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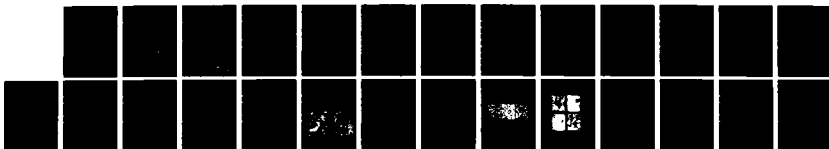
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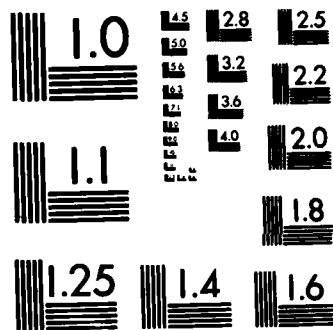
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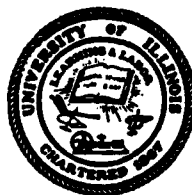
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EFFECT OF COPRECIPITATION PARAMETERS
ON POWDER CHARACTERISTICS
AND ON DENSIFICATION OF PZT CERMAMIC

by

R. C. Buchanan and J. Boy

September, 1982

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Effect of Coprecipitation Parameters on Powder Characteristics
and On Densification of PZT Ceramics

by

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Abstract

Effects of coprecipitation parameters on agglomerate size distribution and XRD crystallinity of PZT (53:47) powders obtained from butoxide precursors, were studied. Parameters found to significantly affect the coprecipitation process included: dilution, pH and coprecipitation rate (controlled by H_2O added to cause exothermic hydrolysis of the butoxides). Effects of rinsing the precipitates with different concentrations of isopropanol/water solutions were also investigated. Results showed that relative agglomerate size distributions developed during coprecipitation persisted throughout subsequent processing. Sintered densities and microstructures were correlated to the agglomerate size distributions. Most compact and uniform microstructures were obtained with uniform (and smaller) agglomerate distributions and 60/40 isopropanol/water rinsing.

I. Introduction

Commercial PZT ceramic powders are formed mainly from calcined mixtures of oxide powders. Difficulties with mixing often lead to local compositional variations with resultant inhomogeneities in the microstructure of the sintered ceramic. This condition in PZT contributes to lack of reproducibility in the electrical and aging characteristics of the sintered ceramic and has the effect of limiting PZT use for the more critical transducer applications.

Alternate methods for powder preparation, including liquid chemical, sol gel, spray drying, and cryogenic techniques have been developed. These achieve intimate mixing through coprecipitation or suspension of the oxide constituents as submicron particles in a finely dispersed and highly reactive state. Subsequent washing, drying, and calcination of the precipitates produce the desired ceramic powders.

Chemical coprecipitation has been perhaps the most widely used of these techniques. In preparing PZT ceramics, the alkoxide precursors of Zr and Ti are preferably used, reflecting difficulties with the use of Ti-nitrate and undesirable side effects with sulfate and chloride precursors. Brown and Mazdiyasni¹ used alkoxide-derived PZT powders prepared from Zr and Ti alkoxide solutions blended with a solution of lead isoamyl oxide in distilled water to bring about decomposition and precipitation of the PZT powder. Excess lead oxide in the form of $Pb(OR'')_2$ was added to compensate for Pb loss during sintering. Calcination of the powder at $\sim 550^\circ C$ for 30-60 minutes produced the PZT phase.

Haertling and Land² prepared high-purity PLZT powders from materials consisting of lead oxide (PbO), lanthanum acetate ($La(AC)_3$), zirconium tetra-n-butoxide (ZNB) and titanium tetra-n-butoxide (TBT). The procedure consisted of weighing out the proper amounts of PbO and butoxides followed by

blending in isopropyl alcohol. Lanthanum acetate solution was added from a plastic squeeze bottle to the blending mixture. This hydrolyzed the butoxides and produced a precipitate of mixed hydroxides. The process was accelerated as a result of the excess heat produced by the exothermic reactions.

Modifications to the basic alkoxide process for producing coprecipitated PZT and PLZT powders with different characteristics, have been described by many authors.³⁻⁶ The effect of subsequent processing steps—washing, drying, calcination, dispersion, and pressing—on powder characteristics and final densification have also been explored.^{5,6} As expected, these steps have been found to significantly influence the final densities achieved.^{7,8} Most important, was the characteristics of the precipitated powders with respect to agglomerate sizes and distribution, and this condition persisted throughout the subsequent processing steps.⁷ Methods for controlling agglomerate sizes during the coprecipitation process have not, however, been well-characterized.

Coprecipitation process has also been used for adding minor ingredients to PZT compositions, either as a sintering aid or for property control. Wittmer and Buchanan⁹ achieved significant reduction in the densification temperature of PZT (53:47) by incorporation of up to 1 wt% V_2O_5 during coprecipitation. The as-dried coprecipitated powders were found to be X-ray amorphous. Powders calcined as low as 350°C produced XRD patterns characteristic of tetragonal PZT, with slight lattice distortion. Sintering of the ceramic was carried out in air at 950°C $\pm$ 10°C for 1-4 hr. Without the added V_2O_5 , a sintering temperature of 1280°C for 4-8 hr in a protective Pb ambient was required. The V_2O_5 was also incorporated as a mill addition subsequent to calcination of the coprecipitated PZT or directly to commercial

(e.g., TAM 5205) PZT powders.⁹ The microstructures obtained for the latter method were generally more uniform.

More recently Tuttle¹⁰ coprecipitated powders of PSZT + Nb₂O₅ using a process similar to that employed by Wittmer and Buchanan.⁹ PbO, SnO₂, and Nb₂O₅ were added to the blending mixture of TBT and ZNB. A 4:1 alcohol:water solution was used as the precipitating agent. Superior electrical properties and high electrocaloric effects were obtained with the fired coprecipitated powders. Fired densities between 92.4% and 95.6% theoretical were obtained by firing at 1380°C using a double-crucible technique.

While not specific as to details, it is clear from the reported work that much sensitivity attaches to the alkoxide precipitation process, when such parameters as pH, dilution, dispersion, temperature and rate of water added to bring about the hydrolysis of the butoxide mixtures, are considered.

The object of this study, therefore, was to investigate the relative influence of these parameters with respect to particle characteristics (agglomerate size distribution, crystallinity) and final densification.

II. Experimental

1. PZT Powder Preparation

The PZT formulation used for this study was Pb(Zr_{0.53}Ti_{0.47})O₃. This composition is close to the morphotropic phase boundary in the PbZrO₃-PbTiO₃ system and is generally known for high coupling coefficients and optimum dielectric properties.

The powders were prepared by coprecipitation from a mixture of electronic grade PbO (Hammond Lead Products, Inc., Hammond, Indiana), tetra-n-butyl titanate (E. I. Dupont de Nemeurs and Co. (Inc.), Wilmington, Delaware), and Zirconium tetra-n-butylate (Dynamit Nobel Co., Norwood, New Jersey).

The PbO powder was first added to a 50/50 butyl/isopropyl alcohol solution which was magnetically stirred to keep the PbO in suspension. The mixed butoxides (and a few drops of (Darvon 7) wetting agent) were added subsequently. Coprecipitation took place when a solution of deionized water in isopropanol was added to the stirred mixture. This was manifested by a significant thickening of the suspension. Transfer of the mixture to a high-speed blender and blending for 15-20 mins. produced an essentially stable gel which was then filtered, washed, and vacuum dried.

Variables which were monitored or controlled during the coprecipitation process included: dispersion, dilution, pH, water content and rate of addition, temperature, blending time and the filtering, washing and drying cycles. Dispersion here refers to homogeneity in blending to keep all solids in suspension. Dilution refers to the volume-percent suspended solids, which generally ranged from 2.0 to 7.0 vol%. The pH was maintained in the range of 4.0 to 9.0 by addition of glacial acetic acid or ammonium hydroxide to the precipitating solution. The water content of the precipitating solution varied from 25 to 67 vol%, corresponding to 12.5 to 33 vol% total liquid content. The H₂O solution was added at a rate of 6 to 32 ml/min. Temperature rise, due to the exothermic hydrolysis reaction was negligible during coprecipitation, but increased 5-10° during the blending cycle. The precipitate was washed either with: a) deionized water, b) 60/40 isopropanol/water solution, or c) isopropanol during filtration to remove the reaction products of the alkoxides. A final rinse with isopropanol to remove excess water, where appropriate, was carried out with some batches. Vacuum drying of the precipitate was done at room temperature and subsequently at 300°C to remove the organic residues.

2. Processing

Calcination of the precipitated powders was carried out at 800°C/4 hr, to develop the PZT phase and promote growth in the agglomerate and grain sizes.

Milling of the batches consisting of the calcined powders with 0-0.75 wt% V_2O_5 was done in polypropylene jars with ZrO_2 balls for 5 hr in a 60/40 isopropanol/water medium. An equivalent 1.0 wt% PVA in solution, as a binder, was added for the final hour of milling. The milled slurry was dried using a laboratory spray-dryer (Buche Model 190—Brinkman Instrument Company).

Pellets were pressed from the spray-dried powder at 207 MPa and fired on Pt foil at 950°C/10 min, or in a closed double-crucible (Pb atm) at 1280°C/4 hr, for samples without V_2O_5 .

Since the study was primarily concerned with variations in the coprecipitation parameters, processing variables were kept to a minimum.

3. Characterization

The vacuum-dried and calcined powders were analyzed by X-ray diffraction using a Phillips Norelco Diffractometer with filtered $Ni_{K\alpha}$ radiation. The powders were also subjected to DTA and TGA analysis using a Dupont 1090 Thermal Analyzer System. Particle-size distribution measurements were made using a Micromeritics X-ray Sedigraph Analyzer.

SEM analysis of fracture sections of dried, pressed and fired compacts were made using a JEOL JSM U-3 Scanning Electron Microscope equipped with an ORTEC Energy Dispersive Analyzer. Density measurements on the fired samples were made by geometric and mercury displacement techniques.

III. Results and Discussion

1. Powder Preparation

Most parameters monitored during the PZT coprecipitation process affected, in varying degrees, the characteristics of the precipitated powders.

Solutions of relatively low solids content (PbO: 2.0-3.0 vol%) were needed in order to maintain the PbO suspension, to minimize temperature fluctuations and to obtain a uniform precipitate. With higher solids content, larger agglomerate sizes and partially crystalline precipitates were obtained. This could be attributed to increased viscosity of the mixture as well as to uneven dispersion and temperature fluctuations arising from the exothermic hydrolysis reactions.

The effect of pH (4.5-8.5) on agglomerate size distribution of the dried precipitates is illustrated in Fig. 1. Average agglomerate sizes are shown to be lower for the acid powder (0.5 μm vs 0.8 μm for pH 8.5), and this also corresponded to a lower viscosity in the precipitating medium. A much coarser distribution is shown for the single-step process in Fig. 1. This process, generally cited in the literature, combines the coprecipitation and blending steps. Relatively poor dispersion and a significant rise in temperature ($\sim 40-60^\circ$) was noted for this technique and explains in part, the coarser distribution shown.

The effect of water content and rate of addition on the amorphous character of the precipitate, as determined by XRD, is illustrated in Fig. 2. H_2O contents of 33 and 67 vol% in the precipitating solutions were equivalent to 16.5 and 33 vol% respectively of the total liquid content. The rates of addition varied from 6.0-32 ml/min. For the air-dried powders only residual PbO XRD peaks were detected, and these were largely absent for the higher water contents and rates of addition. The amorphous nature of the powders could be attributed to the generally smaller agglomerate sizes of the precipitates, as illustrated in Fig. 3, where the low and high water content precipitates are compared. The distributions represent single-step coprecipitation with pH ~ 8.5 . Figure 3 also shows the size distributions after calcination.

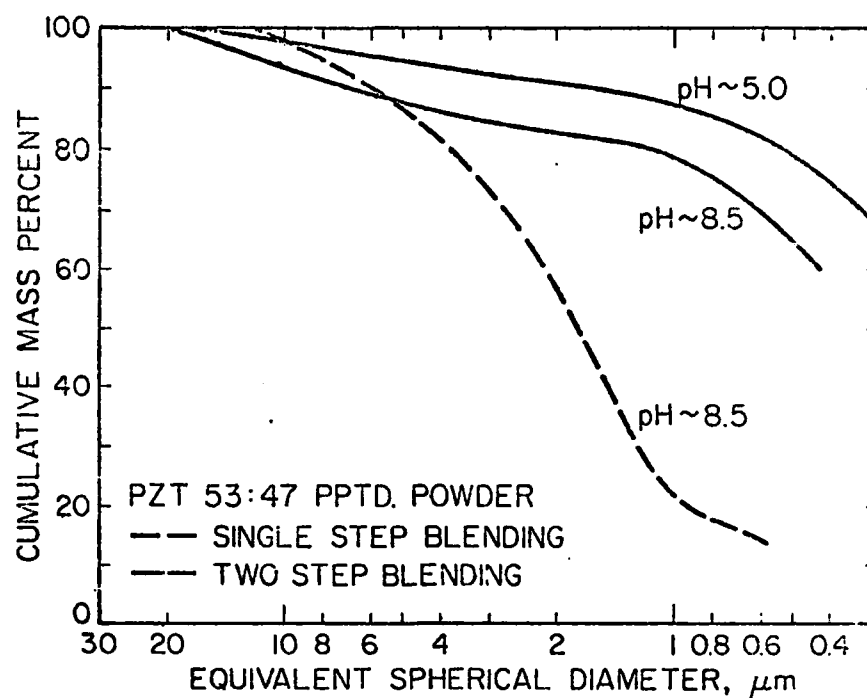


Fig. 1. Effect of pH and blending technique on agglomerate size distribution for PZT (53:47) coprecipitated powders.

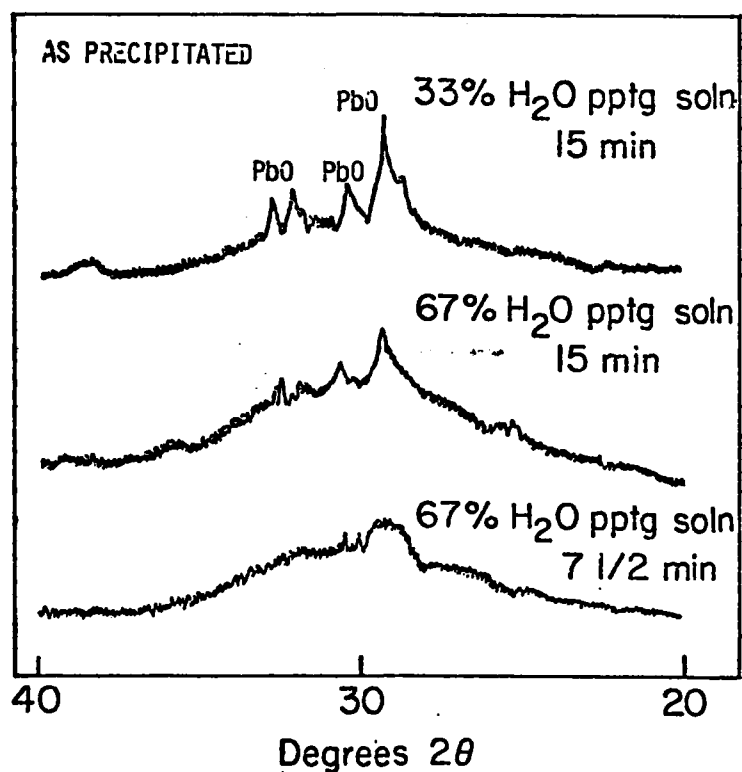


Fig. 2. Effect of H₂O content in precipitating solution and

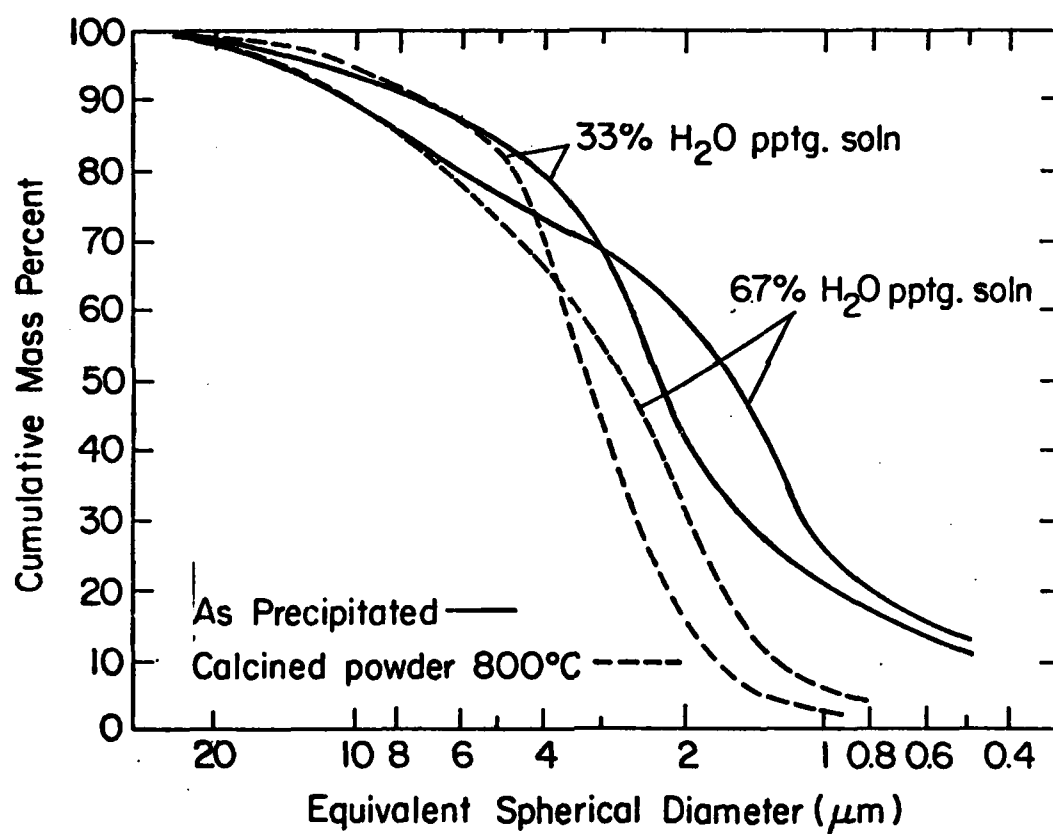


Fig. 3. Influence of H₂O content of precipitating solution on agglomerate size distribution for PZT powders.

As would be expected, there was an increase in the agglomerate sizes after calcination, but relative distributions persist. Vacuum drying of the precipitated powders at $\sim 300^\circ\text{C}$ resulted in a sharpening of the PbO XRD peaks and initial growth of the PZT phase. DTA analysis showed elimination of the residual organics to be most rapid at $\sim 290^\circ\text{C}$, as indicated by maximum TGA weight loss and a DTA exothermic reaction peaks.

DTA heating and cooling curves for the water and/or alcohol rinsed coprecipitated powder are presented in Figs. 4a and 4b. The DTA analyses showed exothermic reaction peaks occurring at approximately 200°C , 300°C , and 530°C . These peaks were attributed respectively to: a) loss of residual alcohol, b) elimination of the butoxide decomposition products, and c) formation of the PZT phase. TGA analysis showed the first two peaks to be associated with weight loss, the maximum loss occurring near 300°C .

From Fig. 4a it is evident that H_2O rinsing suppressed the exothermic peaks at $\sim 200^\circ\text{C}$ and 300°C . In contrast (Fig. 4b) rinsing with (isopropyl) alcohol is seen to enhance the lower peak and shift the second peak to higher temperatures. With the alcohol/water solution rinse only the lower temperature peak was suppressed. These observations are consistent with elimination of the alcohol at $\sim 200^\circ\text{C}$ and butoxide byproducts at $\sim 300^\circ\text{C}$, as indicated. Other powder characteristics (agglomerate size distribution, crystallinity) also varied with rinse conditions, as observed previously for ZrO_2 powders.¹¹

In terms of densification, the H_2O rinsed powders gave significantly lower fired densities than the alcohol/water rinsed, which gave the highest ($> 97\%$ theoretical density). These observations are also consistent with fired densities obtained by Haberko¹¹ on water and alcohol washed stabilized ZrO_2 powders. The differences were attributed to the weaker agglomerates formed with alcohol washing as a result of lower surface ionic absorption.

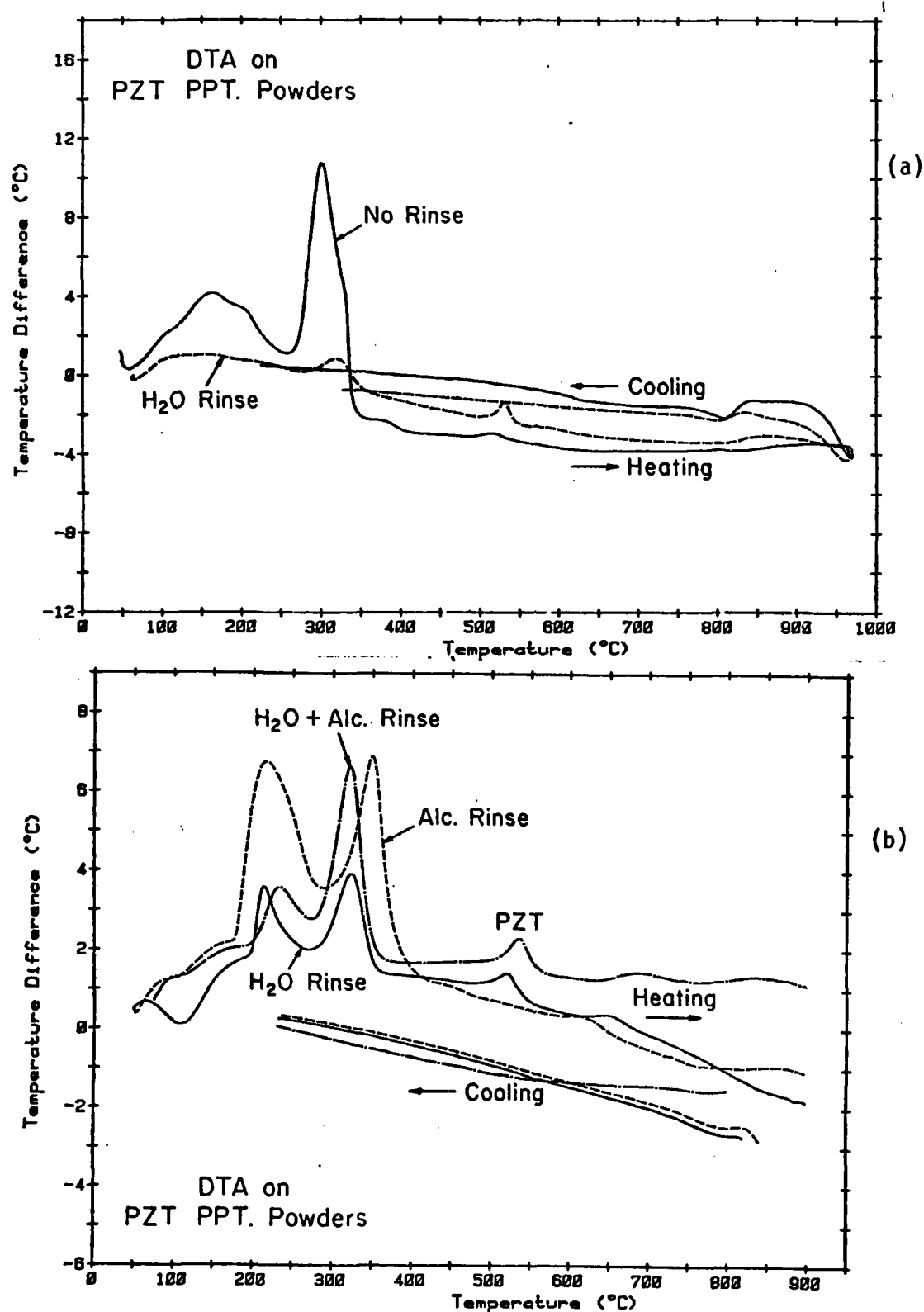


Fig. 4. DTA heating and cooling curves for PZT coprecipitated powders from butoxide precursors showing: a) H₂O and no rinse conditions, b) H₂O alcohol/H₂O and alcohol rinse conditions.

2. Processing

As indicated, growth in agglomerate and particle sizes will generally occur during calcination.¹² Providing hard agglomerates are not formed, the size distributions can be readily reduced by subsequent milling. This condition is illustrated in Fig. 5, where the agglomerate size distribution was substantially reduced by ball milling for up to 12 hrs subsequent to calcination. Prolonged milling will, however, introduce contamination.

SEM fracture micrographs for the dried precipitate (pH = 5), and after calcination, milling and pressing (207 MPa), are shown in Fig. 6. The pressed section shows a significantly more uniform and smaller particle size distribution (avg. PS ~ 0.3 μ m), compared to the dried precipitate, although some agglomerates persist. These agglomerates, formed during coprecipitation were enhanced by calcination and partially reduced by the milling steps. Pressing largely preserved the distributions, although different agglomerate configurations may be formed during spray-drying.

Figure 7 shows the sintering behavior for PZT samples fluxed with small amounts of V_2O_5 . Rapid densification was obtained at ~ 950°C, attributed to liquid phase rearrangement and solution precipitation. The liquid was determined to be a lead vanadate phase of approximate composition $V_2O_5 \cdot 4 PbO$.⁹ The rapid densification tended also to preserve the pressed microstructure in contrast to the double-crucible sintering of unfluxed PZT at ~1280°C/4 hr.

Figure 8 shows the effect of water content (single step process) on the pressed and fired densities. Lower pressed but higher fired densities were obtained for samples coprecipitated at the higher water contents when added rapidly. This is in line with the generally smaller and more uniform agglomerate size distributions noted for these precipitates. This relative distribution effect evidently persisted throughout the various processing

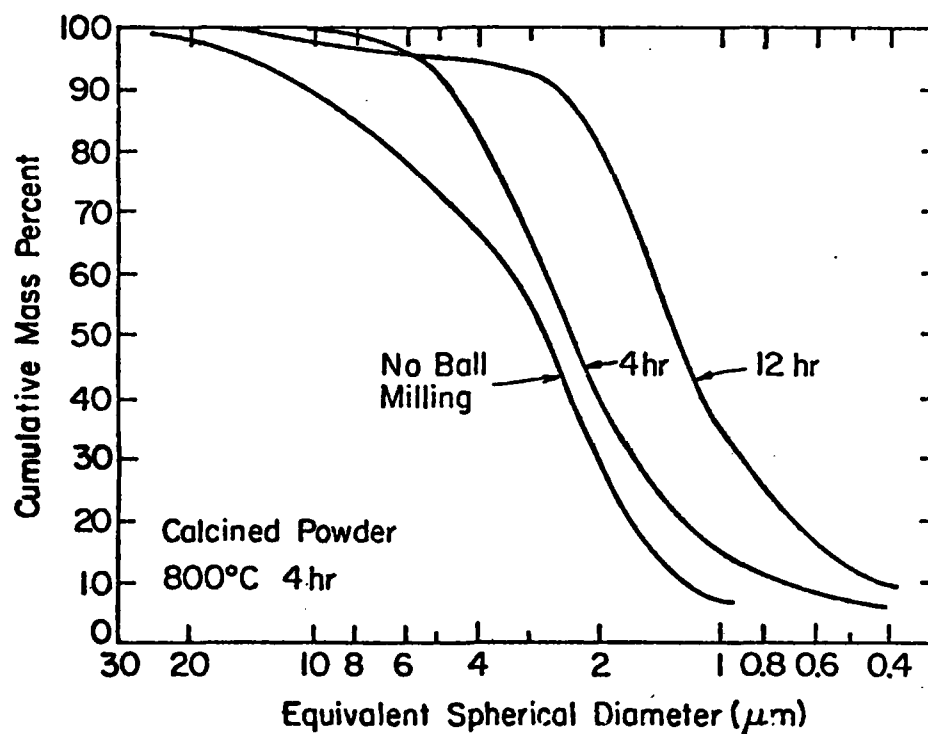


Fig. 5. Effect of milling time on agglomerate size distribution for calcined PZT powders.

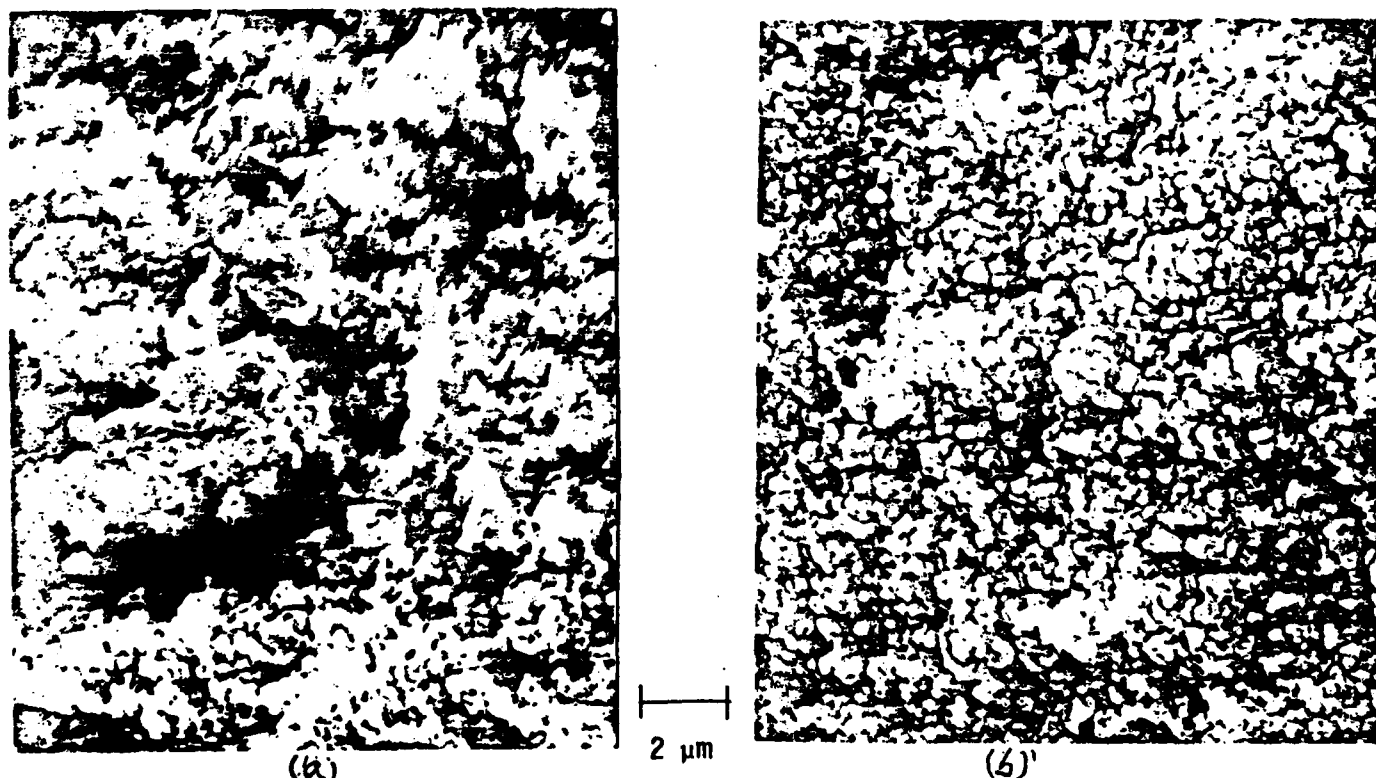


Fig. 6. SEM photomicrographs (5000X) for fracture sections of dried (A) and pressed (B) unfired PZT powders.

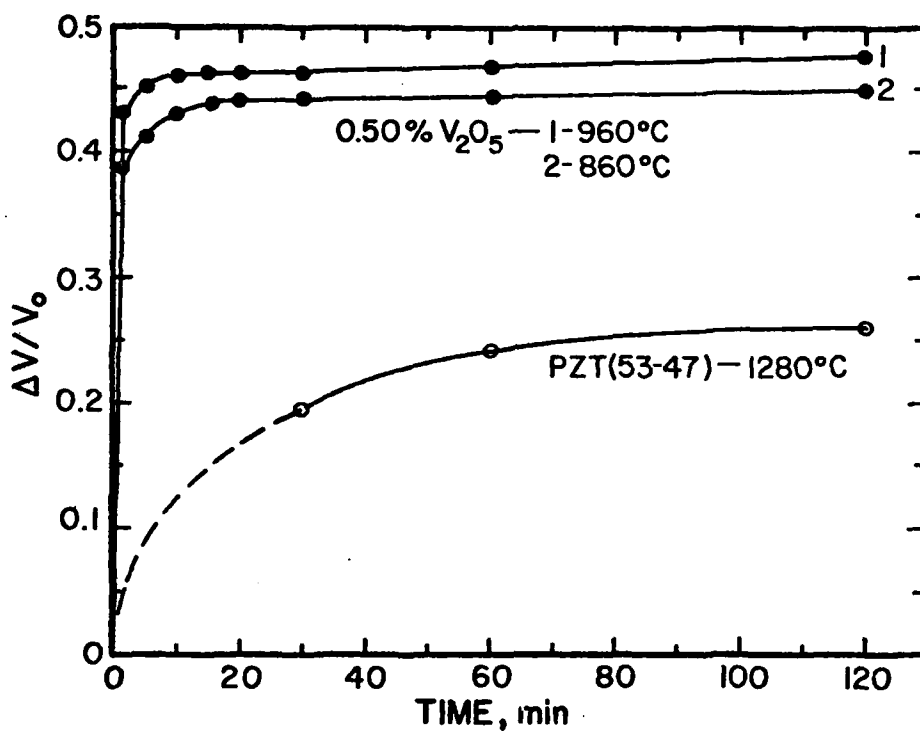


Fig. 7. Effect of V_2O_5 addition on densification of PZT (53:47) ceramic.

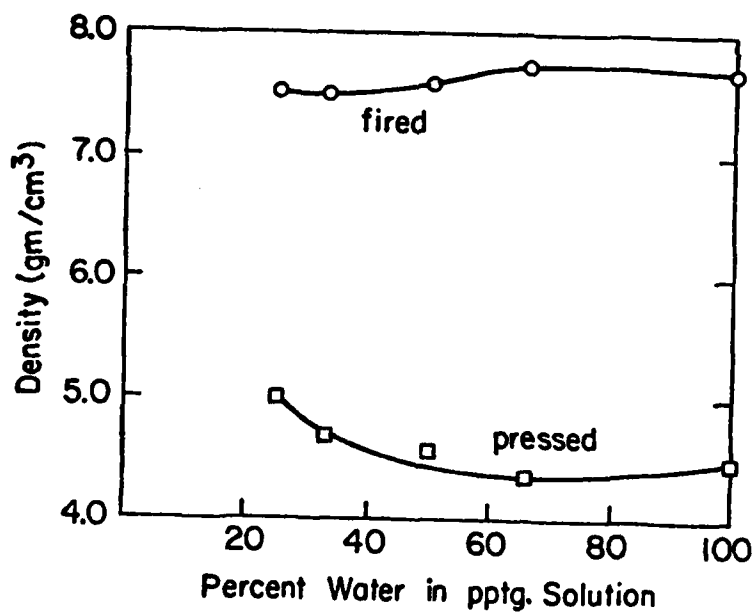


Fig. 8. Effect of water content in precipitating solution (agglomerate size distribution) on pressed and fired densities for PZT ceramic.

steps. This condition is illustrated in Figs. 9 and 10, which show SEM photomicrographs of unfired (pressed) PZT samples and also thermally etched sections of the fired samples, as a function of water content used during coprecipitation. The 25% H_2O fired sample showed a broader spectrum of grain size distribution compared to the 67% H_2O sample, with intermediate distributions for the 33% and 50% H_2O samples. The data correlates directly with the agglomerate size distribution observed in (Fig. 10) for the 25%, 33%, and 67% H_2O pressed samples. Similar variations would be expected, and have been observed^{13,14} for the different parameters affecting agglomerate size distribution, including spray drying.

IV. Conclusions

1. This study has shown that with PZT coprecipitation from butoxide precursors, powder characteristics such as agglomerate size distribution and XRD crystallinity were primarily fixed during the coprecipitation process.
2. The parameters which most sensitively affected coprecipitation, were found to be dilution, pH, and precipitation rate (controlled by H_2O addition). Low solids content (~ 3 vol% PbO), acidic pH (~ 4.5) and ~ 33 total H_2O vol%) were found to promote small, uniformly sized agglomerates. Subsequent rinsing of the precipitates by different concentration of alcohol/water solutions also affected, in particular, the DTA powder characteristics.
3. Relative agglomerate size distributions for the precipitated powders persisted throughout although the actual distributions varied according to subsequent calcination and milling. Final sintered densities and microstructures could readily be correlated with the agglomerate size distributions. Alcohol/water (60/40 vol ratio) had the most positive effect on densification followed in turn by no rinsing, alcohol and water rinsing. This latter was related to surface absorption characteristics of the powders.

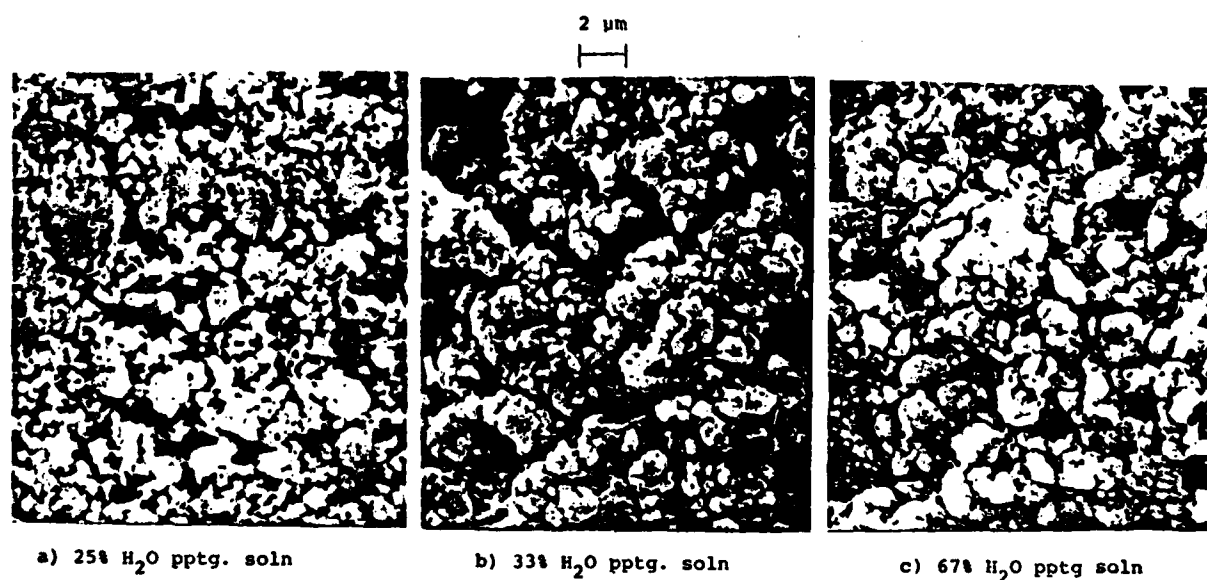


Fig. 9. SEM fracture micrographs of Unfired PZT (calcined 800°C) + 0.75 wt% V₂O₅ mill addition (5000 X).

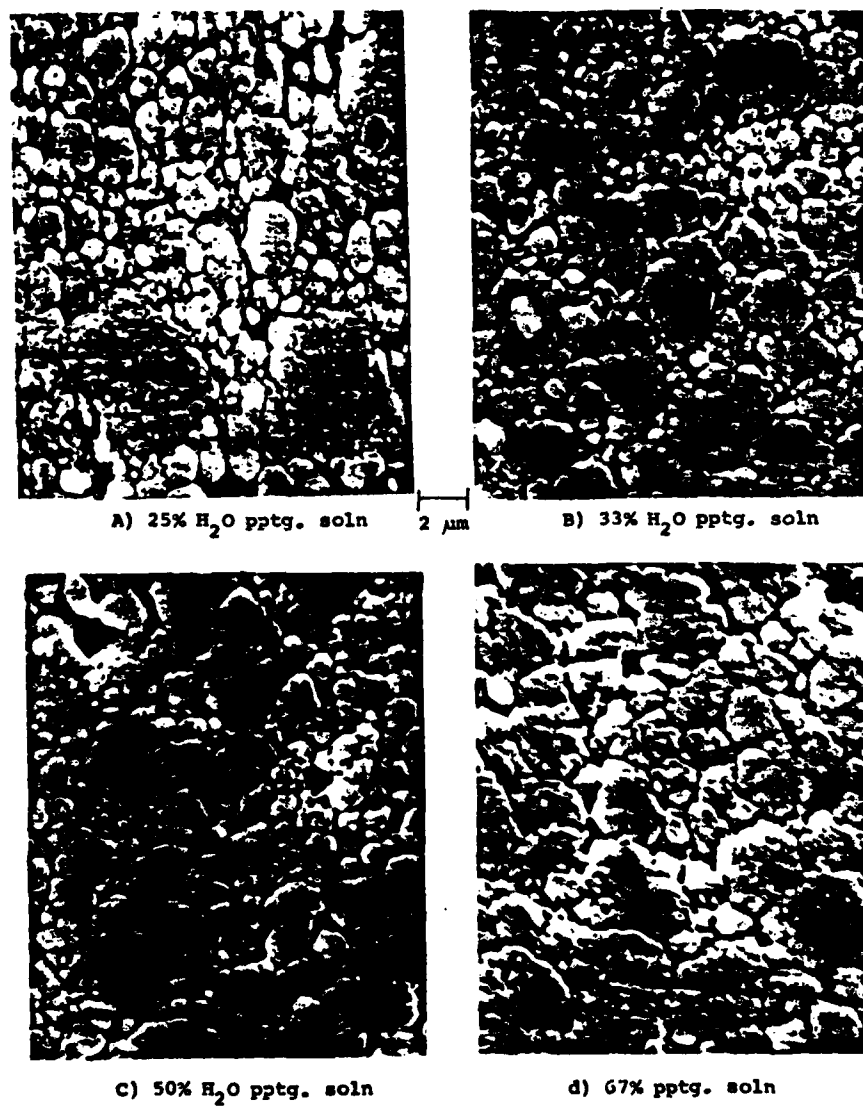


Fig. 10. SEM micrographs of thermally etched (PZT + V₂O₅) samples sintered at 950°C/30 min (5000X).

V. Acknowledgement

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Summary of Work Accomplished
Under Contract No. US NAVY-N-00014-80-K-0969

1. Reports

Report issued under this contract include the following:

- a. R. C. Buchanan and S. Pope, "Optical and Electrical Properties of Yttria Stabilized Zirconia (YSZ) Crystals," (ONR Report #5), University of Illinois, Urbana, IL (September 1981).
- b. R. C. Buchanan and J. Boy, "Effect of Coprecipitation Parameters on Powder Characteristics and On Densification of PZT Ceramics," (ONR Report #6), University of Illinois, Urbana, IL (September 1982).
- c. R. C. Buchanan and D. Wilson, "Development of Optically Transluscent Yttria Stabilized Zirconia (YSZ) Ceramics Below 1300°C with Alumina and Borate Additives." (ONR Report #7), University of Illinois, Urbana, IL (November 1982).

3. Papers

- a. R. C. Buchanan and S. Pope, "Optical and Electrical Properties of Yttria Stabilized Zirconia (YSZ) Crystals," Accepted, J. of Am. Ceram. Soc., 1982.
- b. R. C. Buchanan and J. Boy, "Effect of Coprecipitation Parameters on Powder Characteristics and On Densification of PZT Ceramics," submitted to J. of Am. Ceram. Soc., 1982.