Styrene-acrylic latex binder for room temperature lamination of ceramics green tapes

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Lamination of green tapes is an important process in the manufacture of layered ceramic structures, such as multilayer capacitors, inductances, substrates, etc. [1–3]. Furthermore it has also become important for structural applications, such as heat exchangers, solid oxide fuel cells (SOFC) and for rapid prototyping by Laminated Object Manufacturing (LOM). The basic method used for lamination is the thermo-compression method, in which the different layers are stacked and pressed at a temperature above the adhesive's glass transition point, where the adhesive was pre-coated on the surface of green tapes. A new method allows lamination at room temperature under low or slight pressures and without any adhesive on the surface of green tapes, because the green tape can be adhered by the pressure sensitive adhesion of latex binder when the T_g of the latex binder is below room temperature. Furthermore, good bonding properties of the laminated sheet material ensure good adhesion in the interlayer and avoid de-lamination during organic removal or sintering.

Tape casting is the most widely used technique for fabrication of green sheets for LOM. In general, tape-casting slurry is a typical organic system. Because the volatility and toxicity of organic solvents are harmful to the environment and health, water-based tape casting is widely accepted by the ceramics research domain [5–9]. Kristoffersson [4] and her colleagues studied the solid volume fraction, green density, laminated properties of alumina ceramics by water-based tape casting with a pure acrylic latex binder system. They had fabricated good quality tapes and laminated specimens when the T_g of the latex binder was below room temperature.

The focus of this paper is to introduce a new method of lamination at room temperature, and to study the mechanism of lamination as a function of fraction of binder and plasticizer.

The alumina powder used in this experiment had a median size of 0.8 μm (produced by ALCOA). The binder of aqueous tape casting process was a commercial styrene-acrylic latex that has low foam and viscosity to produce flexible green tapes with good mechanical properties and lamination behaviors. It was a nonionic latex, stable at pH 7. The latex was a dispersion of fine latex particles of $d_{50} \approx 300$ nm and solid loading was about 50 wt%. The glass-transition temperature (T_g) of the latex was about 273 K, which allows it to form a film at room temperature. The slurry compositions with forming-film additive fraction, latex binder fraction and plasticizer (glycerol) fraction were given in Table I.

Tape-casting experiments were carried out on a laboratory tape caster. This device enables the production of tapes of 120 mm width and 1000 mm length by fixing a container with two blades. Micrometer screws allowed the gap of the blades to be from 100 up to 1000 μm, and the gap was set to be 700 μm, and the casting speed was about 10 mm/s in this experiment. The green tape was stripped from the support polyester film after casting for about 5 hrs.

The adhesive strength of dried green tapes was tested by a 180° stripping experiment [5] as shown in Fig. 1. The green tapes were cut into some strips of 10 mm × 200 mm. First, we fixed a strip on the platform with the strong adhesive, and laminated another strip on the first strip, then compressed naturally three times with a steel roller whose weight is usually about 2.0 kg. Finally, we stripped the upper green tape at the speed of 5 mm/s and noted ten values of F in the effective range about 75 mm. The adhesive strength was calculated using the following formula:

$$\sigma_{180^\circ} = H/B \qquad (1)$$

where H is the average value of F(N), B(m) is the width of the green tape.

Latexes act as a binder and form films by drying when the water phase evaporates. Finally; the polymeric particles coalesce and form a polymeric network. The drying process was one of the most important steps in tape-casting processing. It enabled to obtain a solid tape from a liquid slip. The forming-film conditions of the latex binder system should include: (1) the compressive force for latex particles results from solvent evaporation proceeds, and (2) the glass transition temperature of latex is lower than work temperature [5–6].

Preliminary lamination tests on green tapes had showed that the polymer latex content of 20 vol% was not enough to obtain adhesion strength between layers,

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TABLE I The compositions of aqueous suspensions andσ_{180° of green tape

Sample	Alumina fraction (vol%)	Latex binder (vol%)	Plasticizer (vol%)	Water+other additive (vol%)	σ_{180° (N/m)
1#	43	20	2	35	About 0
2#	43	30	3	24	20
3#	43	40	4	13	90
4#	43	40	0	17	About 0
4#	43	40	8	9	Tape destroyed

Note: the vol% of latex binder is the complete emulsion including water, and the fifth column does not include the water in emulsion.

Figure 1 The measurement of adhesive strength between green tapes by 180 ° stripping.

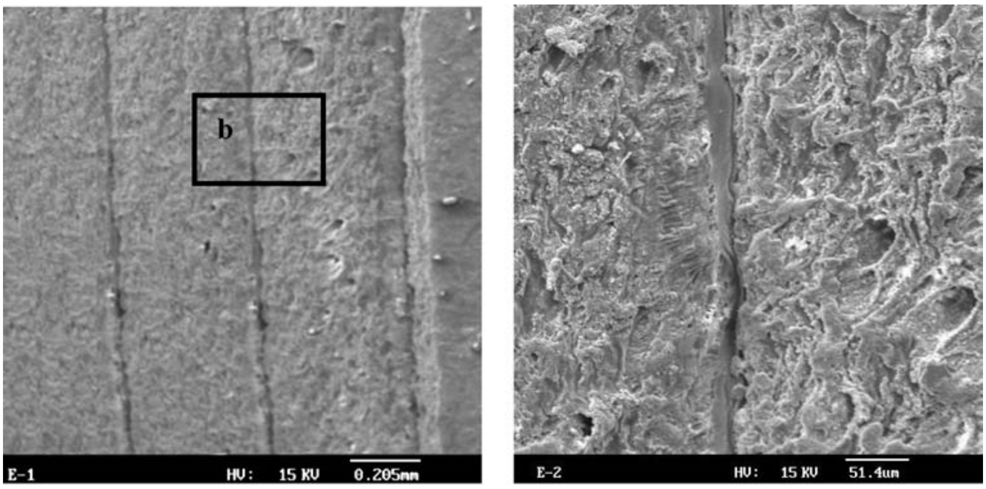
even if the pressure was very large (excess 50 MPa). The laminated specimens which held homogeneous interface microstructure were fabricated by 3# green tape in suitable pressure (the laminated pressure value was decided by experiments), as shown in Fig. 2.

If the T_g of latex was located in the range of 213–253 K, the adhesive strength of the latex binder would be the most suitable for pressure-sensitive-adhesion binder [5]. In this experiment, the T_g of the latex binder was only 273 K; therefore, the adhesive strength of latex was not strong enough so that it could act as the pressure-sensitive-adhesion binder. But the latex still possessed considerable adhesive force, which enables us to laminate the green tape on specific conditions when adding plasticizer to decrease the T_g of the latex binder.

The experimental results showed the relationship between adhesive strength and latex binder fraction, the ratio of plasticizer to binder in suspensions. As shown in Table I, the adhesive strength of green tapes increased with the increasing of latex volume fraction at a fixed ratio of the plasticizer. But the adhesive strength was about 0 between green tapes when the latex fraction was less than 20% of entire suspensions. Therefore, a guarantee of laminated green tapes was that the fraction of this latex binder should be higher than about 30 vol%; for results refer to samples 1#, 2#, and 3# in Table I.

Mixing plasticizer into latex in aqueous suspension is a valid way to decrease the T_g of the latex system. The plasticizer is a low molecular weight and in-volatile composite; therefore, the plasticizer can reduce the mutual action among the polymer chains so as to improve the flexibility of polymer chains. The reason is that the plasticizer remains in green tapes after drying, which decreases the T_g of the latex binder system. Glycerol was a good plasticizer for this latex binder system. Adequate content of glycerol could increase the flexibility of green tapes, and improve the adhesive strength between green tapes. When the ratio of glycerol to latex binder exceeded 1/5, the adhesive strength of the green tapes would become strong but the tensile strength of green tape would decrease with excessive amount of plasticizer. Therefore, the stripped experiment result was the green tapes were firstly destroyed by tensile force, as shown in 5# of Table I.

In short, the experiment results showed that three factors decided the pressure-sensitive-adhesion property of the latex binder at room temperature: (1) the glass transition temperature of latex should be very low; (2) the content of latex binder in suspensions should be high; (3) the suitable ratio of glycerol plasticizer to this latex binder in slurry.



(a) (b)

Figure 2 (a) The interface of laminated green part by 3# tapes and (b) the amplificatory portion in (a) photograph of interface.

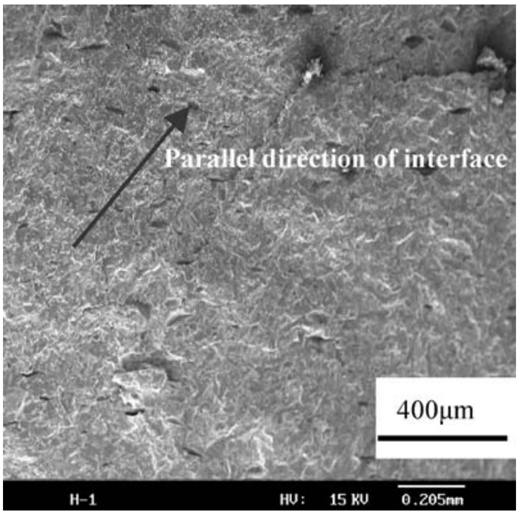


Figure 3 The fracture surface of sintered lamination sample by 3# tapes.

The laminated green part was sintered at a heating rate of 10 °C/min to 1600 °C in air after binder removal, and held the 2 hrs. Although the sintering rate was relatively fast, the laminated green sample had not developed delamination in the sintering process. Especially, the sample that was laminated by 3# tapes, showed a higher sintering relative density of 97% theory density. As shown in Fig. 3, a little pore is present in the matrix of sintered bodies, and the interface was almost disappearing between two layers.

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