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Aqueous Tape Casting Of Ceramic Powders: A Review

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ABSTRACT: Slurry formulations and water based tape casting parameters of ceramic powders are reviewed. Additives include binders, such as polymers of the cellulose ethers, vinyl or acrylic type, plasticizers such as glycols and dispersants such as polyacrylic ammonium salts. Alumina powders were mostly used. Hydrophobic ceramic powders require even water reactive powders, such as aluminum nitride, to be processed aqueously. An alternative to organic solvent based system is the non-toxicity and non-inflammability of water based systems. An alternative to organic solvent based systems is the non-toxicity and non-inflammability of water based systems .on the other hand, aqueous slurries are complex multiphase systems, which are very sensitive to variations in processes. The mathematical design of experiment was used for process improvement. Tape casting is a well-established technique that is used to make large scale ceramic substrates and multilayered structures. A slurry consisting of the ceramic powder contained in a solvent, together with dispersants, binders and plasticizers is cast onto a stationary or moving area .in particular, the research aim in aqueous tape casting must be to make the product insensitive to environment variables, product deterioration and manufacturing imperfections. This technique can also be achieved by using robust design experiment.

KEYWORDS*:*Ceramic Powders, Tape Casting, Slurries

I. INTRODUCTION

Tape casting has been a well-established method used to make ceramic substrates and multilayered structures on a massive scale. Slurry comprised of the ceramic powder contained in a solvent, together with dispersants, binders and plasticizers, is cast onto a stationary or moving area. The cast tape would then be dried and eventually sintered to get a desired final shape with a standard thickness in the range of 100-250 µm. A variety of non-aqueous organic solvents, such as alcohol, ketones or hydro carbons are used for preparing highly concentrated suspensions with reproducible rheological properties and drying behavior depending on the composition of the ceramic materials [1] .

In recent times considerable attention has been paid to the environmental and health implications of the tape casting process. Furthermore, slurry formulations have appeared in the literature using water as a solvent instead of organic liquids. Non-aqueous solvents have low boiling points and prevent hydration of the ceramic powder, but require special toxicity and inflammability measures. Organic solvent recovery systems are usually required for controlling compound emissions into the atmosphere. At the other hand, an aqueous system seems to have advantages of incombustibility, non-toxicity and low cost, associated with the large amount of experience using water in similar ceramic powder processes, such as slip casting. Moreover, other colloidal processing processes, such as those used in paint or magnetic tape production, have, due to safety considerations, shifted from organic to water based systems [2].

A tape casting slurry must be calibrated to produce tapes that meet certain quality requirements, such as (i) no drying defects; (ii) stability to allow dried sheet manipulation; (iii) micro structural homogeneity; (iv) strong thermo compression (lamination) capability; (iv) fast pyrolysis (burnout); and (v) high mechanical strength after sintering. It requires careful selection of the slurry additives along with precise monitoring of many parameters for processing.

Notable differences between non-aqueous and aqueous tape casting apply to perturbation sensitivity to operation. An organic solvent-based slurry is much more volatile and irritating to process but it is easy to achieve solid, uniform green

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bands. Aqueous slurry has less resistance to minor changes in drying, casting composition, or film thickness conditions. Only when all variables are controlled extremely well, it produces crack-free, uniform green tapes. The goal of such a work is to examine the efforts made for the tape casting process to establish aqueous systems as a reliable alternative to organic solvent based systems [3].

II. METHODOLOGY

Slurry formulation:

The range of the water soluble binders, plasticizers and dispersants is limited to a few system compared with non-aqueous solvents. Table 1 describe the combination used to cast alumina and mullite with aqueous tape.

For the formulation of a tape casting slurry, some basic rules may be inferred: (i) the ratio among organic components and ceramic powder should be as low as possible (ii) the amount of solvent must be set the minimum to ensure homogeneous slurry (iii) the volume of dispersant must be the minimum needed to ensure the consistency of the slurry (iv) the ratio of plasticizer to binder must be set to tape flexible, resistant and easy to release [4].

Table.1: Aqueous Slurry Additives for Tape Casting

Powder	Binder	Plasticizer	Dispersant
Alumina +MgO	Acrylic Polymer	PEG+BBP	Condensed aryl sulfonic acid
Alumina +talc	PAA	Glycerol	NH_4PMA + Dispex A 40 NH_4PM + Dispex A40
Alumina	PUR PVA PVA_c NH_4PA	Glycerol Glycerol Glycerol+DBP	POENPE POENPE POENPE
Alumina	Cellulose		NH_4PMA

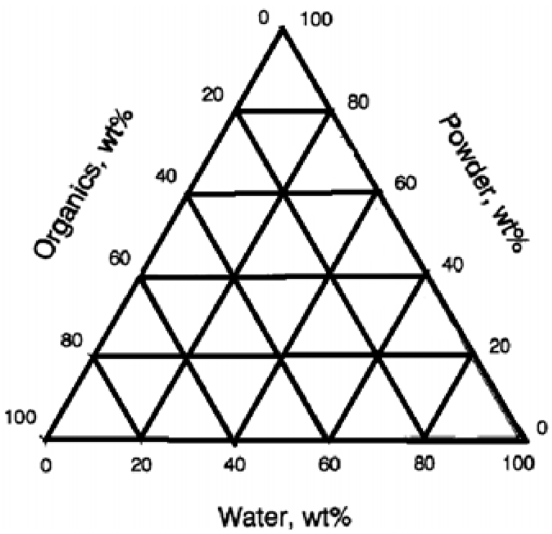

Fig.1: Aqueous Slurry Formulation for Tape Casting

A compilation of the aqueous slurries compositions is illustrated in Figs. 1 & 2. The top of the triangle in Fig. 1, according to the first two criteria mentioned above. The goal to be reached should be. In other words, the minimum amount to produce slurry with acceptable properties should be used in water and in organic additives. Ceramic powder charges range from around 25% to nearly 80%. The organic additives are always above 18 wt. percent, whereas the

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water content varies from less than 20 to 70% of wt. As shown in Fig.2, the organic components were used in very different ratios [2][3].

Nonetheless, this definition isn't absolute: some additives have multifunctional features. For example, a certain binder can exhibit plastizing or dispersing effects. In the slurry types, two investigators use neither dispersant nor plasticizer. In the other situations, the binder refers to at least half the organic component of the slurry. In any case, the dispersant content would be the lowest of all three organic additives (< 20% of wt.). In general, plasticizers were used up to about 50 wt. each.

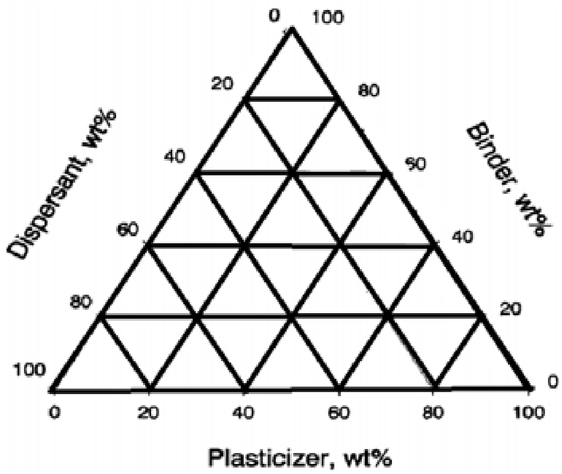

Fig.2: Organic Additive Formulations Used in Aqueous Slurries for Tape Casting

Solvent:

The solution dissolves the organic materials and is evenly dispersed throughout the slurry. It is the vessel that holds dispersion of the ceramic particles until it evaporates and leaves a thin tape on the container. A non-aqueous suspension dries easily and provides high density green sheets and a fine appearance on the surface. An aqueous suspension has the drawbacks of high latent heat evaporation and poor drying characteristics and many quality issues need to be resolved.

It has been made a comparison of aqueous and non-aqueous slurries for tape casting to assess the effect of changing solvent systems on the processing of slurry and on the consistency of green tape. The alumina powder was used, and the other slurry components and processing parameters were kept stable. Green bands from both systems had similar physical properties, although the aqueous systems were more prone to process disturbances [5].

Powder:

To increase reliability in ceramic processing, and especially in aqueous tape casting, a well-characterized powder is needed. The powder must be of a small particle size to achieve effective particle packing. But, the smaller the particle size, the higher the actual surface area which is not convenient because there are higher tape shrinkages and higher additive concentrations are required. Alumina powder is widely used. Most of the time, it is used with addition of the grain growth inhibitors.

The incompatibility with powders resistant to hydration, such as CaO or MgO, should be considered to be a disadvantage of the use of water based systems in tape casting. This can, however, be solved by hydrophobic ceramic powders. This technique allows water-reactive powders to be processed in aqueous media [6].

Binder:

After the solvent evaporates into organic bridges between the ceramic particles, the binder gives power to green tapes. The tapes can be manipulated easily before sintering and retained in the desired shapes.

Organic binders are dissolved as an emulsion, or spread in water. Long chain polymer molecules are the most soluble binders. The molecule backbone consists of covalently bonded atoms, including carbon, oxygen, and nitrogen. They are attached to the backbone are side groups often positioned along the length of the molecule at frequent intervals. In addition, the chemical structure of the side groups determines which liquids the binder can dissolve. If the side groups are strongly polar, water solubility is encouraged. The binders ' polymeric molecules consist of fewer components, the monomers. The number of monomers in a polymer is called the polymerization degree, DP. The number of sites on

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which a monomer is changed is called the degree of substitution, DS. The molar ratio between side groups and one monomer unit is called the average molar substitution degree, MS [7].
In aqueous tape casting of ceramics two classes of substances were primarily used as binders: cellulose ethers and polymers of vinyl or acrylic form. Cellulose is a naturally occurring polysaccharide formed by monomers of the ring type with a modified glucose structure. Fig. 3 displays the structure of a molecule of the cellulose type; it is depicted as a polymeric chain formed with a number n of cellulose units.

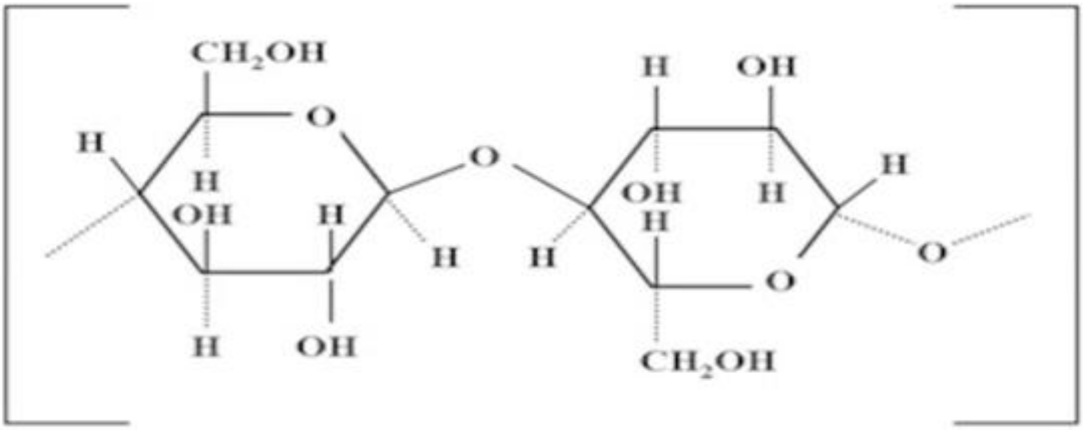

Fig.3: Structure of the Cellulose

The latter composed of two units of anhydroglucose. Each anhydroglucose ring has three free hydroxyls, which can be replaced through chemical reactions by different side groups.
Dividing the cellulose ethers into ionic and non-ionic types is a common practice. Ionic cellulose ethers such as NaCMC contain electrical-charged substituents and are used as polyelectrolytes instead. Unlike MC and HEC, non-ionic cellulose ethers bear no charge and are primarily used as binders. In the group, whose character predominates, copolymers are ordered with ionic and non-ionic substituents. Non-ionic cellulose ethers are further subdivided because of their differing solubility: for example, MC is soluble in cold water; HEC is soluble in both cold and warm water.
The general formulation for additives of the vinyl kind is given in Fig. 4. The vinyls are distinguished by a linear backbone composed of carbon-carbon bonds, with a side group attached to every other atom. When two side groups are attached to the carbon atom it is called acrylics. Some additives of the vinyl and acrylic type used in the processing of aqueous tape casting are given in Table 2. These may also be subdivided into ionic polymers and non-ionic ones. The former are the poly ammonium salts (acrylic acids), acting as poly-electrolytes. The latter are used as binders, instead

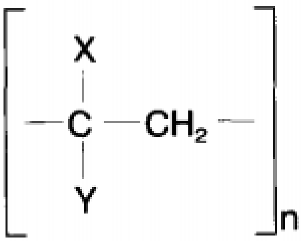

Fig.4: General Formula of Vinyl and Acrylic-Type-Derivative

Table.2: Vinyl and Acrylic-Type Polymers Used in Aqueous Tape Casting

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Compound		Side group	
		X	Y
PVA	Poly vinyl alcohol	-H	-OH
PVAc	Poly vinyl acetate	-H	$-OOCCH_3$
PVP	Poly vinyl pyrrolidine	-H	$-NC_4 H_8$
PAA	Poly acrylic acid	-H	-COOH
PMAA	Poly methacrylic acid	$-CH_3$	-COOH
PMMA	Ply methyl methacrylic acid	$-CH_3$	$-COOCH_3$

Plasticizer:

Plasticizers are additives that soften the dry or semidried binder. Compared with binders, they are organic substances of low molecular weight and are soluble in the same liquid. Binders and plasticizers are closely combined after drying. The plasticizer prevents the close coordination and bonding of the binder molecules, thereby increasing the tape's strength and workability. The plasticizer tends to reduce the strength while softening the binder.

The plasticizer's most important effect is to reduce the temperature T_g of gel forming, at room temperature or less. Tapes with gelled liquids dry much slower because the liquid does not float to the surface during the drying process. Within the gel structure, water has to leave the body through diffusion. One advantage of a gelled system is that the binders will not nigrate to the drying surface. It would be if the moving liquid brings this down there as it falls to the surface.

After casting, the binder plus plasticizer system cannot cling strongly to the casting surface, and must decompose without leaving residues. There is also an optimal benefit of versatility, obtained by choosing the appropriate binder / plasticizer device and changing the relative concentrations properly. If the concentration of the plasticizer is gradually increased to improve durability, the porosity might decrease until the pores vanish. Therefore, the interparticle distances will improve and the green density would decline [8].

III. PROCESSING AND EQUIPMENT

Milling and mixing:

Most of the aqueous tape casting processes recorded is conducted using a two-stage milling / mixing technique. The first stage refers to the milling phase in which a low viscosity slurry is prepared, consisting of water, dispersant and powder. Agglomerates are dissolved during milling, and dispersants are evenly distributed on the ceramic particle surfaces. In the second stage, there is mixing and homogenization in which in the aqueous slurry plasticizer and binder are dissolved.

Some modification has been specified in that standard procedure. Several investigators carried out a one-step milling with all of the components. In the first stage other researchers have already incorporated the plasticizer. According to previous research with non-aqueous slurries, the order of attachment of the components is important although the active mechanisms remain partly unexplained. There has been no clear research on the aqueous slurries [9].

Casting and drying:

Relative movement between a "doctor blade" and a support is achieved by casting tapes. There are two possible solutions: either the blade moves over a fixed support, or the support moves under a fixed blade. The first method is discontinuous and typically used in manufacturing on a small scale or for laboratory operations. The latter is a continuous one, used in most tape casting processes on a production scale [10].

IV. CONCLUSION

The alternative to the common non-aqueous tape casting is the use of water-based systems. Nontoxicity and non-inflammability seem especially beneficial. Nevertheless, all of the evidence previously reported indicates that many aspects of the aqueous tape casting process have yet to be understood. The use of additives arose from empirical observations, rather than from an understanding of the physicochemical processes occurring on the surface of the particles and their interactions.

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Statistically formulated experiments are a powerful tool in ceramic processing development. In particular, the research goal in aqueous tape casting should be to make the product resistant to environmental variables, product degradation and manufacturing imperfections. Use robust design experiments, the approach can be achieved.

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