

Applying ceramic labels to glass

Wenke Hu and Gary Smay present findings from a study to define the stresses that are generated when an applied ceramic label is fused onto a soda-lime-silica glass surface, and explore how this can affect the performance of the decorated item.

Using conventional methods, the source material for an applied ceramic label (ACL), consisting of a glassy frit, colourant and dispersion media, is applied onto the surface of room temperature bottles, which are then heated to about 625°C. At this temperature, the ACL melts, is fused to and becomes an integral part of the glass surface.^{1,2} The labelled bottles are then cooled to room temperature at a rate that is consistent with normal annealing practices.

The suitability of using a ceramic label on either refillable or non-refillable glass containers depends primarily on the coefficient of thermal expansion (CTE) of the ACL relative to the glass substrate. If the CTE of the ACL and glass exactly match, no residual stress will be generated in either the ACL or the underlying glass surface. However, this ideal situation is seldom realised in normal commercial practice. Therefore, since some degree of mismatch routinely exists, it is imperative that the CTE of the ACL is less than that of the glass so that a compressive stress will be present in the ACL.²⁻⁶

It has been stated in the literature that under these conditions, the CTE difference can be relatively large with no adverse effects on the performance of the decorated glass article.⁴ This assertion was initially troubling because it was assumed that with large differences in the CTE, high magnitude tensile stresses would be generated in the surface of the glass immediately beneath the ACL. It was postulated that the presence of high tensile stresses could adversely affect the performance of the decorated item.⁷ This concern was the genesis of the current study.

Experimental procedure

A finite element computer stress analysis (FEA) model was utilised to determine the magnitude of the stress that is created in both the glass and the ACL when the CTE of the ACL and glass are different. The model consisted of a two-dimensional solid beam with a thin ACL layer situated on the surface of a much thicker glass substrate. The boundary conditions consisted of a fixed support on both ends of the beam. The mesh size ranged from 5.1 microns to 10.2 microns, values that provided an accurate assessment of the stresses throughout the ACL thickness and the underlying glass substrate.

ACL thicknesses of 25.4 microns, 38.1 microns and 50.8 microns were used, which are comparable to typical commercial values. Glass substrate thicknesses were 2.29mm and 1.27mm, representative of the sidewall thickness of refillable and non-refillable glass containers, respectively. The CTE of the glass was 88, 90 and 92 ($\times 10^{-7} \text{C}^{-1}$), typical of current soda-limesilicate glass container compositions. The CTE of the ACL ranged from 86 to 102 ($\times 10^{-7} \text{C}^{-1}$), which provided both positive and negative 'fits' relative to the CTE of the glass substrate.

It was assumed that the CTE for both the glass and the ACL were linear from the 'lower critical point or transformation point'² of the ACL to room temperature. It was further assumed that both the ACL and glass substrates were fully elastic below the ACL 'transformation

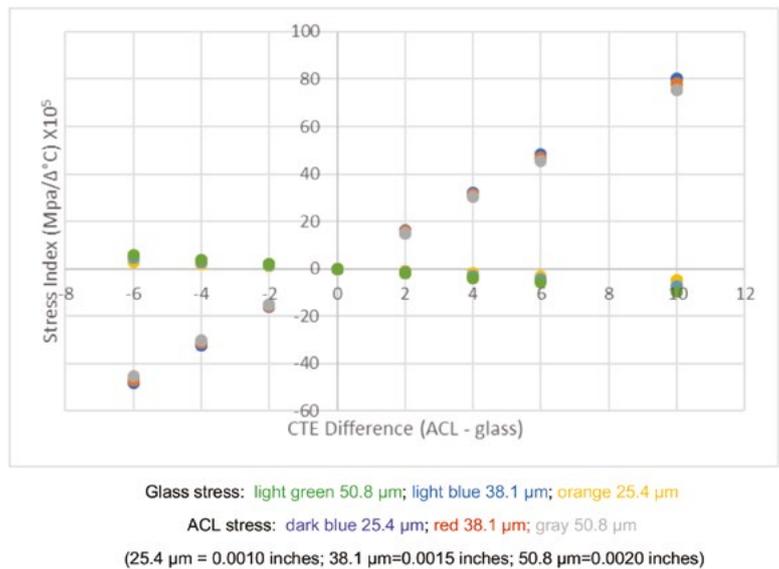


Figure 1: ACL and glass stresses for different ACL thicknesses (glass thickness = 2.29mm).

point' and that the elastic modulus of the ACL was equal to that of soda-lime-silica glass. This latter assumption was necessary because of the absence of data for the elastic modulus of typical ACL materials in the published literature.

The output of the FEA models provided a stress index in units of $\text{MPa}/\Delta^\circ\text{C}$, the difference in temperature being the value from the transformation point to room temperature. Stress magnitudes can be calculated by multiplying the stress

indices by the total temperature interval from the ACL transformation point to room temperature. However, in this paper, only the stress indices will be discussed as the temperature range could vary in actual commercial practice depending on the operation of the decorating lehr.

Results

The results of the FEA calculations are summarised in Tables I, II and III for ACL thicknesses of 25.4 microns, 38.1 microns and 50.8 microns, ▶

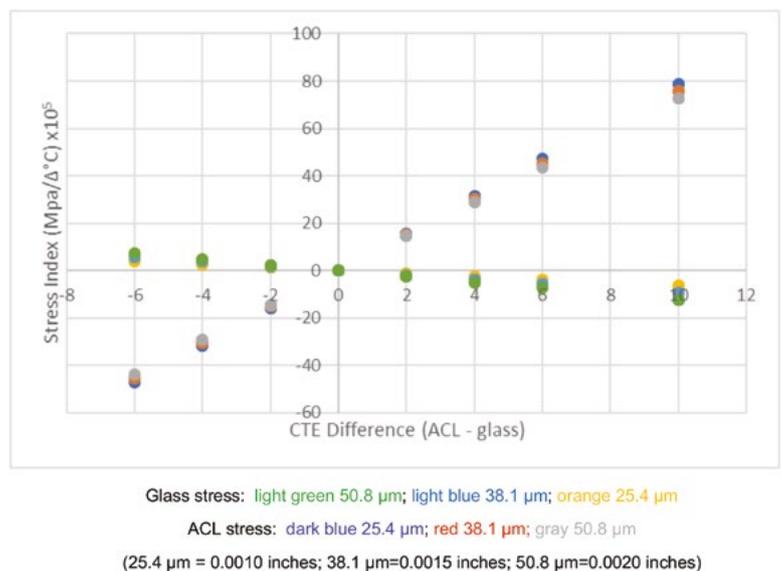


Figure 2: ACL and glass stresses for different ACL thicknesses (glass thickness = 1.27mm).

respectively. In each of these tables, the data are grouped into three sub-sections according to the CTE of the glass. When the CTE of the ACL and glass were equal, the results are highlighted in yellow.

As shown by the data in Table I, the overall magnitude of both the glass and ACL stress indices increased with increases in the difference between the CTE of the ACL and glass. This was consistent with previous studies.^{10,11}

The ACL stress indices were slightly lower for a glass thickness of 1.27mm compared to a glass thickness of 2.29mm. The glass stress indices showed the opposite trend. Similar results were obtained for ACL thicknesses of 38.1 microns and 50.8 microns, as shown in Table II and Table III, respectively. Also, as noted in Tables I, II and III, as the ACL thickness increased, the ACL stress indices decreased while the glass stress indices increased. All of these results are consistent with the thin layer theory of Hutchinson, et al.¹⁰ which states that stress in a thin layer increases when the thickness ratio of the thin layer and substrate decreases (a decrease in this ratio was observed in this study when either the ACL thickness decreased or the glass thickness increased).

Finally, the absolute magnitudes of the ACL stress indices were much greater compared to the stress indices of the glass. For a glass thickness of 2.29mm, the ACL stress indices were approximately 8 to 16 times greater compared to the stress indices in the glass. This difference was approximately 6 to 12 times for a glass thickness of 1.27mm. These differences were attributed to the response of the relatively thin ACL layer compared to the much thicker glass substrate when an equivalent strain is placed on the decorated surface.

The data in Tables I, II and III are graphically shown in Figure 1 and Figure 2 for glass thicknesses of 2.29 mm and 1.27 mm, respectively. The large differences in the ACL and glass stress indices are clearly seen. In addition, the nearly identical results for both the ACL and glass stress indices for various ACL thicknesses are also evident.

Discussion

The type and magnitude of stress that is generated in a glass substrate due to the presence of an ACL is

easily and conveniently analysed by use of suitable optical retardation instruments.^{8,9} It is normal practice to assume that the stress type in the ACL is opposite of that which is observed in the glass. Such an assumption is justified based on the differences in the CTE of the ACL and glass and how these two components contract when cooled from the ACL transformation point to room temperature.

The magnitude of the stress in the ACL, however, is an entirely different matter. Historically, it has been assumed that the magnitude of the stress in the ACL would be at least equal to and, since the ACL is much thinner compared to the thickness of the glass substrate, most likely numerically greater than the magnitude of the stress in the glass. Unfortunately, this assumption cannot be directly verified by polariscopic observations due to the opaque nature of the ACL. In the current FEA study, it was confirmed that the absolute magnitude of the ACL stress was numerically greater compared to the stress created in the glass. While this comparison has never been presented in the published literature, it is consistent with comments made by previous investigators who suggested that significant differences existed although no direct calculations or measurements were given.^{4,10,11}

The discovery of this stress

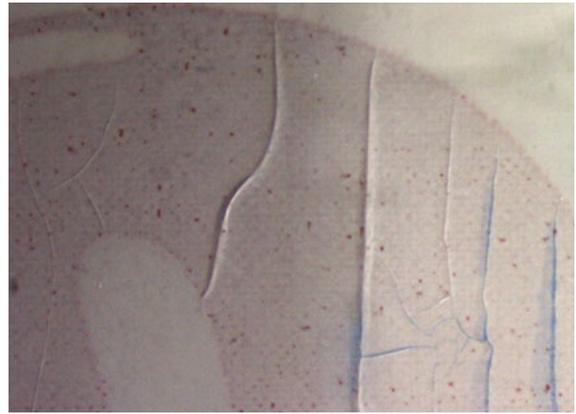


Figure 3: View from inside surface of the glass showing microcracks in an ACL decoration.

difference has two significant implications. First, these results confirm that large differences in the CTE of the glass and ACL can be tolerated with no adverse effects, provided the CTE of the ACL is less than that of the glass.⁴ Even if a large magnitude of compressive stress is present in the ACL, a much lower tensile stress will be generated in the underlying glass substrate. It is anticipated that the presence of only minor tensile stresses in the glass would have very little, if any, effect on the overall performance of the decorated item contrary to our initial concern.

Second, if the CTE of the ACL is greater compared to the glass, then relatively large tensile stresses will be generated in the ACL even though the magnitude of the compressive stresses that would be observed in the glass will be of moderate or even low magnitude. Such a condition could potentially cause breakage even though the magnitude of the measured stresses in the glass would not be alarming, as inferred by Andrews.⁴

A recent breakage problem is representative of this latter situation. A decorated, non-refillable bottle had failed ▶

ACL and Glass Substrate Stresses

(ACL thickness = 25.4 mm; glass thicknesses = 2.29 mm and 1.27 mm)

ACL Thickness (µm)	ACL CTE (°C) ⁻¹	ACL Stress Index [Mpa/Δ°C]		Glass Stress Index [Mpa/Δ°C]	
		t = 2.29 mm	t = 1.27 mm	t = 2.29 mm	t = 1.27
25.4	8.60E-06	-0.00032	-0.00032	0.00002	0.00003
	8.80E-06	-0.00016	-0.00016	0.00001	0.00001
	9.00E-06	0.00000	0.00000	0.00000	0.00000
	9.20E-06	0.00016	0.00016	-0.00001	-0.00001
	9.40E-06	0.00032	0.00032	-0.00002	-0.00003
	1.00E-05	0.00080	0.00079	-0.00005	-0.00006
25.4	8.60E-06	-0.00016	-0.00016	0.00001	0.00001
	8.80E-06	0.00000	0.00000	0.00000	0.00000
	9.00E-06	0.00016	0.00016	-0.00001	-0.00001
	9.20E-06	0.00032	0.00032	-0.00002	-0.00003
	9.40E-06	0.00048	0.00047	-0.00003	-0.00004
	9.80E-06	0.00080	0.00079	-0.00005	-0.00006
25.4	8.60E-06	-0.00048	-0.00047	0.00003	0.00004
	8.80E-06	-0.00032	-0.00032	0.00002	0.00003
	9.00E-06	-0.00016	-0.00016	0.00001	0.00001
	9.20E-06	0.00000	0.00000	0.00000	0.00000
	9.40E-06	0.00016	0.00016	-0.00001	-0.00001
	1.02E-05	0.00080	0.00079	-0.00005	-0.00006
CTE of glass and ACL are equal					

Table 1: ACL and glass substrate stresses (ACL thickness = 50.8mm; glass thicknesses = 2.29mm and 1.27mm).

at a very low level of applied force. Inspection of the fracture origins revealed that dwell marks were present indicative of the presence of micro-cracks in the decorated glass surface. These micro-cracks had significantly weakened the glass surface and were the sole cause of the breakage problem. Further examination of unbroken exemplar bottles revealed that the micro-cracks were present in the ACL on the as-produced bottles (see Figure 3).

Polariscopic analysis revealed the presence of a compressive stress in the glass with a magnitude of 28kg/cm² (400psi). Using the assumption that the tensile stress in the ACL was approximately the same magnitude, then it was difficult to fathom how 28kg/cm² (400psi) of tension would cause the ACL to spontaneously crack. However, applying the factors shown in the current FEA analyses, the tensile stress in the ACL would actually have been on the order of 168kg/cm² (2,400psi) to as much as 336kg/cm² (4,800psi) in magnitude. Such high tensile stresses are definitely sufficient to spontaneously cause micro-cracks to develop in the ACL layer as was observed in the breakage problem. ●

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ACL Thickness (μm)	ACL CTE (α) ⁻¹	ACL Stress Index [Mpa/Δ°C]		Glass Stress Index [Mpa/Δ°C]	
		t = 2.29 mm	t = 1.27 mm	t = 2.29 mm	t = 1.27
38.1	8.60E-06	-0.00031	-0.00030	0.00003	0.00004
	8.80E-06	-0.00016	-0.00015	0.00001	0.00002
	9.00E-06	0.00000	0.00000	0.00000	0.00000
	9.20E-06	0.00016	0.00015	-0.00001	-0.00002
	9.40E-06	0.00031	0.00030	-0.00003	-0.00004
	1.00E-05	0.00078	0.00076	-0.00007	-0.00009
38.1	8.60E-06	-0.00016	-0.00015	0.00001	0.00002
	8.80E-06	0.00000	0.00000	0.00000	0.00000
	9.00E-06	0.00016	0.00015	-0.00001	-0.00002
	9.20E-06	0.00031	0.00030	-0.00003	-0.00004
	9.40E-06	0.00047	0.00045	-0.00004	-0.00006
	9.80E-06	0.00078	0.00076	-0.00007	-0.00009
38.1	8.60E-06	-0.00047	-0.00045	0.00004	0.00006
	8.80E-06	-0.00031	-0.00030	0.00003	0.00004
	9.00E-06	-0.00016	-0.00015	0.00001	0.00002
	9.20E-06	0.00000	0.00000	0.00000	0.00000
	9.40E-06	0.00016	0.00015	-0.00001	-0.00002
	1.02E-05	0.00078	0.00076	-0.00007	-0.00009
CTE of glass and ACL are equal					

Table 2: ACL and glass substrate stresses (ACL thickness = 38.1mm).

ACL Thickness (μm)	ACL CTE (α) ⁻¹	ACL Stress Index [Mpa/Δ°C]		Glass Stress Index [Mpa/Δ°C]	
		t = 2.29 mm	t = 1.27 mm	t = 2.29 mm	t = 1.27
50.8	8.60E-06	-0.00030	-0.00029	0.00004	0.00005
	8.80E-06	-0.00015	-0.00015	0.00002	0.00002
	9.00E-06	0.00000	0.00000	0.00000	0.00000
	9.20E-06	0.00015	0.00015	-0.00002	-0.00002
	9.40E-06	0.00030	0.00029	-0.00004	-0.00005
	1.00E-05	0.00076	0.00073	-0.00010	-0.00012
50.8	8.60E-06	-0.00015	-0.00015	0.00002	0.00002
	8.80E-06	0.00000	0.00000	0.00000	0.00000
	9.00E-06	0.00015	0.00015	-0.00002	-0.00002
	9.20E-06	0.00030	0.00029	-0.00004	-0.00005
	9.40E-06	0.00045	0.00044	-0.00006	-0.00007
	9.80E-06	0.00075	0.00073	-0.00010	-0.00012
50.8	8.60E-06	-0.00045	-0.00044	0.00006	0.00007
	8.80E-06	-0.00030	-0.00029	0.00004	0.00005
	9.00E-06	-0.00015	-0.00015	0.00002	0.00002
	9.20E-06	0.00000	0.00000	0.00000	0.00000
	9.40E-06	0.00015	0.00015	-0.00002	-0.00002
	1.02E-05	0.00076	0.00073	-0.00010	-0.00012
CTE of glass and ACL are equal					

Table 3: ACL and glass substrate stresses (ACL thickness = 50.8mm).

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