interlayer and a laminated window containing polyurethane that exhibits improved ballistic properties.

Table 2: Comparative Properties of Interlayer Materials used in Laminated glasses

Interlayers / *Properties*	*Poly-vinylbutyral*	*Poly-carbonate*	*Poly-urethane*	*Standard metho*
Specific density(g/cc)	1.08	1.2	1.05	ASTM D792
Refractive index	1.485	1.586	1.496	ASTM D-542
Hardness (Shore D)		80	Variable (20A to95A, 50D- 80D)	ASTM D 2240/ASTM D676-59T*
Tensile strength	320 Kg/cm 228.1 mPa	52 mPa	20-62 mPa 2000-12000 psi	ASTM D638/ ASTM D412-61T*
Elongation at break percentage	250	3%	800-270	ASTM D638/ASTM D412-61T*
Water absorption (%)	0.45	0.15		Immersion 24 hrs, ASTM D-570
Adhesion with Glass	Poor	Good	Excellent	Boil test IS 2553,1990
Light stability	Poor	Poor	Excellent	IS 2553,1990
Glass Transition temperature (^{0}C)	145 -161	Variable (-60-)	22	ASTM D3418
Haze(%)	<0.5	<0.3		ASTM D-1003

4. Future Prospects

Presently most of the laminated glasses are being manufactured by using Polyvinyl butyral film. Several researchers have made efforts in developing alternative interlayers which are mainly based on polyurethanes, polycarbonates, EVA, acrylates etc. but limited success is obtained. Hence it can be concluded that more efforts should be made to find out alternative materials to PVB film. The performance of the laminated glass is not evaluated for compression, flexural, impact i.e. mechanical properties and for service performance. It can further be concluded that lots of efforts are required to gain data for modeling studies. Very few studies have been made on modeling of fracture analysis which can be helpful in the development of interlayer and as well as for their evaluation. It is therefore proposed that more efforts should be made in developing different models especially for fracture analysis.

5. Conclusion

Laminated glass furnishes solutions to many architectural design problems and offers increased protection from the effects of disasters such as hurricane, earthquake and bomb blast. Benefits of laminated glasses include: Safety and Security, Sound Control, Solar Energy Performance, Ultraviolet Screening and protection from injury and damage during natural disasters. Specially designed polymeric interlayers in laminated glass helps to offer the required properties. In this review different polymeric interlayers used in the preparation of laminated glasses have been discussed and compared. Physical properties of different types of glasses, fabrication procedures such as heat and pressure method and UV Curing method have also been detailed. Laminated glass is increasingly in demand because of environmental threats and security. There is always a need for materials which are characterized by superior penetration and spall resistance and which at the same time have good clarity, strength and integrity.

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