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Statistical optimization of melt-quenching process parameters for multiple properties of ternary barium phosphate glasses

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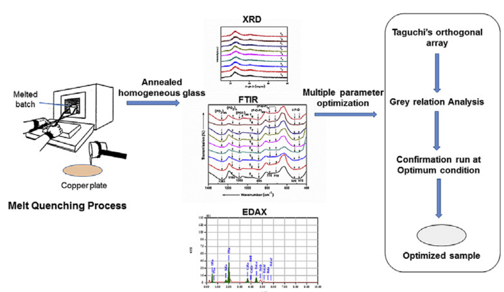
HIGHLIGHTS

- Optimal parametric combination is obtained by statistical design of experiment.
- Taguchi's orthogonal array reduces time and cost of experimentation.
- Significant parameters affecting the properties were obtained using ANOVA method.
- Reliability and repeatability of the method is also tested.

GRAPHICAL ABSTRACT



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ABSTRACT

In the present work simultaneous optimization of density, refractive index and hardness (multiple performance characteristics) of ternary barium phosphate glasses has been carried out using Taguchi based grey relational analysis. Effect of the parameters such as CaF_2 content, melting temperature and melting time on the response parameters of $(50 - X)BaO\text{-}XCaF_2\text{-}50P_2O_5$ ($X = 0, 10, 20$ mol%) glasses prepared according to the experimental layout of Taguchi's standard orthogonal array has been studied. Highest grey relational grade is the performance criteria used in this study to indicate the optimum level of process parameters needed for the best multiple performance characteristics. Analysis of variance (ANOVA) conducted on grey relational grade shows that under 95% confidence level, CaF_2 content is the only parameter significantly affecting multiple performance characteristics. Experimental values of response parameters obtained by conducting the confirmatory experiment at optimum level of process parameters are closer to the optimal set of predicted values.

1. Introduction

Synthesis of glasses with superior properties suited for numerous technological applications is a field of active research because future developments in medical technology, information processing and photonics depend crucially on the development of glass materials with improved mechanical and physical properties. Among oxide glasses (silicate and germanate glasses), phosphate glasses show excellent properties such as high refractive indices, high thermal expansion coefficients, high solubility of active ions, high electrical conductivity, low melting temperature, low softening temperature and low glass transition temperature, which make them suitable for variety of applications like host glasses for

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pulsed lasers, optical data transmission, solid state batteries, sensing technology and preparation of glasses with special properties [1–5]. Barium phosphate ($BaO-P_2O_5$) glasses show more homogeneity and wide glass formation region (35–100 mol% P_2O_5) compared to other alkali earth phosphate glasses [6]. $BaO-P_2O_5$ glasses with higher concentration of barium oxide (≥50 mol%) depict the properties like low photoelastic constants, high solubility of active ions and good water durability [7]. Barium and calcium metaphosphate glasses are used as matrix for the preparation of metal nanoparticle embedded glasses due to its high metal solubility [8]. But low chemical durability, poor thermal stability against crystallization and hygroscopic nature of phosphate glasses limit its extensive practical application [2,3,5,7].

In recent years there has been an extensive research on improving physical, mechanical, optical and electrical properties of oxide glass forming systems such as phosphate, silicate and borate glasses by adding halogen containing components to them. Addition of small amount of fluorine containing components to the glass batch reduces the melting temperature and time of synthesis of glasses [9,10]. It has been observed that addition of calcium fluoride (CaF_2) to lead phosphate and alkali phosphate glass batches lowers the viscosity of glass melts and thereby reduces the liquidus temperature to a substantial extent and also makes the glasses more corrosion resistant [11,12]. It further improves the durability of glasses by reducing the hydroxyl content [13]. Similarly, the addition of calcium fluoride to barium phosphate glasses is expected to reduce the melting temperature of them by reducing their viscosity and also make them more durable by reducing hydroxyl content. In addition to that, replacement of barium oxide (BaO) by calcium fluoride (CaF_2) in the batch composition may also improve the corrosion resistance and thermal stability of the glasses due to incorporation of Ca^{2+} ions into the glass matrix, which are having larger field strength (0.69) compared to Ba^{2+} ions(0.51) [14,15]. Present study mainly focuses on the preparation of ternary barium phosphate glasses with CaF_2, without degrading its properties such as hardness, refractive index and density.

More than 90% of practical glasses in both volume and number of types at present are prepared by melt-quenching process. The advantages of the melt - quenching technique like high flexibility of the geometry and large flexibility of composition of glasses make it easiest and suited method for the preparation of glasses [16]. Density (physical), refractive index (optical), hardness (mechanical) etc. are the basic properties of glasses considered for the industrial glass production and also for the selection of appropriate glasses for technological applications [17–19]. Previous studies show that glass composition and thermal history have significant impact on these properties of melt-quenched glasses [20–25]. In all the above mentioned studies, variation in glass properties with respect to change in composition and cooling rate are investigated either by simulation or by conventional trial and error method. In trial and error method, studies are carried out by varying only one parameter at a time keeping others constant. When the properties are affected by multiple parameters, conventional method requires a large number of experiments.

A material with improved properties can be obtained by optimizing parameters that influence the properties of that material using design of experiment (DOE). Statistical design of experiment is an important tool to understand the relationship among the various controllable parameters and identify the important parameter that influences the properties of materials [26]. Taguchi design of experiment, one of the approaches of DOE developed by Genichi Taguchi uses robust design to optimize the individual performance characteristics (response parameters) by reducing the variation in a process [27]. In our previous study, the level of process parameters needed to obtain a glass of highest refractive index has been successfully optimized using Taguchi method [28]. But multiple performance characteristics, which determines the quality of a product cannot be optimized simultaneously using Taguchi method [29]. Instead grey relational analysis based on grey system theory developed by Deng in 1982, which deals with poor, incomplete, and uncertain information and evaluate the interrelationships among the multiple performance characteristics is a suitable method to optimize complicated multiple performance characteristics [30]. Therefore, researchers used Taguchi based grey relational analysis in different fields, which requires less number of experiments for optimization of multiple performance characteristics [31]. It simplifies the procedure for determining optimum process parameters to obtain a high quality product. But there is no report on scientific study of simultaneous optimization of multiple performance characteristics of glasses prepared using melt quenching process. Thus, in the present work efforts are made to simultaneously optimize multiple performance characteristics such as density, refractive index and hardness of calcium fluoride added ternary barium phosphate glasses prepared by melt quenching process using Taguchi based grey relational analysis.

2. Experimental details

Ternary barium phosphate glasses of $(50-X)BaO\text{-}XCaF_2\text{-}50P_2O_5$ ($X = 0, 10, 20$ mol%) were prepared by melt-quenching process using reagent grade barium carbonate ($BaCO_3$), calcium fluoride (CaF_2) and ammonium dihydrogen phosphate ($NH_4H_2PO_4$) supplied by Alfa Aesar. A batch of 15 g of each composition was weighed, mixed well in agate mortar for 30 min and then calcined for 2 h at 400 °C in a muffle furnace to remove ammonia and water. The calcined powders were transferred into silica crucible and melted in a PID controlled high temperature furnace for 1–3 h in the temperature range 1000–1200 °C depending on the experimental layout of Taguchi's orthogonal array (Table 2). Homogenized melt was quenched in air on copper plates, immediately annealed at 400 °C for 3 h to remove the residual thermal stresses developed during the quenching and then furnace cooled to room temperature. To measure hardness and refractive index, prepared samples were grounded and polished using emery papers of different grades and alumina powder (0.05 micron) suspension.

X-ray diffraction (XRD) patterns of prepared samples were obtained by using JEOL, JDX-8P-X-ray diffractometer with Cu-K_α radiation. Powdered samples were scanned at a rate of 4°/min in the range of 10°–80° diffraction angle (2θ) and X-ray tube was operated at 40 kV and 30 mA. Energy dispersive analysis of X-ray (EDS) was carried out using scanning electron microscope (JEOL JSM 6380LA system) to determine the elemental composition of the glasses. FTIR analysis was carried out using FTIR spectrometer (JASCO-4200, Japan) and spectrum was obtained in the wave number range 400–1400 cm^{-1}. Samples were powdered and well mixed with KBr to make pellets and these pellets were placed in a spectrometer to obtain FTIR spectra of samples. Density (ρ) of prepared glass samples was obtained at room temperature by Archimedes principle using xylene as immersion liquid. Measurements were repeated for three specimens from the same batch and average value was obtained for each composition. An Abbe (MAR-

Table 1
Levels of parameters used in the experiment.

Process parameters	Symbol	Low	Centre	High
CaF_2 content, mol%	A	0	10	20
Melting temperature (temperature), °C	B	1000	1100	1200
Melting time (time), h	C	1	2	3