# Formulation Of Lamination Conditions And Interface Studies Using Acrylic Binder System For LOM

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## Abstract:

A multicomponent binder-plasticiser system based on acrylates was formulated for making flexible tapes of Alumina (58 volume %), to be used for Laminated Object Manufacturing. Optimum lamination parameters were arrived at by viscosity measurement of the binder-plasticiser system and dilatometry of the compounded tapes . Lamination was done at five different temperatures (75, 85, 90, 95 and 110$^{\circ}$C) with solvent (toluene-butanol) spray and under three different pressures ( below 50 psi). The interlaminar strength was measured and interface studies were made using SEM .The shrinkage along the Z direction during lamination was in the range of 1.7-3.5%. Highest interlaminar strength and defect free lamination could be achieved at the intermediate temperature of 90$^{o}$C, due to sufficient reduction of polymer viscosity and presence of solvent.

## Introduction:

Laminated Object Manufacturing (LOM) is a layered manufacturing technique which is used to create solid 3D shapes of materials by sequential cutting and laminating 2D sheets or tapes of the same material. The principle behind this is essentially same with most of the other Rapid Prototyping techniques i.e. to fabricate components directly from CAD solid models .Ceramic tapes are made and cut in specific CAD defined contours in 2D and ultimately laminated to realise a 3D shape[1]. The materials issues are to make suitable flexible tapes as feed material, formulation of a thermoplastic organic system for the making of these tapes, formulation of optimum lamination conditions and interface studies, and finally binder burnout and defect free sintering.

The LOM technology has been successfully used in making complicated shapes of different ceramics (Alumina, Silicon Carbide, Silicon Nitride etc) and composites [2].While considerable reports dealing with the lamination process of Alumina and other ceramics are available [3] , not much details are available with respect to binder formulation, solids loading, viscosity of the polymer system and their effect on lamination conditions and formation of defect free good interface. LOM was tried to make shapes of alumina. We have formulated a specific binder-plasticiser system based on polyacrylates to make alumina tapes.The selection of the organic system was based on the thermal properties of the organic polymers. Our objective was to formulate a suitable lamination condition for LOM based on this binder system. Optimisation of lamination conditions by investigating viscosity of the organic system as a function of temperature, dilatometric studies for shape integrity during binder burnout, interlaminar strength and interfacial defect elimination are reported here.

## Experimental:

A tape casting slurry of Alumina ( ACC-India A-16 grade, average particle size:0.6 microns, specific surface area:8-10 sq.m/gm) was made using an acrylate binder and polythelene glycol and benzyl butyl phthalate as plasticiser in toluene- butanol solvent (in azeotropic ratio) medium using gleceryl trioleate as dispersant. The alumina powder dried at 100-110°C for 30 minutes was mixed with the dispersant which was disssolved in the solvent system along with 0.25 %by weight talc used as grain growth inhibitor. It was then milled in a pulverisette ( speed 144 rpm)with high purity alumina as grinding medium. for 3 hours. Then the binder and the plasticisers were added to form a viscous slip. Once again it was subjected to milling in the pulverisette for 3 hours. The slip was vacuum degassed and sieved through a nylon mesh to remove any large particles or undissolved binder, if present. The viscosity of the slip was measured to be 1790 cps in a Brookfield viscometer (DV-II type, spindle no. 25) at 60 rpm. It was then cast in a batch type double-blade caster (casting rate is 10 cm/min) onto a silicone coated polyester film . Then the tape was subjected to drying for 10 hours, the tapes of thickness 0.55mm were peeled off the film and cut to desired shapes for subsequent measurements. The solid loading was 58% by volume with respect to the total organic binder content after solvent removal.

Table 1: Slip formulation used for Alumina tape casting

| **Constituents** | **Materials** | **Volume %** |
|---|---|---|
| Ceramic Powder | Alumina | 29.52 |
| Solvent | Butanol-Toluene mix | 49.14 |
| Binder | Polymethyl Methacrylate | 9.08 |
| Plasticiser I | Polyethelene Glycol | 3.50 |
| Plasticiser II | Pthalete | 5.26 |
| Dispersant | Gleceryl Trioleate | 3.50 |

The viscosity of the binder-plasticiser-dispersant system was measured in rotational viscometer (Brookfield Viscometer DV-II type, spindle no. 32 ) at a temperature range of 35-100°C at 8 different temperatures and at different rpm's at each temperature. Lamination was done using solvent spray [4] and by the application of pressure and temperature. Tapes were cut into rectangular pieces of 1cm × 1 cm . Lamination was done at 5 different temperatures (75, 85 ,90 ,95, 110°C) and under three different pressures (1.75 , 2.2, 3.0 kg/sq.cm). Two tapes of the said dimension were joined together after solvent spraying and then placed in an apparatus fabricated in-house for lamination. The tapes were kept in the particular pressure-temperature condition for 10 minutes. The change in the thickness of the laminates parallel to the pressure axes ( the z direction) after lamination were measured

The interlaminar strength was measured using an arrangement as shown in fig.1.Two laminated tapes, after lamination using different pressure-temperature conditions were used. Each face of the laminate were stuck to two identical rectangular brass plate assemblies using a two component epoxy resin. The brass plate assembly (fig.1) was hung and the load applied. The

particular load at which delamination occurs was noted. The laminate interfaces were studied using SEM (S360 Cambridge Scanning Electron Microscope) after gold plating.

Dilatometric measurements were made in a laminated sample (height 4.85 mm). The sample was heated to 800°C at 2°C/min in the dilatometer and then cooled to room temperature.

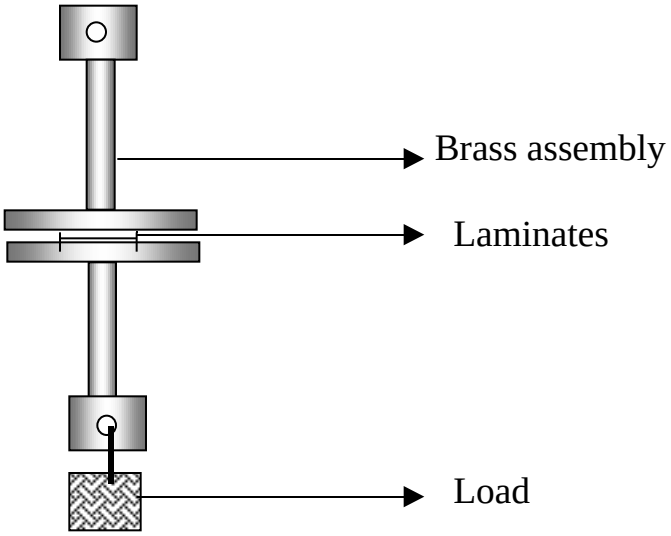



Fig 1: Experimental arrangement for interlaminar strength measurement

## Results and Discussions:

Flexible tapes of uniform thickness (0.55 mm ) were obtained after casting followed by drying. The tapes had a good surface finish and were easy to handle. The loss on ignition showed the total organic content was 16.48%, which corresponds to 41.96% by volume of the starting formulation without the solvent.

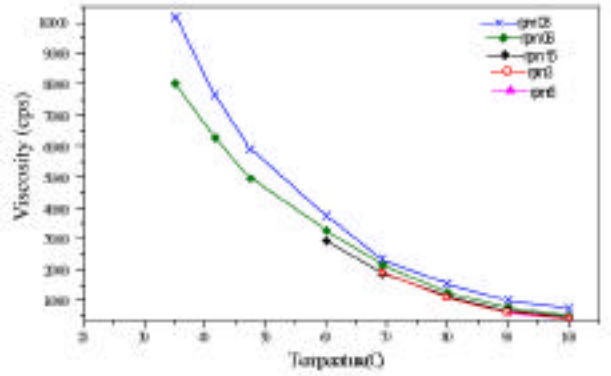



Fig 2: Variation viscosity of the organic binder-plasticiser system with temperature at different shear rates

The viscosity data of the organic components show that viscosity reduces sufficiently after 60C (shown in fig.2). Viscosity comes down most significantly in the range 75-100°C, and after 100°C viscosity came under the instrument detectable range. Also the viscosity shows increase with decreasing shear, which implies pseudoplastic behaviour of the organic system. From the viscosity data lamination temperature range was fixed to be between 75-110°C.

Attempt was made to find out the viscosity of the compounded ceramic-polymer mix using a capillary rheometer (CFT-500C type, Shimadzu Corporation) with capillary diameter 1mm, but no signifiacant sagging and softening was observed till 200°C and a load of 500 KN.

The lamination was to be done on green ceramic tapes, so the pressure to be applied depended on the allowable dimensional change of the laminate after laminating and retention of shape of the green tape. After laminating at different lamination pressure-temperature conditions it was found that the dimensional change along the pressure axis (Z axis) was between 1.7-2.6% without deshaping of the laminates ( Table 2). With increasing pressures the dimensional change values increased, so the pressure range for lamination was chosen between 1.75-3.0 kg/sq.cm.

Table 2: Room temperature Interlaminar strength and percentage shrinkage after lamination of the laminate as function of lamination temperature and pressure:

| Pressure / Temp. (°C) | 1.75 (kg/sq.cm) | | 2.2 (kg/sq.cm) | | 3.0 (kg/sq.cm) | |
|---|---|---|---|---|---|---|
| | Laminar Strength (kg/sqcm) | % shrinkage | Laminar Strength (kg/sq.cm) | % shrinkage | Laminar Strength (kg/sq.cm) | % shrinkage |
| 75 | 1.64 | 1.73 | 1.66 | 1.75 | 1.74 | 2.01 |
| 85 | 1.92 | 1.92 | 1.94 | 1.99 | 1.96 | 2.21 |
| 90 | 2.12 | 2.15 | 2.42 | 2.27 | 2.68 | 2.58 |
| 95 | 2.02 | 2.12 | 2.18 | 2.17 | 2.32 | 2.31 |
| 110 | 1.81 | 1.89 | 1.87 | 1.93 | 1.91 | 1.99 |

The interlaminar strength data (Table 2) showed that the strength was maximum at the temperature 90°C for all applied pressures. The strength increased with temperature till 90°C and then decreased . With increasing the pressure during lamination the strength data shows increase for all the temperature conditions. Strength was found to be highest at a pressure of 3.0 kg/sq.cm. The SEM figures (at three different temperaures at a load of 3.0 kg/sq.cm) also showed the best interface was observed in the pressure-temperature condition of 90°C and 3.0 kg/sq.cm (Fig. 4 b). Both at higher and lower temperatures showed clear clear delamination (Fig 4a, 4c). At lower temperatures the organics did not soften enough to give good lamination even at higher pressures. At temperatures above 90°C due to evaporation of solvent ( azeotropic b.p is 105°C)

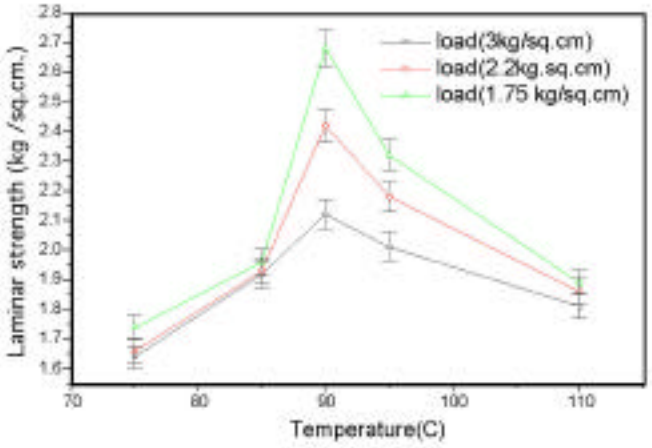
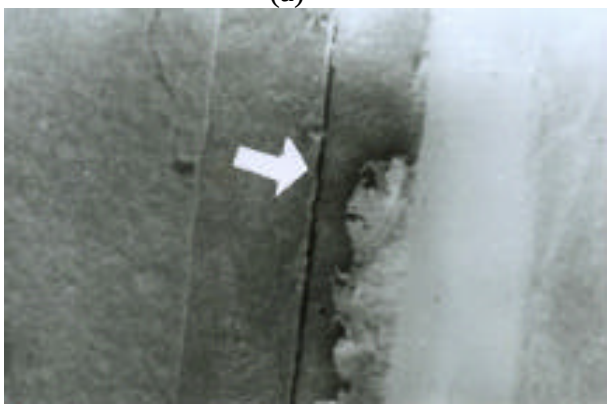
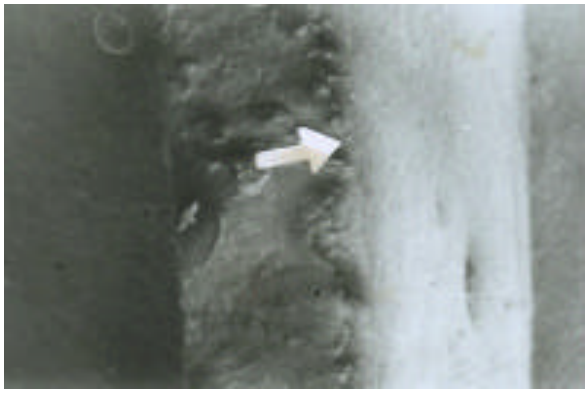
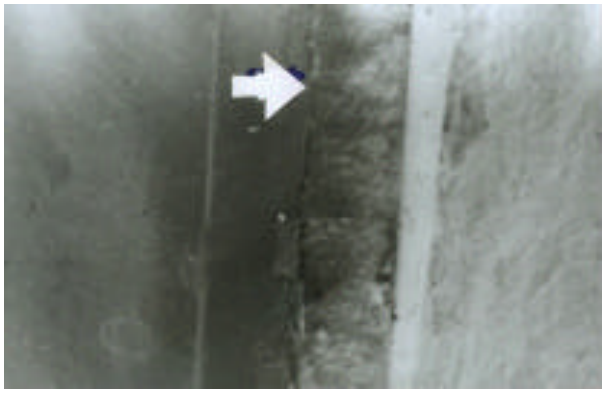



Fig 3:Interlaminar strength Vs lamination temperature at different lamination pressures

(a)







(b) 500µm (c)

Fig: 4 :SEM micrograph of interfaces obtained at 3 kg/sq.cm and at different temperatures (a):110˚C (b): 90˚C (c):75˚C

lamination was poor. Though both the strength data and SEM studies showed with increasing pressure during lamination ensures better interface, but due to the factors like deshaping of the tapes and significant dimensional changes during lamination the pressure was limited to the value of 3.0 kg/sq.cm.

The result of the dilatometric measurements of a laminated (at 90°C and 3.0 kg/sq.cm) sample of height 4.85 mm is shown in Fig.5. There is no significant dimensional change or shape loss in the sample till 450°C. On further heating the contraction due to the binder softening and decomposition is observed. During cooling the sample showed the usual contraction of a porous ceramic sample ( $\alpha = 4.2 \times 10^{-6}$/K). This shows that our binder formulation is suitable for making complex ceramic shapes without need for creating support structures.

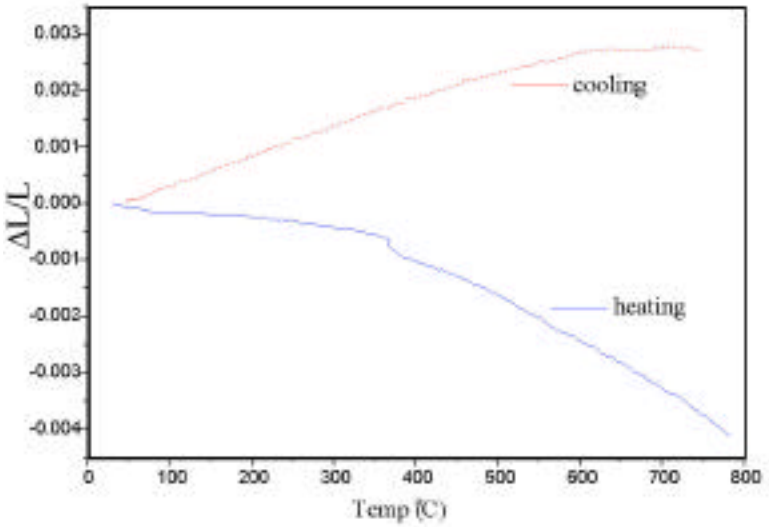



Fig 5: Thermal strain of the laminated alumina body as functionof temperature during heating and cooling

The binder burnout studies, retention of net shape and studies on laser cuttability of green ceramic tapes are currently under progress.

## Summary and Conclusion:

Flexible alumina tapes were produced an acrylic binder and a non-aqueous solvent system. Based on the viscosity of the organic system at different temperatures lamination temperature was chosen. Lamination was done using solvent spray and under applied load. Optimum lamination conditions were arrived at by measuring the interlaminar strength and studying the interface using SEM. Defect free strong interface was achieved at a temperature of 90°C and under a load of 3.0 kg/sq.cm. It was seen that with increase of load interface was better but dimensional change in the Z direction was more with increasing load. At temperatures less than 90°C the organic system was not soft enough to give good lamination and at temperatures higher the sprayed solvent evaporates as its boiling point is 105°C.

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