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Process modeling of gelcast Si_3N_4 ceramics using multi gene genetic programming

Gurabvaiah Punugupati[a] *, Kishore Kumar Kandi[b], P. S. C. Bose[c], C. S. P. Rao[d]

Research Scholar, Department of Mechanical Engineering, National Institute of technology, warangal-506004, India

Abstract

The influence of solid loading, monomer content and ratio of monomer to cross linking agent on the flexural strength and the porosity Si_3N_4 ceramic sintered body were investigated. The high strength porous Silicon Nitride (Si_3N_4) ceramics were prepared using gelcasting by increasing the monomer content in the slurry and flexural strength of the ceramic composite increases as the solid loading increases and flexural strength decreases as the monomer content increases. The porosity of the ceramic composite decreases as the solid loading increases and porosity increases as the monomer content increases. Flexural strength and porosity were determined as objective functions and a mathematical model is generated using Multi Gene Genetic Programming (MGGP).

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*Keywords:*Gelcasting; sintering; flexural strength; porosity ; Multi gene genetic programming

1. Introduction

1.1 Gelcasting

Silicon nitride (Si_3N_4) ceramics are considered as the acceptable material for their beneficial mechanical, chemical and physical properties at room and elevated temperatures [1, 2]. Different types of manufacturing methods are available to prepare Si_3N_4 ceramic composites. Among all gelcasting process is considered to be efficient. Gelcasting is an innovative approach to prepare the porous ceramic composites used in manufacturing of high

* Corresponding author. Tel.:+91-9951725237.
*E-mail address:*guru.punugupati65@gmail.com

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quality and complex shaped ceramic parts for numerous industries. The ideas are borrowed from the traditional ceramic processing [1]. During the gelcasting process, the binder forms a strong, continuous gel network, which permanently fixes the ceramic particles in their positions, hindering the formation of in-homogeneities during subsequent drying and sintering [3-7].

1.2 Multi Gene Genetic Programming (MGGP)

An increasingly popular technique is the multi gene genetic programming (MGGP). In a standard genetic program, the representation used is a variable-sized tree of functions and values. Each leaf in the tree is a label from an available set of value labels. Each internal node in the tree is label from an available set of function labels. The entire tree corresponds to a single function that may be evaluated. Typically, the tree is evaluated in a leftmost depth first manner. A leaf is evaluated as the corresponding value. A function is evaluated using an argument that is the result of the evaluation of its children. Genetic algorithms (GA) and genetic programming are similar in most other respects, except that the reproduction operators are tailored to a tree representation [8, 9]. A multigene individual consists of one or more genes, each of which is a traditional GP tree. Genes acquired incrementally by individuals in order to improve fitness (e.g. to reduce a model's sum of squared errors on a data set). The overall model is a weighted linear combination of each gene. Optimal weights for the genes are automatically obtained (using ordinary least squares to regress the genes against the output data). The resulting pseudo-linear model can capture nonlinear behavior [10].

2. Experimental Details

2.1Preparation of gelcasting Si_3N_4 ceramics

Fig. 1 shows the detailed flowchart of the gelcasting process.

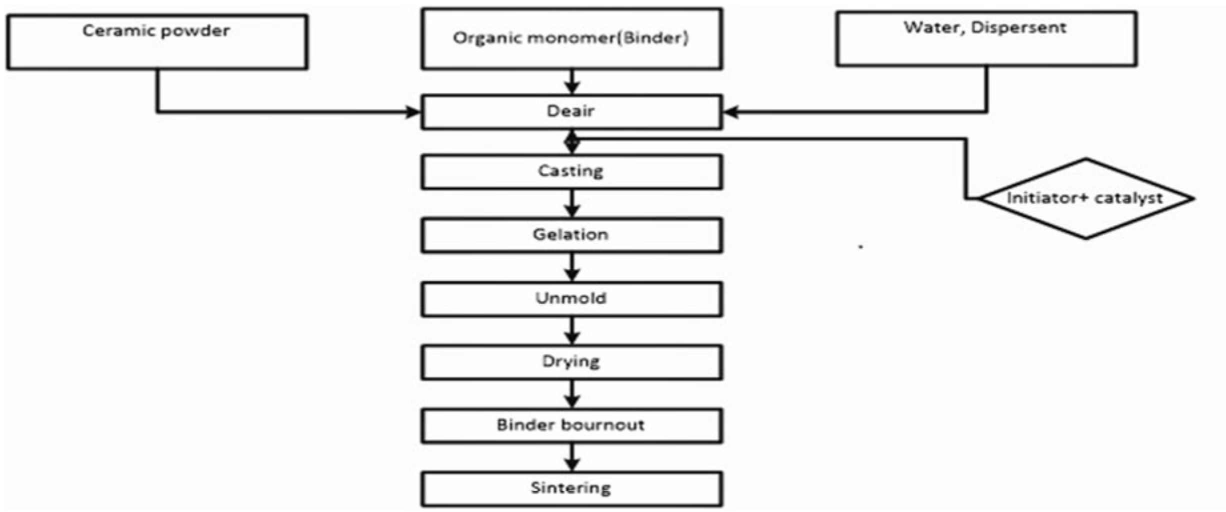


Fig. 1. Flow chart of gelcasting process

During the gelcasting preparation of porous Si_3N_4 ceramics, in the first step, dispersant (Dolapix A88, 1 wt%, based on silicon nitride) and monomers (MAM-methacrylamide and MBAM- N, N^1-methylenebisacrylamide) were completely dissolved in distilled water by magnetic stirring, and the premix solution was served as a dispersing media for the ceramic powders. The next step was to add silicon nitride powder and suitable sintering additives (2 wt%, Al_2O_3; 1 wt%, Y_2O_3) into the premix solution, and the slurry was degassed for 15–20 min. After that APS (Ammonium persulfate) and TEMED (Tetramethylethylenediamine) are added to the premix slurry which acts as initiator and the catalyst. The slurry was cast into a nonporous rectangular glass mold. The mold was left for 15–20 minutes to polymerize to form gelled body after casting and then the gelled part was removed from the mold and dried to remove the solvent system. In order to avoid the occurrence of crack and warpage caused by rapid drying, the samples were dried in a commercial dryer at controlled humidity conditions. Finally, sintering was performed at a heating rate of 2 ^{O}C/min at 550 ^{O}C for binder burn out and heating rate 10 ^{O}C/min and 1 hour holding time at 1700 ^{O}C under nitrogen atmosphere, and then porous Si_3N_4 ceramics were achieved.

Design of experiments comprises of set of experiments which are to be carried out in a sequential manner for evaluating the response measurements [11]. 27 runs full factorial method is chosen since the method provides a wider covering region of parameter space and good consideration of variables interaction in the model. The ranges of manufacturing parameters used are given in table 1.

Table 1.factors and levels

S. No	Factors	Levels		
1	Solid loading (vol %)	30	40	50
2	Ratio of monomer to cross linking agent (MAM:MBAM)	3:1	6:1	9:1
3	Monomers content (Wt %)	5	10	15

The measured values of flexural strength and porosity for 27 experiments conducted as per full factorial for training dataset. The 27 ceramic samples are shown in the Fig. 2. The dispersant, TEMED and APS are kept constant.

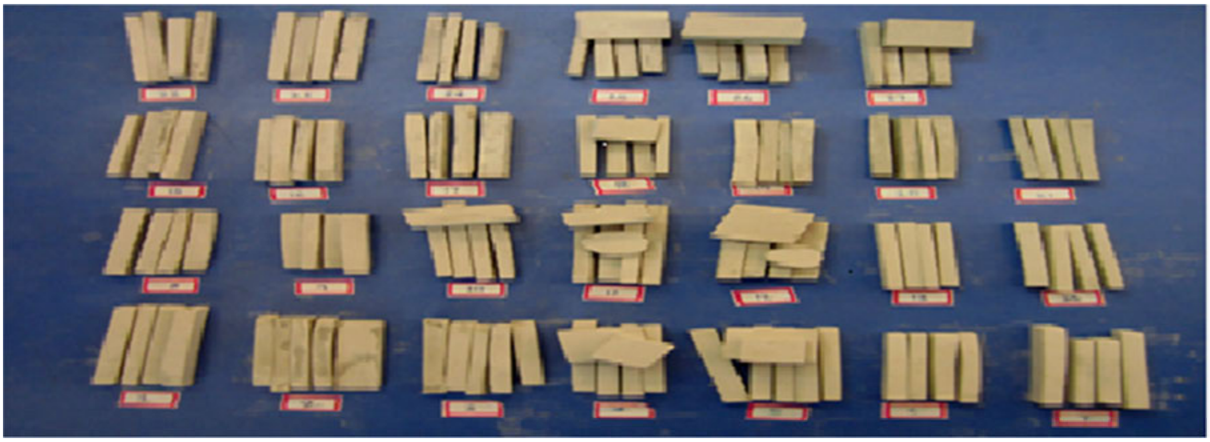


Fig. 2. Ceramic composite samples

The materials and compositions are shown in table 2.

Table 2. Materials kept constant

S.No	Material	Quantity
1	Al_2O_3(Aluminum Oxide)	2 wt% of solid loading
2	Y_2O_3(Yttrium Oxide)	1 wt% of solid loading
3	Dispersant	1 wt% of solid loading
4	APS	0.8 wt% of monomers
5	TEMED	0.5 wt% of monomers

3. Material characterizations

3.1 Three point bending test

Flexural strength is the ability of the material to withstand bending forces applied perpendicular to its longitudinal axis [12].The room-temperature flexural strength of sintered body was determined by three-point bending test. The specimens were machined into test bars, and flexural strength was measured on specimen bars of dimensions length x width x length are 45x4x3 mm^3 respectively. For a 3-point bend test of a rectangular bar,the stress at fracture using the bend test is known as the flexural Strength or modulus of rupture and is given by

$$\sigma_f = \frac{3pl}{2bh^2} \quad (1)$$

Where

σ_f =Flexural strength (Mpa), p= Fracture load (N), l= Length between outer supports (mm)
b= Specimen width (mm), h= Specimen height (mm)

3.2 Porosity measurement

The porosity of sintered body was measured by the Archimedes' displacement method with distilled water. The bulk density (ρ_0) and the true density (ρ) of the sintered bodies were determined by Archimedes' principle water as the medium.

The porosity (P) was calculated as follow:

$$P = (1 - \frac{\rho}{\rho_0}) \quad (2)$$

The bulk density (ρ_0) of sintered bodies is determined by using following formula.

$$\text{Bulk density} = \frac{D}{(w1 - w2)} \quad (3)$$

Where D= dry weight, w_1= Soaked weight, w_2= suspended weight

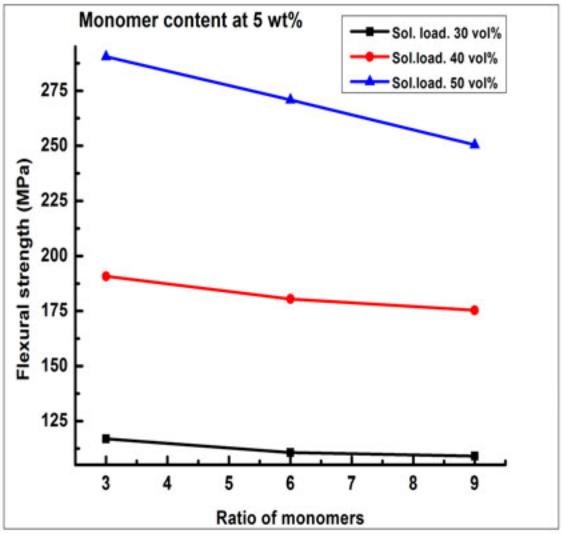
4. Results and discussion

4.1 Effect of manufacturing parameters on the flexural strength

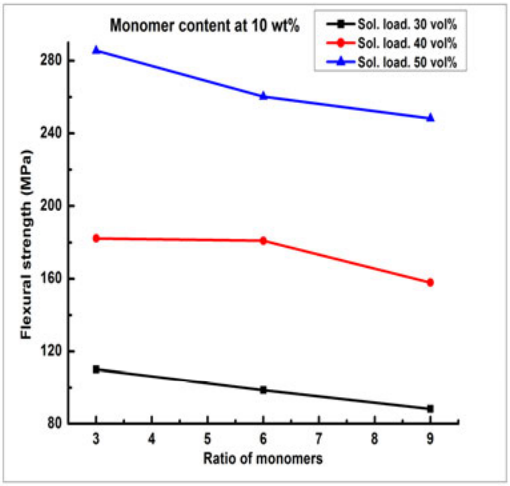
Table 3. Experimental data set

S. No	Solid loading (vol %)(x_1)	Ratio of monomer to cross linking agent (MAM:MBAM) (x_2)	Monomers content (wt%)(x3)	Flexural strength (MPa)	Porosity (%)
1	30	3:1	5	116.93	43.01
2	30	3:1	10	110.07	44.03
3	30	3:1	15	102.03	46.5
4	30	6:1	5	110.69	45.7
5	30	6:1	10	98.69	48.1
6	30	6:1	15	85.03	51.76
7	30	9:1	5	108.98	46.0
8	30	9:1	10	88.229	51.01
9	30	9:1	15	68.64	53.0
10	40	3:1	5	190.76	33.36
11	40	3:1	10	182.21	35.08
12	40	3:1	15	173.32	38.04
13	40	6:1	5	180.46	36.1
14	40	6:1	10	180.9	36.7
15	40	6:1	15	149.81	39.2
16	40	9:1	5	175.32	37.4
17	40	9:1	10	157.88	40.7
18	40	9:1	15	120.73	42.23
19	50	3:1	5	290.50	25.01
20	50	3:1	10	285.52	26.5
21	50	3:1	15	273.05	27.0
22	50	6:1	5	270.9	27.3
23	50	6:1	10	260.3	28.0
24	50	6:1	15	250.26	29.7
25	50	9:1	5	250.51	30.5
26	50	9:1	10	248.23	31.1
27	50	9:1	15	229.07	32.5

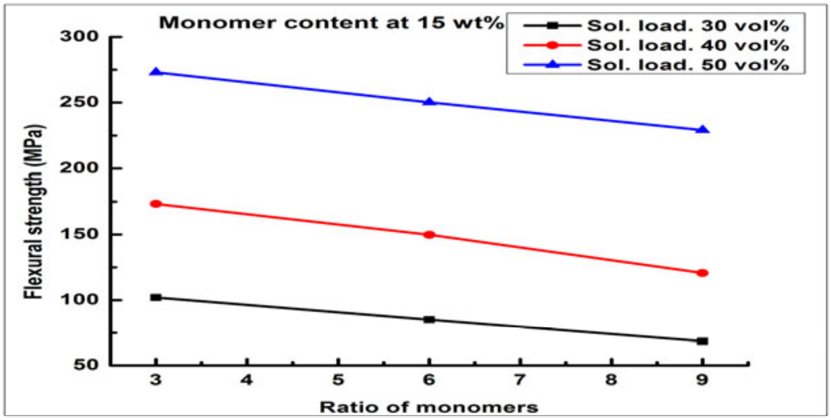
Effect of ratio of monomer to cross linking agent at different solid loadings and constant monomer content on flexural strength are shown in Fig.3 (a), (b), (c).



(a)



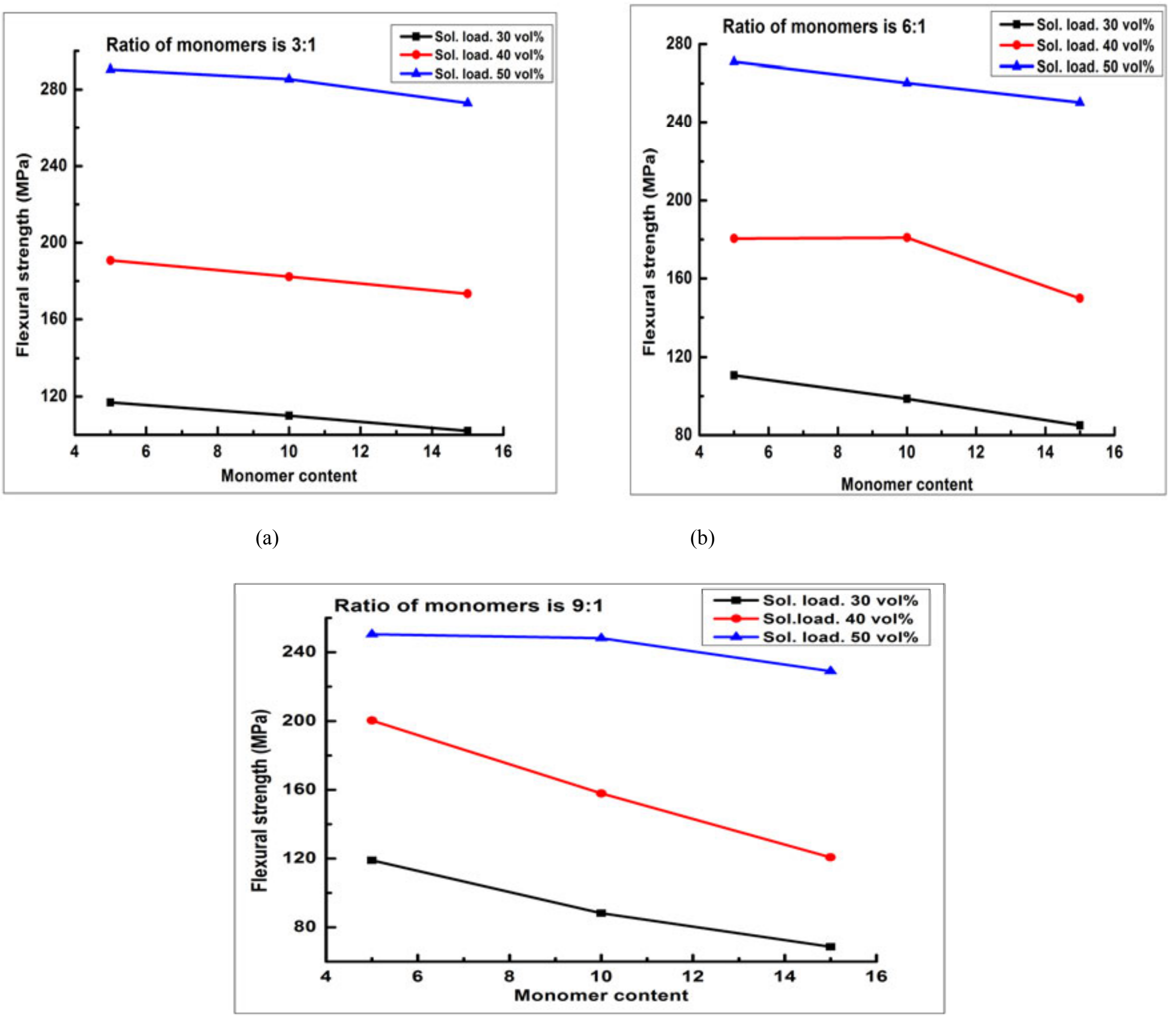
(b)



(c)

Fig. 3. Variation of flexural strength as the function of ratio of monomer s (a) Monomer content 5 wt%, (b) Monomer content 10 wt%, (c) Monomer content 15 wt%

Effect of monomer content at different solid loadings and constant ratio of monomer to cross linking agent on flexural strength are shown in Fig. 4 (a), (b), (c).



(a) (b)

(c)

Fig. 4. Variation of flexural strength as a function of monomer content, (a) at monomers ratio 3:1, (b) at monomers ratio 6:1, (c) at monomers ratio 9:1

From Fig.3 (a), (b), (c) the flexural strength of ceramic composites increases as the solid loading increases and Flexural strength decreases as the ratio of monomer to cross linking agent increases at constant monomer content and solid loading respectively.

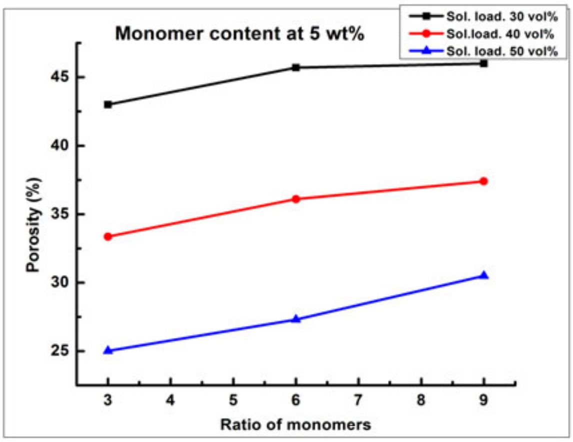
From Fig. 4 (a), (b), (c) the flexural strength of ceramic composites increases as the solid loading increases and Flexural strength decreases as the monomer content increases at constant ratio of monomer to cross linking agent and solid loading respectively.

Solid loading has great influences on the flexural strength of porous Si_3N_4 ceramics, and the reasons are as follows:

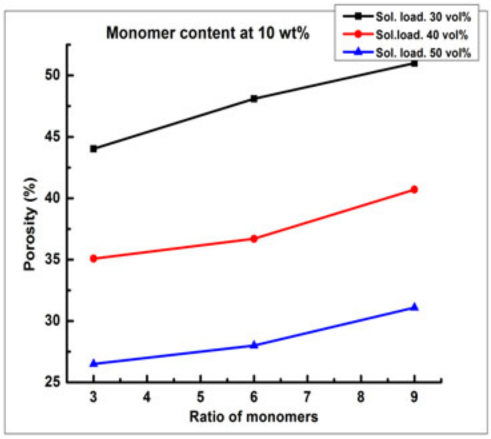
- The spaces between Si_3N_4 particles in slurry are affected by the solid loading.
- Solid loading has a great influence on the shrinkage during drying and sintering.
- Solid loading also affects the density of the sintered body.

4. 2 Effect of manufacturing parameters on the porosity

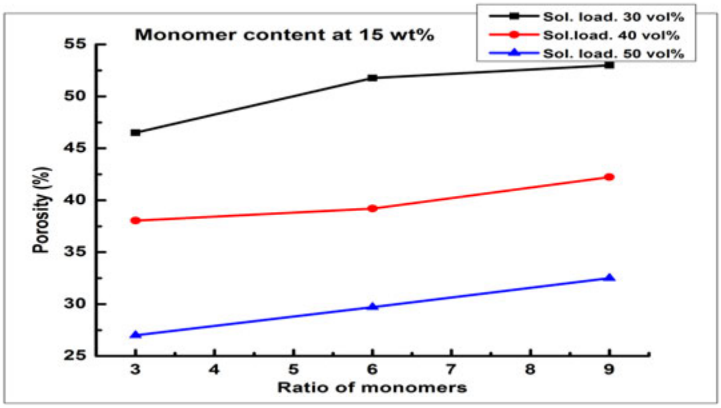
Effect of ratio of monomer to cross linking agent at different solid loadings and constant monomer content on porosity are shown in Fig. 5 (a), (b), (c).



(a)



(b)



(c)

Fig. 5. Variation of porosityas the function of ratio of monomers (a) Monomer content 5 wt%, (b) Monomer content 10 wt%, (c) Monomer content 15 wt%

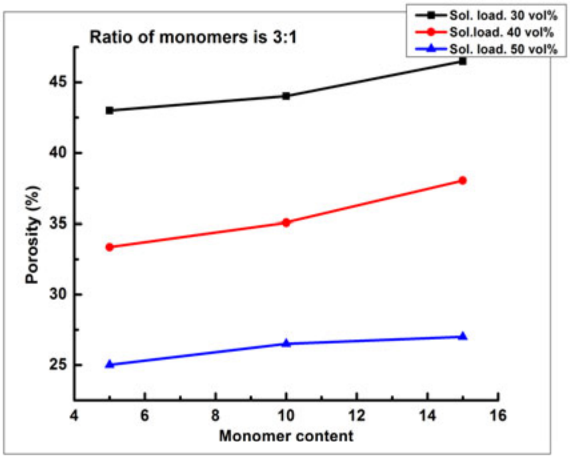
Effect of monomer content at different solid loadings and constant ratio of monomer to cross linking agent on porosity are shown in fig.6 (a), (b), (c).

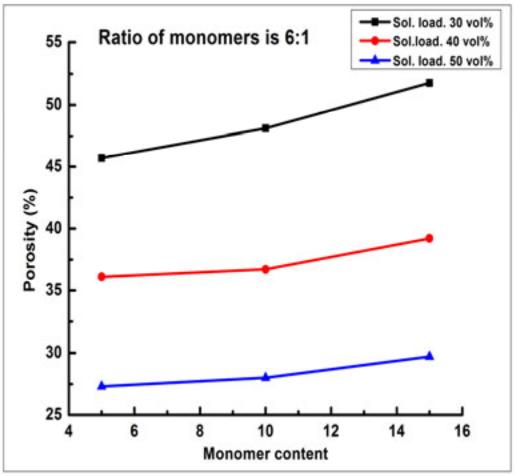
From Fig. 5 (a), (b), (c) the porosity of ceramic composites decreases as the solid loading increases and porosity increases as the ratio of monomer to cross linking agent increases at constant monomer content and solid loading respectively.

From Fig. 6 (a), (b), (c) the porosity of ceramic composites decreases as the solid loading increases and porosity increases as the monomer content increases at constant ratio of monomer to cross linking agent and solid loading respectively.

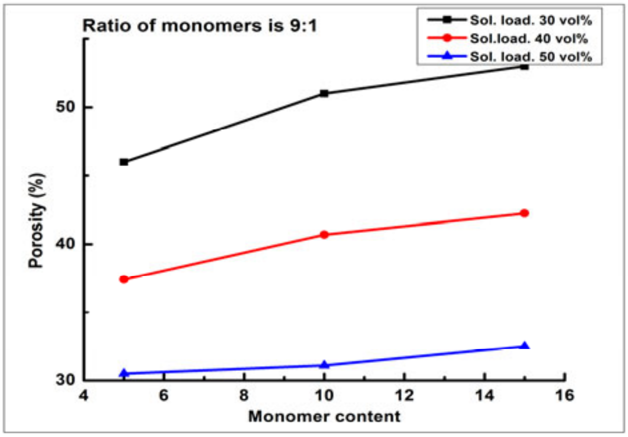
Porosity of sintering body increases with the increase of monomer content. In the experiment, the porous ceramics is prepared by only increasing monomer content in the slurry without other organic additives during gel casting. Here, monomer and crosslinking agent not only can form macromolecular network to hold the ceramic particles together, but also can play the leading role in the formation of pores during the preparation of porous Si_3N_4 ceramics.

The increase of crosslinking agent makes the crosslink density of cross-linked polymer gels in green body increase, the distribution of ceramicsparticles is more uniform, the drying shrinkage is smaller, and thus the porosity of sintering body increases.





(a) (b)



(c)

Fig. 6. Variation of porosityas a function of monomer content, (a) at monomers ratio 3:1, (b) at monomers ratio 6:1, (c) at monomers ratio 9:1

4.3 Implementation of MGGP

The training data set for implementation of MGGP constitutes table 3. MGGP, being a stochastic search technique, makes no prior assumptions about the actual model form. The structure and complexity of the model evolve automatically. The populations of models of initial tree generation for flexural strength and porosity are first initialized. The terminal set T and the function set F were defined as: T= {x_1, x_2, x_3} and F= {+, -, *, /}. Based on these experiments, the final parameters used to generate the models are given in Table 4.

Table4. MGGP Control Parameters

Terminal set	{x_1, x_2, x_3}
Functional set	{+, -, *, /}
Population size	100
Number of generations	100
Crossover probability (%)	85
Mutation Probability (%)	10
Elitism Probability (%)	05
Selection method	Tournament selection

The following expressions for flexural strength and porosity were found to have the best fitness value.
Overall MGGP model for flexural strength (MPa)

$$F.S = 0.06213x_2 - 0.06221x_1 + 0.5736x_3 - 0.06221x_1x_2 + \frac{8.4x_2x_3}{10^5} + 0.007062(x_3 - x_1 + 3.286(x_3^2 + 2x_1 - x_2)) - \frac{8.4x_1(x_1+(x_1+x_2)(x_2-x_3))}{10^5} + 0.0147x_3(3\,x_3 - 5.621) + 0.1244x_1^2 + 0.0039x_1^2(x_2 - x_3) + 0.02601x_3(x_1 - x_2x_3) + 23.69 \qquad (4)$$

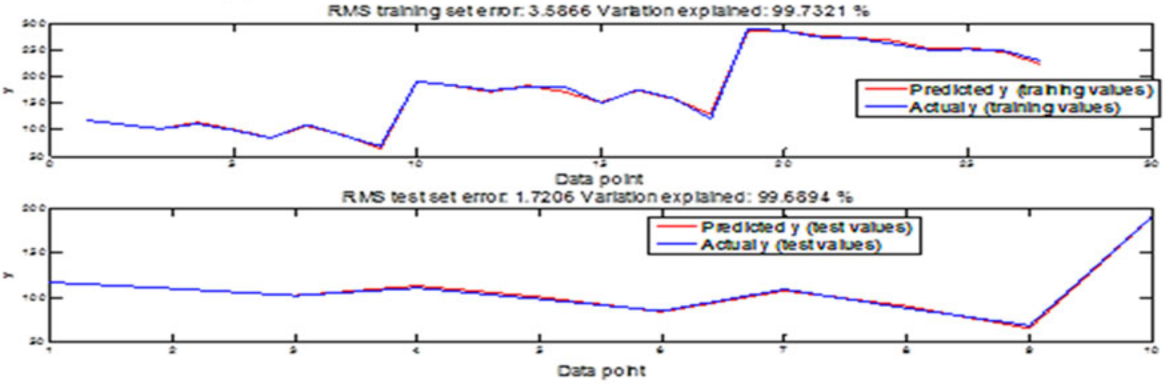


Fig.7. Predicted vs Actual values of flexural strength

Overall MGGP model for porosity (%)

$$P = \frac{4.304x_1^3}{10^5} + \left(\frac{1.27x_2^2 - 1.27x_2}{10^5}\right)x_1^2 + (-0.0002738x_2^2 - 0.006391x_2 - 0.01717x_3 - 0.9792)x_1 + 0.9793\,x_3 + 0.01717\,x_2x_3 - 0.01278\,x_2^2 + 65.99 \qquad (5)$$

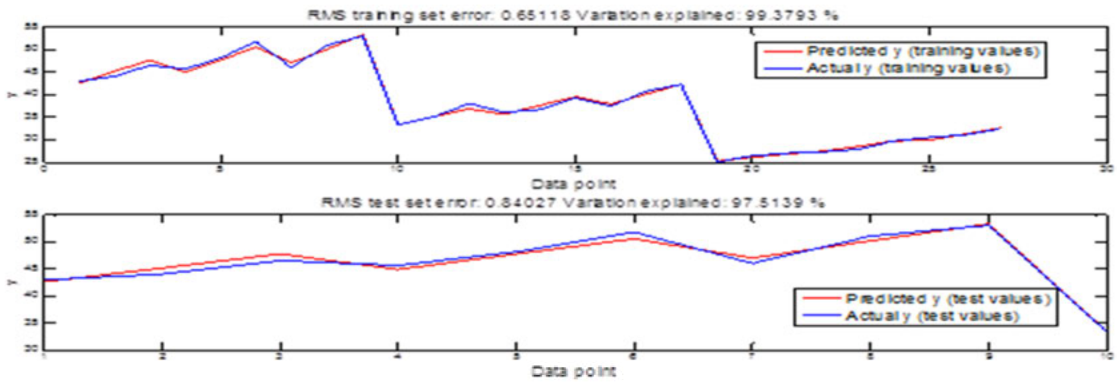


Fig. 8. Predicted vs Actual values of porosity

A comparison of the predicted models and the experimental values for the validation datasets of flexural strength and porosity are shown in Fig. 7 and Fig. 8 respectively. MGGP predicts the responses with high accuracy. Very high values of correlation coefficient (R^2) for flexural strength and porosity of validation sets are obtained and found to be 0.9937 and 0.9751 respectively. These indicate that the developed models satisfactorily represent the outputs.

Conclusions

The following conclusions were drawn based on the present investigations.
1. Si_3N_4 ceramic samples were prepared by Gelcasting method with varying solid loading, ratio of monomer to cross linking agent and monomer content at 3 levels using full factorial experimentation. Flexural strength and porosity of gelcast ceramic composite were measured. It found that

- Flexural strength increases as solid loading increases and decreases as ratio of monomer to cross linking agent and monomer content increases.
- Porosity decreases as solid loading increases and increases as ratio of monomer to cross linking agent and monomer content increases.

2. Experimental data based on full factorial experiments were used to develop empirical models for flexural strength and porosity in terms of manufacturing parameters solid loading, ratio of monomer to cross linking agent and monomer content, with an efficient evolutionary algorithm namely, Multi Gene Genetic Programming.
3. Multi Gene Genetic Programming is a domain independent methodology which does not assume any prior functional form of the solution and hence it can accurately model the complex relationships of the process.

References

[1] Juanli Yu , Hongjie Wang , Jian Zhang , Dahai Zhang ,Youlan Yan, Gelcasting preparation of porous silicon nitride ceramics by adjusting the content of monomers, J Sol-Gel SciTechnol (2010) 53:515–523.
[2] Jianqingdai, Zhipengxie, Young huang, Liguo ma, Longjiezhou, The contributing factor and elimination of macro pores in si3n4 green bodies formed by water-based gelcasting, Journal of materials science letters 18 (1999) 1739 – 1742.
[3] Jing-yizhang, Feng ye, Effect of agarose content on microstructures and mechanical properties of porous silicon nitride ceramics produced by gelcasting, Journal of Zhejiang university-science a (applied physics & engineering 2010 11(10):771-775.
[4] Chunrong Zou , Changrui Zhang, Bin Li, Siqing Wang, Feng Cao, Microstructure and properties of porous silicon nitride ceramics prepared by gel-casting and gas pressure sintering, Materials and Design 44 (2013) 114–118.
[5] Jyoti Prakash Nayak and Japes Bera, Bioactive Porous Silica Ceramics Prepared using Rice Husk Ash by Gelcasting Method, Advanced Materials Research Vol. 548 (2012) pp 12-16.
[6] Wen Zhang, Hongjie Wang, ZhihaoJin, Gel casting and properties of porous silicon carbide/silicon nitride composite ceramics, Materials Letters 59 (2005) 250– 256.
[7] Xing he bosu, Xinguizhou, Junhe yang, Bin zhao, Xianyingwang, Guangzhi yang, Zhihong tang, Hanxunqiu. Gelcasting of alumina ceramics using an egg white protein binder system.Ceramics – Silikaty 55 (1) 1-7 (2011).
[8] Koza, J. R. Genetic programming: On the Programming of Computers by Means of Natural Selection, MIT Press, Cambridge, MA, 1992.
[9] S.N.Sivanandam, S.N.Deepa. Introduction to Genetic Algorithms.Springer-Verlag Berlin Heidelberg 2008.
[10] J.O. Orove, N.E. Osegi, B.O. Eke, A Multi-Gene Genetic Programming Application for Predicting Students Failure at School.
[11] VikasSonkar, Kumar Abhishek, SauravDattac, SibaSankarMahapatra, Multi-Objective Optimization in Drilling of GFRP Composites: A Degree of Similarity Approach, Procedia Materials Science 6 (2014) 538 – 543.
[12] K. Kishor Kumar, Dr. K. Raja Narender Reddy, S. Sripathy, Evaluation OF Flexural and Water Absorption Properties of Short Kenaf Fiber Reinforced Green Composites, Advanced Materials Manufacturing & Characterization Vol 3 Issue 2 (2013).
[13] Juanli Yu, Hongjie Wang, Hong Zeng, Jian Zhang. Effect of monomer content on physical properties of silicon nitride ceramic green body prepared by gelcasting. Ceramics International 35 (2009) 1039–1044.
[14]Mark A. Janney, Stephen D. Nunn, Claudia A. Walls, Ogbemi O. Omatete, Randy B. Ogle, Glen H. Kirby, April D. McMillan. Gelcasting, the handbook of cermic engineering, Mohamed N. Rahaman, Editor, Marcel Dekker, 1998.
[15] Sara Silva, GPLAB, A Genetic Programming Toolbox for MATLAB, Version 3, 2007.
[16]Stephen J. chapman, Essentials of MATLAB programming, second edition, 2009.