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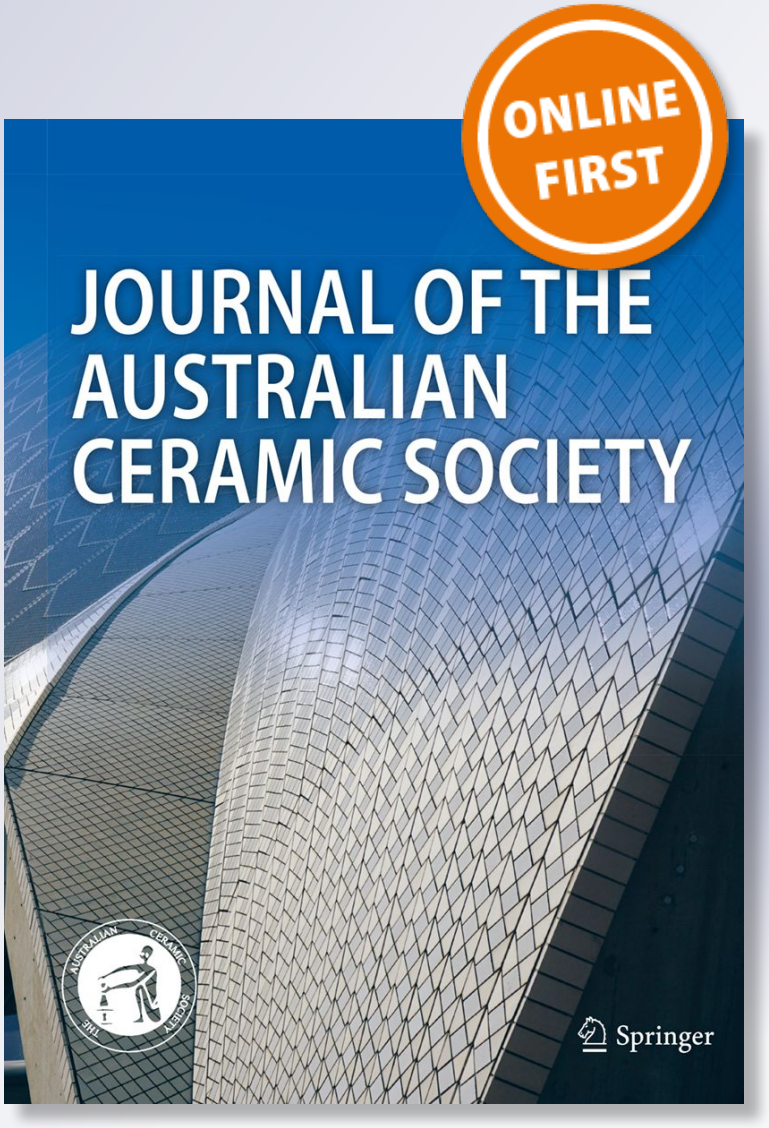

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RESEARCH

Optimization of the physical properties of barium titanates using a genetic algorithm approach

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Abstract Barium titanates are widely used in the electronics industry because of their high dielectric constant and ferroelectric and piezoelectric properties. These properties are related to the physical properties of the material; thus, the optimization of these properties is crucially important. The aim of the current work is to control the values of the processing parameters to produce the optimum density, porosity, firing shrinkage, and green density of $BaTiO_3$. A genetic algorithm was used to fulfill this aim. The modified pechini method was used to prepare barium titanate powders with five different particle size distributions. Eighty samples were prepared using different processing parameters including the pressing rate, pressing pressure, heating rate, sintering temperature, and soaking time. A genetic algorithm (GA) approach has been applied in order to obtain the optimum processing parameters. The results showed that the best value of the density that can be achieved is 6.02 g/cm^3, which is equal to the theoretical density of $BaTiO_3$ using a pressing rate of 3 KN/S, a pressing pressure of around 370–385 Mpa, a sintering temperature of not less than 1400 °C, a soaking time of around 6–8 h, and a heating rate of 2.5 °C/min. The same upper and lower boundary conditions, used to obtain the optimum density, were also employed for the investigation of the porosity, firing shrinkage, and green density. The optimum achieved values were of geometric mean of 6.89%, 3.48 g/cm^3, and 17.01% for the porosity, green density, and firing shrinkage, respectively.

Keywords Physical properties · Barium titanates · Optimum density · Porosity · Firing shrinkage and green density of $BaTiO_3$ · Genetic algorithm approach

Introduction

Barium titanate ($BaTiO_3$) is one of the most important electronic ceramic components and has been of practical interest for more than 60 years. This is because it has good characteristics, which are described as, firstly, its relatively simple crystal structure, secondly, it can exhibit ferroelectric properties at room temperature, and finally because it can be easily prepared as a polycrystalline ceramic. Barium titanates are used in capacitors and employed for many applications such as medical imaging, gas lighters, ultrasonic cleaning, and underwater detection [1–3].

Its dielectric properties are strongly influenced by the physical properties of the ceramics which generally include among others, the density and porosity. The presence of porosity in the ceramic material, for example, has an unfavorable effect on dielectrics, which have a high dielectric constant, since these pores contain air, and the dielectric constant (k) for air is very low. On the other hand, the ceramics which possess a high density seems to have better dielectric properties than the porous ones because of the little existence of porosity. Thus, it is desired to find the processing conditions which led to optimum density [1, 4, 5].

Based on these findings, a lot of time and cost has been spent in order to achieve the optimum density through experimental investigations. Thus, developing a model for conducting optimization is highly desirable.

There are many studies that are focused on the employment of a genetic algorithm (GA) for the optimization of problems, in the joining, and turning processes and additionally, in the field of metals. However, in the ceramics field, a GA is not commonly used and has rarely been documented. The GA

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approach begins with a set of randomly generated individuals in a named population. Each individual is called a chromosome, and every chromosome is evaluated using a certain function named the fitness function. After that, a new population is generated by employing the GA operator (selection, crossover, and mutation) which replaces the old population with the new one. This process is repeated until the optimum solution is obtained [6–9].

In the current work, a GA was used to find the values of the processing parameters that give a $BaTiO_3$ ceramic with optimum density, porosity, firing shrinkage, and green density. This work relies on the results that were achieved through experimental work and that include the preparation of five different batches of barium titanate powders. These batches were prepared by a modified pechini method to obtain different particle size distributions. The prepared samples were pressed at different pressing pressures and at different pressing rates. Subsequently, the pressed samples were sintered at different temperatures, with different heating rates, and at different soaking times. The concept of optimization was employed in this work using a GA approach in order to achieve the optimum values of the key parameters, namely, the density, porosity, firing shrinkage, and green density.

Experimental procedure

$BaTiO_3$ powder was prepared by modified pechini method according to following steps: (100 ml) barium chloride solution was prepared by dissolving the specific amount of $BaCl_2$ in 100 ml of distilled water, and then put on the magnetic stirring with an average speed of 1100 rpm for 30 min. The desired volume of titanium chloride solution was added to the solution of $BaCl_2$ under continuous stirring for additional 30 min. A solution of 100 ml of oxalic acid is prepared by dissolving prepared weight of $C_2H_2O_4$ in 100 ml of distilled water. This solution is then placed on magnetic stirring for 30 min with an average speed of 1100 rpm, it then added drop by drop into the mixed solution of $BaCl_2$ and $TiCl_3$ under continuous stirring to get nutty color precipitate of barium titanyl oxalate (BTO), according to the following reaction:

$$BaCl_2.2H_2O + TiCl_3 + 2C_2H_2O_4 + 2H_2O + 1/2O_2 \rightarrow BaTiO(C_2O_4)_2.4H_2O + 4HCl + 1/2Cl_2 \quad (1)$$

The precipitate (BTO) is then filtered and washed several time with distilled water and then dried at temperature 80 °C for 20 h. Finally, the precipitate (BTO) was fired at 850 °C for 2 h with heating rate of 3 °C/min to be thermally decomposed to yield a white powder of barium titanate. According to the equations below:

$$BaTiO(C_2O_4)2.4H_2O \rightarrow BaTiO(C_2O_4)2 + 4H_2O \quad (2)$$

$$2BaTiO(C_2O_4)2 \rightarrow Ba_2Ti_2O_5(CO_3)(CO_2) + 2CO_2 + 4CO \quad (3)$$

$$Ba_2Ti_2O_5(CO_3)(CO_2) \rightarrow Ba_2Ti_2O_5(CO_3) + CO_2 \quad (4)$$

$$Ba_2Ti_2O_5(CO_3) \rightarrow 2BaTiO_3 + CO_2 \quad (5)$$

Five different batches with different concentrations of barium chloride and titanate chloride were prepared in order to achieve different particle sizes of barium titanate powder. These batches are batch 1, batch 2, batch 3, batch 4, and batch 5.

The experimental population consisted of 80 samples, with each sample considered as a chromosome and every chromosome composed of number of genes. These genes were represented by the input parameters, in the current work; eight input parameters with five levels were used as shown in Table 1.

The fitness function or regression equation was formed using Minitab 17 software for density (y_1). The regression equation obtained for density can be expressed as follows:

$$\begin{aligned} y_1 = {} & -3.7 + 2.057^{*} x_{(1)} + 0.875^{*} x_{(2)} - 0.18^{*} x_{(3)} + 0.00742^{*} x_{(4)} + 0.0086^{*} x_{(5)} - 17.9^{*} x_{(6)} + 0.52^{*} x_{(7)} + 0.184^{*} x_{(8)} \\ & + 0.01050^{*} x_{(1)}{}^{*} x_{(2)} - 0.0984^{*} x_{(1)}{}^{*} x_{(3)} - 0.001220^{*} x_{(1)}{}^{*} x_{(4)} \\ & + 0.000892^{*} x_{(1)}{}^{*} x_{(5)} - 0.73^{*} x_{(1)}{}^{*} x_{(6)} - 0.0395^{*} x_{(1)}{}^{*} x_{(7)} \\ & + 0.0317^{*} x_{(1)}{}^{*} x_{(8)} - 0.0447^{*} x_{(2)}{}^{*} x_{(3)} - 0.000430^{*} x_{(2)}{}^{*} x_{(4)} - 0.000054^{*} x_{(2)}{}^{*} x_{(5)} - 0.484^{*} x_{(2)}{}^{*} x_{(6)} \\ & + 0.0079^{*} x_{(2)}{}^{*} x_{(7)} + 0.00816^{*} x_{(2)}{}^{*} x_{(8)} - 0.000980^{*} x_{(3)}{}^{*} x_{(4)} + 0.000495^{*} x_{(3)}{}^{*} x_{(5)} \\ & + 2.84^{*} x_{(3)}{}^{*} x_{(6)} - 0.107^{*} x_{(3)}{}^{*} x_{(7)} - 0.0273^{*} x_{(3)}{}^{*} x_{(8)} - 0.000005^{*} x_{(4)}{}^{*} x_{(5)} \\ & + 0.0128^{*} x_{(4)}{}^{*} x_{(6)} - 0.000251^{*} x_{(4)}{}^{*} x_{(7)} - 0.000174^{*} x_{(4)}{}^{*} x_{(8)} - 0.0080^{*} x_{(5)}{}^{*} x_{(6)} \\ & + 0.00075^{*} x_{(5)}{}^{*} x_{(7)} - 0.000206^{*} x_{(5)}{}^{*} x_{(8)} + 0.04^{*} x_{(6)}{}^{*} x_{(7)} \end{aligned}$$

Table 1 The processing parameters

Exp. No.	Batch. No.	Press (Mpa)	Press rate KN/S	Sintering Temp °c	Rate of Temp min/s	Soaking Time (h)	Bulk density gm/cm^3	Porosity %	Shrinkage %	Green density gm/cm^3
1	Batch #1	300	5	1000	5.0	3.0	3.55	37.029	8.264	3.440
2		150	15	1200	3.5	1.0	3.891	34.542	8.448	3.310
3		300	3	1300	2.5	4.0	5.343	8.245	14.906	3.440
4		150	3	1000	5.0	2.0	3.321	40.833	5.770	3.318
5		150	5	1100	4.0	1.5	3.311	42.000	6.082	3.318
6		250	5	1200	3.5	3.0	4.587	20.803	9.242	3.396
7		200	10	1000	2.5	4.0	4.442	23.212	8.780	3.347
8		200	5	1000	5.0	3.0	3.324	40.784	7.853	3.347
9		200	5	1000	5.0	2.0	3.633	35.651	5.742	3.347
10		250	15	1000	5.0	3.0	3.467	38.408	6.927	3.392
11		350	15	1200	3.0	4.0	5.000	12.943	10.062	3.490
12		350	3	1100	4.0	1.5	3.703	34.488	5.923	3.498
13		150	10	1100	5.0	1.0	2.664	52.747	5.178	3.316
14		200	20	1000	5.0	1.0	2.635	53.229	4.268	3.313
15	Batch #1	200	5	1100	4.0	1.5	3.410	39.355	6.080	3.347
16		250	10	1100	4.0	1.5	3.475	38.475	5.861	3.396
17	Batch #2	150	15	1200	3.5	3.0	4.312	25.372	9.165	3.299
18		150	5	1000	2.5	4.0	4.237	26.617	8.749	3.306
19		150	3	1000	5.0	2.0	2.682	52.448	5.726	3.306
20		200	15	1150	4.0	1.0	2.737	52.149	6.054	3.340
21		150	10	1200	3.5	1.0	3.764	36.458	8.439	3.299
22		200	5	1300	2.5	4.0	5.243	9.906	14.888	3.343
23		200	10	1200	3.0	4.0	4.742	18.229	10.042	3.343
24		250	15	1200	3.5	3.0	4.585	20.837	9.242	3.396
25		250	10	1000	5.0	3.0	2.852	48.624	8.197	3.399
26		250	3	1000	5.0	2.0	3.555	36.946	5.765	3.403
27		250	15	1300	2.5	4.0	5.315	8.710	14.678	3.396
28		350	3	1000	3.0	1.0	2.758	50.800	7.146	3.483
29		350	5	1200	3.0	4.0	4.914	14.372	10.093	3.481
30		150	10	1000	5.0	1.0	2.584	53.076	3.829	3.299
31		350	5	1200	3.5	3.0	4.842	15.568	9.351	3.481
32	Batch #3	300	5	1300	2.5	4.0	5.326	8.528	14.729	3.403
33		250	5	1000	3.0	1.0	2.593	52.453	5.162	3.379
34		350	15	1200	3.5	3.0	4.600	20.588	7.802	3.450
35		150	3	1000	2.5	4.0	4.175	26.647	8.795	3.291
36		150	3	1300	2.5	4.0	5.107	12.166	14.558	3.291
37		150	10	1000	5.0	3.0	2.781	49.803	8.105	3.289
38		250	5	1200	3.5	1.0	3.913	30.581	8.571	3.379
39		250	5	1200	3.0	4.0	4.757	17.980	10.037	3.379
40		250	15	1000	5.0	2.0	3.302	42.149	5.754	3.375
41		200	3	1300	2.5	4.0	5.242	9.923	14.597	3.347
42		200	3	1200	3.5	3.0	4.262	26.202	9.203	3.347
43		200	10	1200	3.0	4.0	4.742	18.229	9.906	3.320
44		150	3	1000	3.0	2.0	2.793	49.475	7.793	3.291
45		300	3	1150	4.0	1.0	2.703	51.099	6.090	3.417

Table 1 (continued)

Exp. No.	Batch. No.	Press (Mpa)	Press rate KN/S	Sintering Temp °c	Rate of Temp min/s	Soaking Time (h)	Bulk density gm/cm^3	Porosity %	Shrinkage %	Green density gm/cm^3
46		150	5	1100	5.0	1.0	2.627	52.362	5.129	3.290
47		150	20	1000	5.0	1.0	2.569	53.325	3.825	3.286
48	Batch #4	200	5	1000	2.5	4.0	4.013	29.338	8.775	3.340
49		300	15	1150	4.0	1.0	2.690	52.315	6.204	3.406
50		300	3	1300	2.5	4.0	5.287	9.176	14.773	3.407
51		300	10	1200	3.5	3.0	4.325	25.156	9.329	3.407
52		150	15	1300	2.5	4.0	5.000	13.943	14.462	3.266
53		150	3	1000	5.0	3.0	2.693	52.265	6.907	3.268
54		150	5	1000	5.0	2.0	3.000	47.166	5.882	3.267
55		350	3	1200	3.5	1.0	4.113	27.478	8.540	3.446
56		350	3	1000	4.0	3.0	3.481	38.176	9.595	3.446
57		350	10	1200	3.0	4.0	4.814	16.033	10.072	3.437
58		200	3	1000	5.0	3.0	3.223	43.461	8.042	3.340
59		200	5	1000	3.0	2.0	3.009	47.016	7.576	3.340
60		150	10	1000	5.0	1.0	2.538	53.840	3.918	3.267
61		250	3	1300	2.5	4.0	5.192	10.754	14.809	3.361
62	Batch #5	150	3	1000	3.0	2.0	2.784	49.754	7.710	3.253
63	Batch #5	150	5	1200	3.5	3.0	4.175	26.647	9.314	3.253
64		250	5	1000	5.0	2.0	2.813	49.272	5.731	3.320
65		150	15	1300	2.5	4.0	4.985	13.192	14.792	3.247
66		250	10	1200	3.5	1.0	3.965	30.136	8.598	3.335
67		250	3	1000	3.0	2.0	3.136	44.906	7.690	3.336
68		250	5	1200	3.0	4.0	4.614	20.355	10.035	3.336
69		250	15	1300	2.5	4.0	5.125	11.867	14.707	3.335
70		300	3	1000	2.5	4.0	4.150	27.063	8.816	3.396
71		350	3	1000	3.0	2.0	3.322	41.817	7.717	3.406
72		350	5	1200	3.5	3.0	4.362	24.541	9.441	3.403
73		350	3	1200	3.5	1.0	4.125	26.305	8.611	3.406
74		350	15	1300	2.5	4.0	5.231	10.106	14.878	3.403
75		150	15	1000	5.0	1.0	2.538	53.840	3.816	3.247
76		200	10	1100	5.0	1.0	2.627	53.362	5.112	3.297
77		250	10	1200	3.5	3.0	4.287	25.787	9.181	3.320
78		200	3	1000	3.0	2.0	2.811	49.305	7.746	3.306
79		300	5	1200	3.0	4.0	4.714	18.694	9.975	3.396
80		300	3	1000	3.0	2.0	3.211	43.661	7.612	3.396

where y_1: density, x_1: soaking time, x_2: rate of pressing, x_3: rate of sintering, x_4: sintering temperature, x_5: magnitude of pressing, x_6: D_{10}, x_7: D_{50}, x_8: D_{90}.

For the porosity, the regression equation is

$$\begin{aligned} y_2 = {} & 161 - 35.5\,x_{(1)} - 14.85\,x_{(2)} + 1.9\,x_{(3)} - 0.139^{*}\,x_{(4)} - 0.111\,x_{(5)} + 336\,x_{(6)} - 7.5\,x_{(7)} - 5.14\,x_{(8)} - 0.225\,x_{(1)}{}^{*}x_{(2)} \\ & + 1.873\,x_{(1)}{}^{*}x_{(3)} + 0.02161^{*}\,x_{(1)}{}^{*}x_{(4)} - 0.0135\,x_{(1)}{}^{*}x_{(5)} + 9.6\,x_{(1)}{}^{*}x_{(6)} + 0.73^{*}\,x_{(1)}{}^{*}x_{(7)} - 0.476\,x_{(1)}{}^{*}x_{(8)} \\ & + 0.773^{*}\,x_{(2)}{}^{*}x_{(3)} + 0.00768^{*}\,x_{(2)}{}^{*}x_{(4)} + 0.00113\,x_{(2)}{}^{*}x_{(5)} + 7.62\,x_{(2)}{}^{*}x_{(6)} - 0.080\,x_{(2)}{}^{*}x_{(7)} - 0.1527\,x_{(2)}{}^{*}x_{(8)} \\ & + 0.0169\,x_{(3)}{}^{*}x_{(4)} - 0.0065\,x_{(3)}{}^{*}x_{(5)} - 51.4\,x_{(3)}{}^{*}x_{(6)} + 1.79\,x_{(3)}{}^{*}x_{(7)} + 0.643\,x_{(3)}{}^{*}x_{(8)} \\ & + 0.000067^{*}\,x_{(4)}{}^{*}x_{(5)} - 0.205\,x_{(4)}{}^{*}x_{(6)} + 0.0018\,x_{(4)}{}^{*}x_{(7)} + 0.00410^{*}\,x_{(4)}{}^{*}x_{(8)} \end{aligned}$$

For the firing shrinkage (y_3), the regression equation is

$$\begin{aligned} y_3 = {} & -84.3 + 2.55^{*}\,x_{(1)} - 4.045^{*}x_{(2)} + 18.94^{*}\,x_{(3)} + 0.0950^{*}\,x_{(4)} - 0.0133^{*}\,x_{(5)} + 110.0^{*}x_{(6)} - 10.24^{*}\,x_{(7)} - 0.938^{*}\,x_{(8)} \\ & + 0.0072^{*}\,x_{(1)}{}^{*}x_{(2)} + 0.2816^{*}\,x_{(1)}{}^{*}x_{(3)} - 0.00289^{*}x_{(1)}{}^{*}x_{(4)} + 0.00149^{*}\,x_{(1)}{}^{*}x_{(5)} \\ & + 0.70^{*}\,x_{(1)}{}^{*}x_{(6)} - 0.172^{*}x_{(1)}{}^{*}x_{(7)} + 0.0172^{*}\,x_{(1)}{}^{*}x_{(8)} + 0.2998^{*}x_{(2)}{}^{*}x_{(3)} \\ & + 0.002636^{*}\,x_{(2)}{}^{*}x_{(4)} - 0.000251^{*}\,x_{(2)}{}^{*}x_{(5)} + 0.071^{*}x_{(2)}{}^{*}x_{(6)} \\ & + 0.0508^{*}\,x_{(2)}{}^{*}x_{(7)} - 0.0315^{*}\,x_{(2)}{}^{*}x_{(8)} - 0.01663^{*}\,x(3)^{*}x_{(4)} + 0.00202^{*}x_{(3)}{}^{*}x_{(5)} - 12.57^{*}\,x_{(3)}{}^{*}x_{(6)} \\ & + 0.257^{*}\,x_{(3)}{}^{*}x_{(7)} + 0.2494^{*}\,x_{(3)}{}^{*}x_{(8)} - 0.000011^{*}\,x_{(4)}{}^{*}x_{(5)} - 0.0796^{*}\,x_{(4)}{}^{*}x_{(6)} + 0.00444^{*}x_{(4)}{}^{*}x_{(7)} \\ & + 0.000580^{*}x_{(4)}{}^{*}x_{(8)} + 0.0219^{*}\,x_{(5)}{}^{*}x_{(6)} - 0.00002^{*}\,x_{(5)}{}^{*}x_{(7)} - 0.000161^{*}\,x_{(5)}{}^{*}x_{(8)} + 4.21^{*}\,x_{(6)}{}^{*}x_{(7)} \end{aligned}$$

For the green density (y_4), the regression equation is

$$\begin{aligned} y_4 = {} & 2.855 + 0.00135^{*}\,x_{(1)} + 0.000702^{*}\,x_{(2)} + 0.493^{*}\,x_{(3)} + 0.0787^{*}\,x_{(4)} - 0.00611^{*}\,x_{(5)} \\ & + 0.000003^{*}\,x_{(1)}{}^{*}x_{(2)} - 0.00527^{*}\,x_{(1)}{}^{*}x_{(3)} + 0.000148^{*}\,x_{(1)}{}^{*}x_{(4)} + 0.000048\,x_{(1)}{}^{*}x_{(5)} \\ & + 0.000419^{*}\,x_{(2)}{}^{*}x_{(3)} - 0.000031^{*}\,x_{(2)}{}^{*}x_{(4)} - 0.000004^{*}\,x_{(2)}{}^{*}x_{(5)} - 0.0920^{*}\,x_{(3)}{}^{*}x_{(4)} \end{aligned}$$

where (x_1): rate of press, (x_2): pressing pressure, (x_3): D_{10}, (x_4): D_{50}, and (x_5): D_{90}.

The GA parameters selected lead to the optimum solutions which are summarized in Table 2.

Results and discussion

XRD result

Figure 1 shows the result of x-ray diffraction analysis of barium titanate powder calcined at 850 °C with a heating rate of 3 °C/min and soaking time of 2 h. The calculated values of d-spacing and relative intensities obtained from this pattern are

Table 2 The genetic algorithm parameters

GA parameters	Type
Population size	80 Samples
Selection	Roulette wheel selection
Crossover	Two point crossover
Mutation	Uniform mutation
Mutation probability	0.2

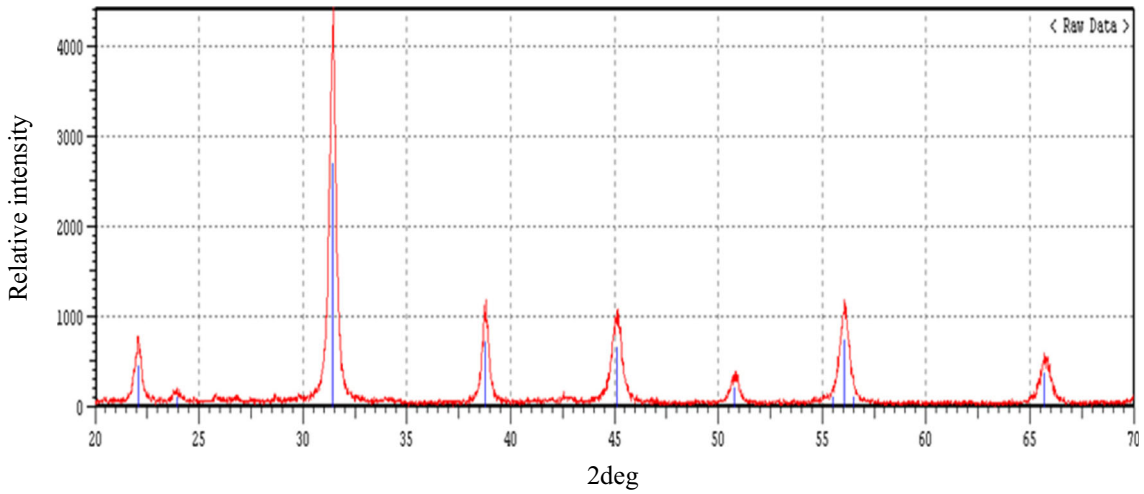

Fig. 1 XRD pattern of barium titanate powder calcined at 850 °C

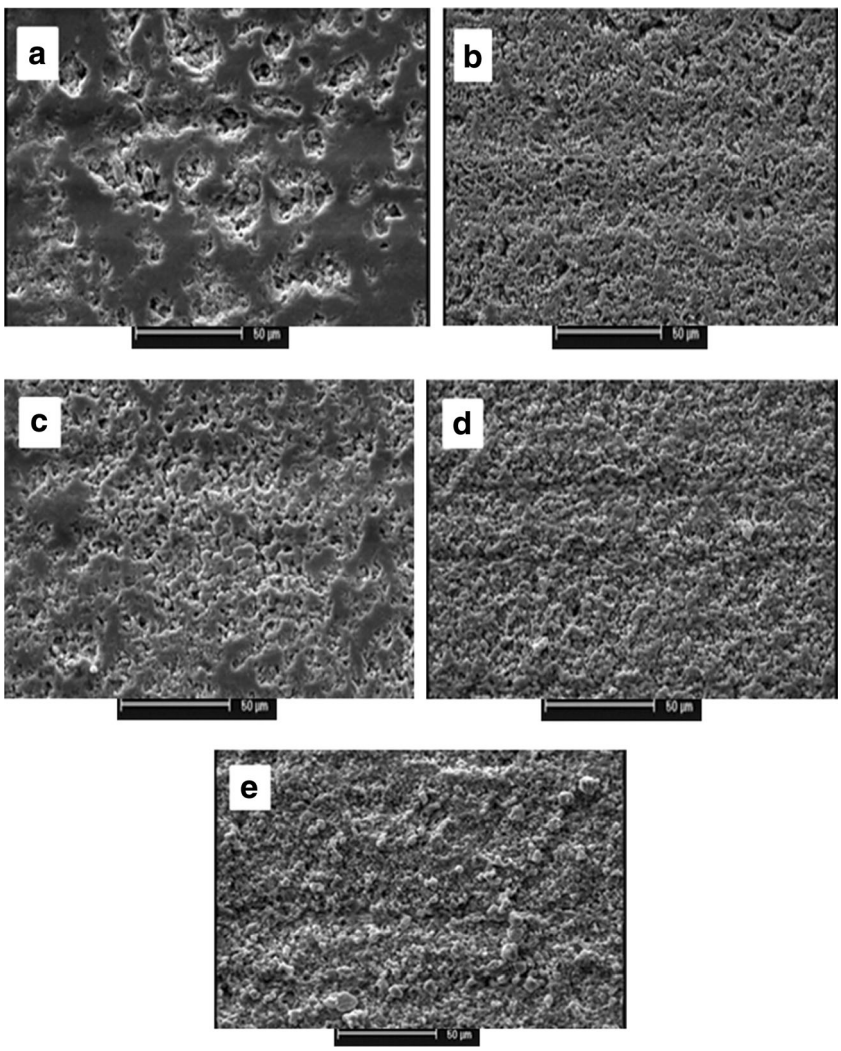

Fig. 2 SEM micrographs for BT samples sintered at 1200 °C for 1 h prepared from a solution of **a** 0.05 M, **b** 0.1 M, **c** 0.25 M, **d** 0.5 M, and **e** 0.97 M

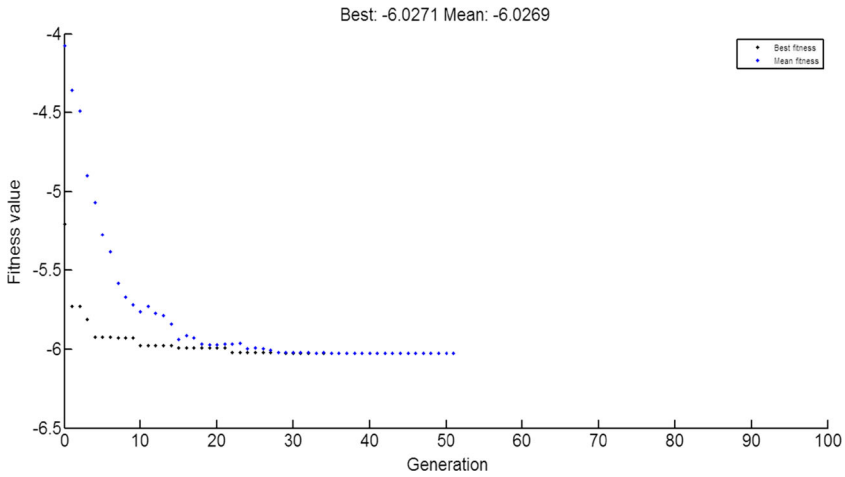

Fig. 3 The generations versus the fitness value

in good agreement with JCPDS Card No. 5-626 for tetragonal $BaTiO_3$.

DTA result

The differential thermal analysis (DTA) result for a sample of barium titanate indicates that there are four endothermic peaks. The first endothermic peak corresponds to dehydration of barium titanyl oxalate (BTO) (loss of water) in the range of 106–178 °C which is represented by the following equation:

$$BaTiO\,(C_2O_4)_2.4H_2O \rightarrow BaTiO\,(C_2O_4)_2 + 4H_2O \quad (6)$$

The second peak involves the thermal decomposition of the dehydrated oxalate in the range of 295–373 °C leading to the formation of intermediate carbonate which occurs in two steps according to eqs. (7) and (8):

$$2BaTiO\,(C_2O_4)_2 \rightarrow Ba_2Ti_2O_5(CO_3)\,(CO_2) + 2CO_2 + 4CO \quad (7)$$

$$Ba_2Ti_2O_5(CO_3)\,(CO_2) \rightarrow Ba_2Ti_2O_5(CO_3) + CO_2 \quad (8)$$

The third peak indicates the result of the decomposition of the intermediate carbonate resulting in the formation of barium titanate in the range of 663–713 °C according to eq. (9):

$$Ba_2Ti_2O_5(CO_3) \rightarrow 2BaTiO_3 + CO_2 \quad (9)$$

While the forth peak may be contributed to the decomposition of barium carbonate in the range of 773–787 °C.

$$BaCO_3 \rightarrow BaO + CO_2 \quad (10)$$

These results are matching with that obtained by Callagher et al. [10], who suggested a three-step mechanism associated with the formation of BT from barium titanyle oxalate.

Table 3 The optimum processing parameters for the different batches

Batch No.	Current iteration	Press (Mpa)	Soaking time (h)	Sintering temperature (°C)	Press rate (KN/s)	Rate of sintering (°C/min)
Batch 1	51	380	6	1405	3	2.5
Batch 2	82	370	6	1433	3	2.5
Batch 3	51	380	6	1440	3	2.5
Batch 4	52	381	7.7	1458	3	2.5
Batch 5	64	385	8	1462	3	2.5

SEM result

All the samples were polished and then chemically etched using a solution composed of 95 ml H2O + 3 ml HCl (32%) + 2 ml HF (40%) for period that ranged from 5 s to

Table 4 The optimum input and output parameters

Batch No.	Current iteration	Press (Mpa)	Soaking time (h)	Sintering temperature (°C)	Press rate (KN/s)	Rate of sintering (°C/min)	Porosity %
Batch 1	51	380	6	1405	3	2.5	7.83
Batch 2	51	370	6	1433	3	2.5	7.33
Batch 3	51	380	6	1440	3	2.5	7.20
Batch 4	51	381	7.7	1458	3	2.5	6.21
Batch 5	51	385	8	1462	3	2.5	6.04

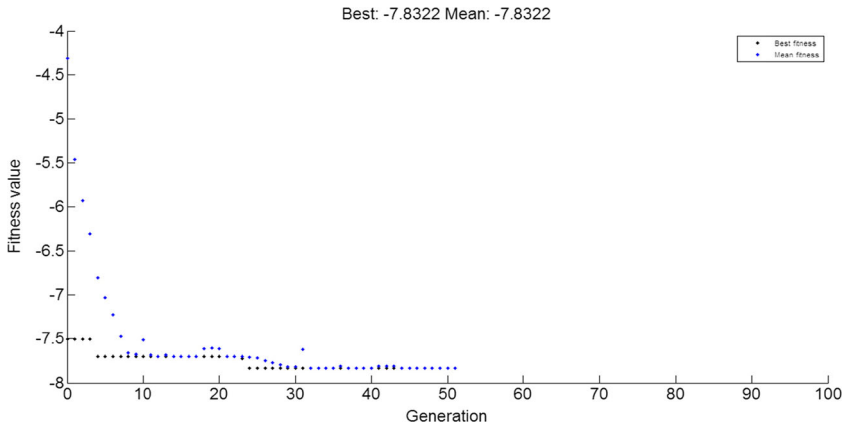

Fig. 4 The generations versus the fitness value for batch 1

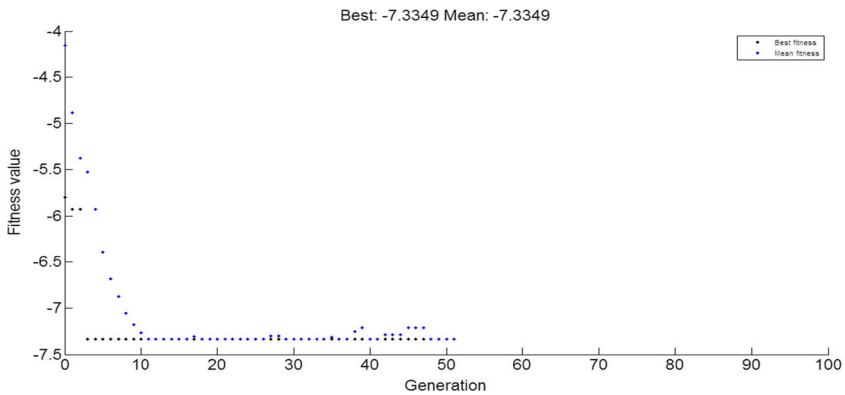

Fig. 5 The generations versus the fitness value for batch 2

2 min. Figure 2a–e shows the SEM micrographs of the surface of samples of barium titanate sintered at a temperature of 1200 °C with the heating rate of 3.5 °C/min for a soaking time of 1 h for the batches 1, 2, 3, 4, and 5, respectively. The SEM image shows that the microstructure of each sample consists of a large network of interconnected grains; these grains have a multi-faceted shape, on average, with homogenous distribution of porosity and with a nearly narrow size distribution. It can be seen that there is a clear effect of the reactant concentration on the average size of the grains and the interconnectivity of the grains. With increasing the concentration of the precursors, the grain size increases. This is due to the larger size of the prepared powder used to prepare the sintered samples. In contract, the interconnectivity increases with

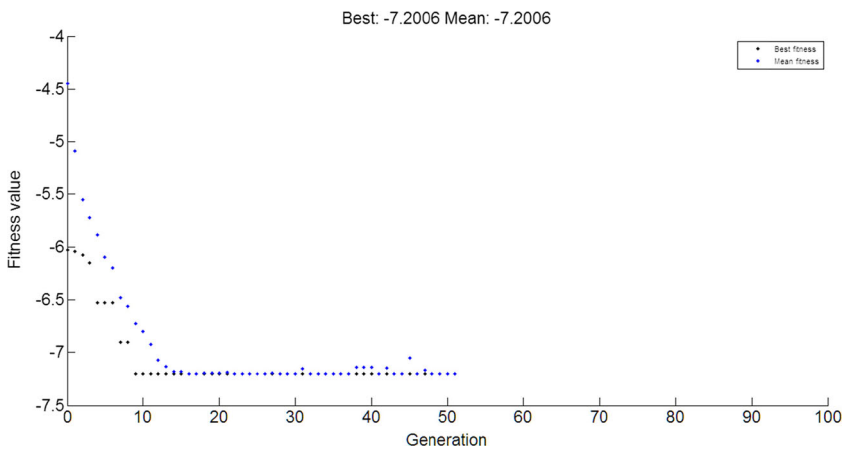

Fig. 6 The generations versus the fitness value for batch 3

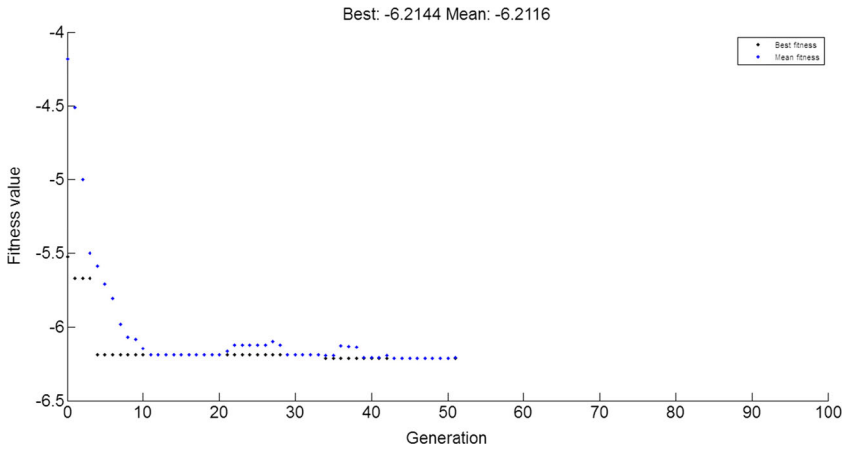

Fig. 7 The generations versus the fitness value for batch 4

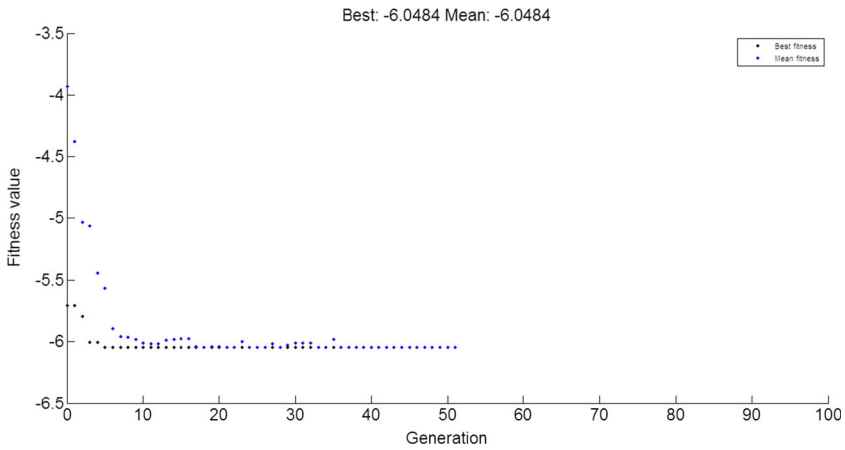

Fig. 8 The generations versus the fitness value for batch 5

decrease of the concentration of precursors. This is because of the finer particle size of the powder obtained at low concentration has higher surface area leading to more efficient sintering process.

The results of the genetic algorithm

The results of a GA can be a representation as a sketch of the fitness value and the number of the generation, generally as the generations progress, the fitness value begins to stabilize, at a certain value that represents the optimum value.

The results of the GA for optimum density

The GA shows that the best fit for the maximization of the density at a population size of 80 is found to be 6.02 g/cm^3 which is equal to the theoretical density of barium titanate. As shown in the Fig. 3, the optimum combinations of input

Table 5 the optimum input and output parameters

Batch No.	Current iteration	Press (Mpa)	Press rate (KN/s)	Green density gm/m^3
Batch 1	76	380	3	3.51
Batch 2	74	370	3	3.49
Batch 3	51	380	3	3.48
Batch 4	51	381	3	3.47
Batch 5	52	385	3	3.44

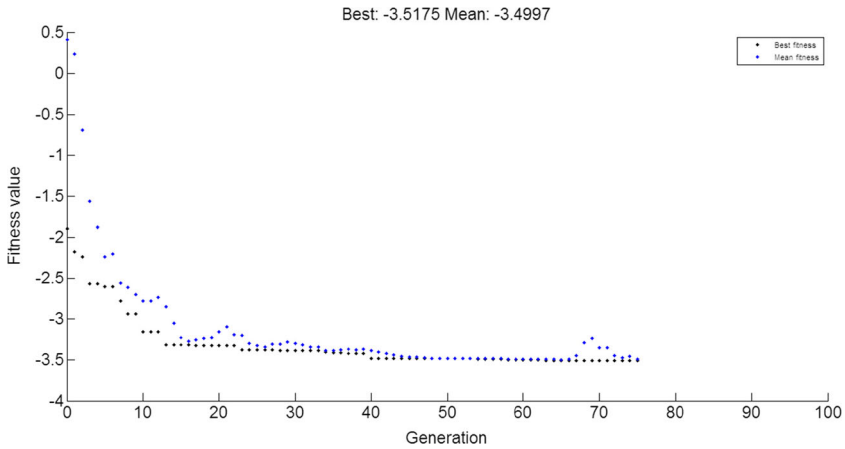

Fig. 9 The generations versus the fitness value for batch 1

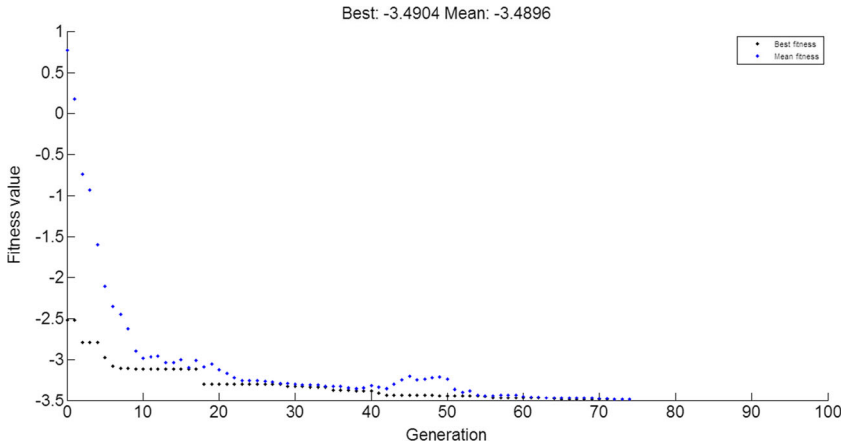

Fig. 10 The generations versus the fitness value for batch 2

parameters for the maximization of the density of each batch of the barium titanate samples are summarized in Table 3. These results suggest that, regardless of the particle size distribution, the pressing and sintering rates must not exceed 3 KN/s and 2.5 °C/min, respectively. Moreover, the forming pressure should be in the range 370–385Mpa.

The soaking time for the fine powder was 6 h while it was 8 h for the coarse powders, and the sintering temperature must

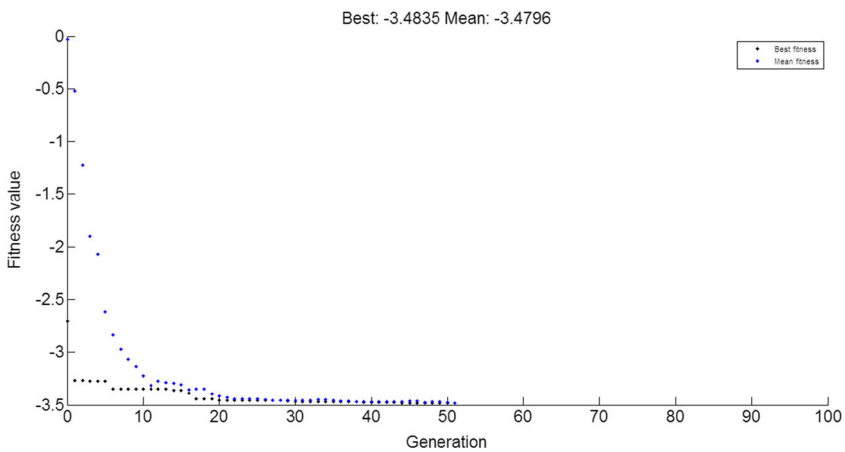

Fig. 11 The generations versus the fitness value for batch 3

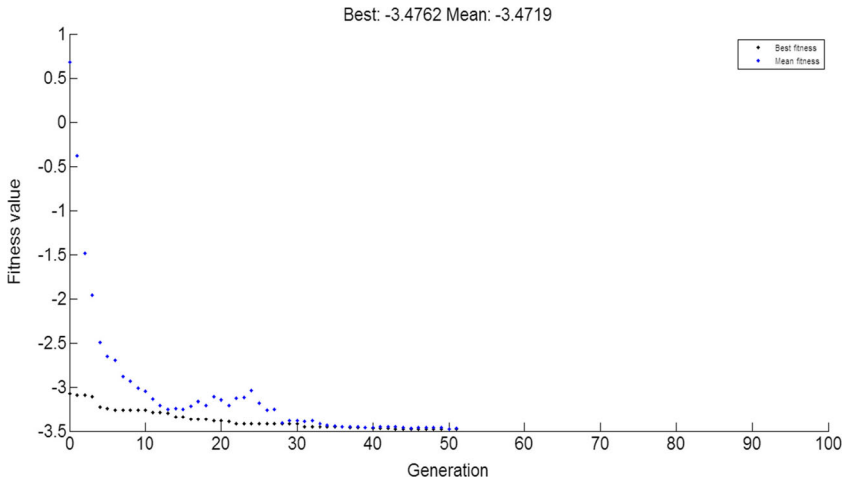

Fig. 12 The generations versus the fitness value for batch 4

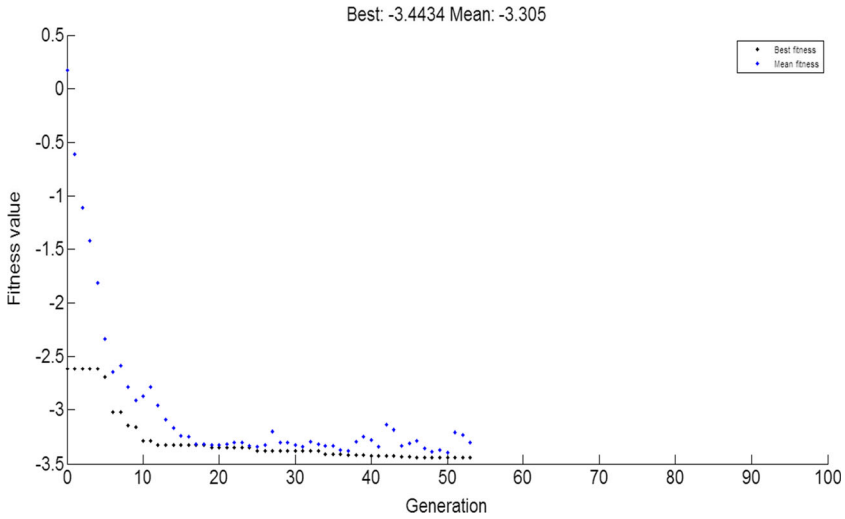

Fig. 13 The generations versus the fitness value for batch 5

not be less than 1400 °C. This is in order to avoid the reconstructive phase transformation for the hexagonal crystal structure. It is obvious that in order to obtain BT ceramics with bulk density equal to the theoretical density, using the traditional ceramic technology, then extreme values of the processing parameters must be selected.

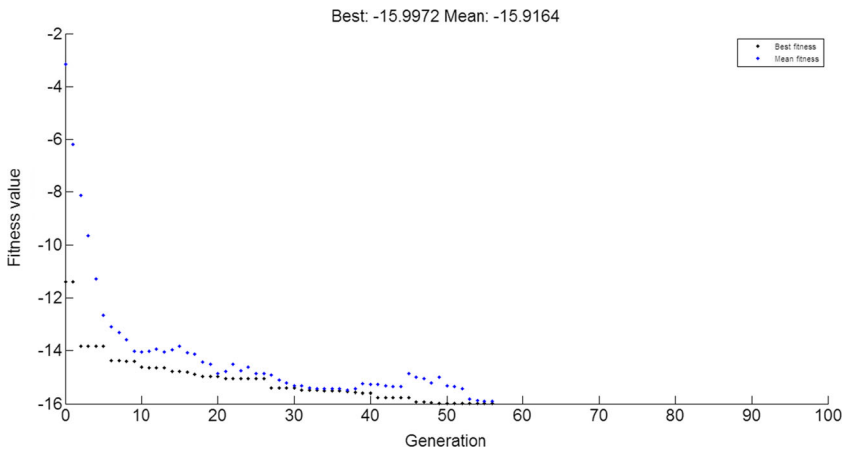

Fig. 14 The generations versus the fitness value for batch 1

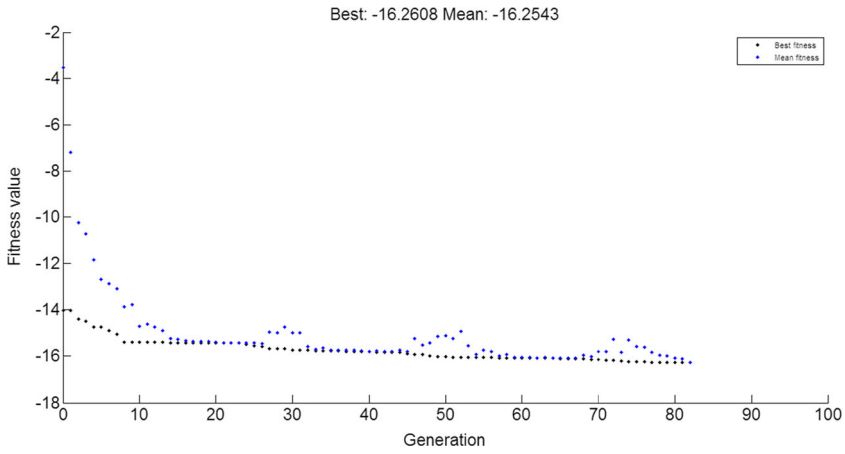

Fig. 15 The generations versus the fitness value for batch 2

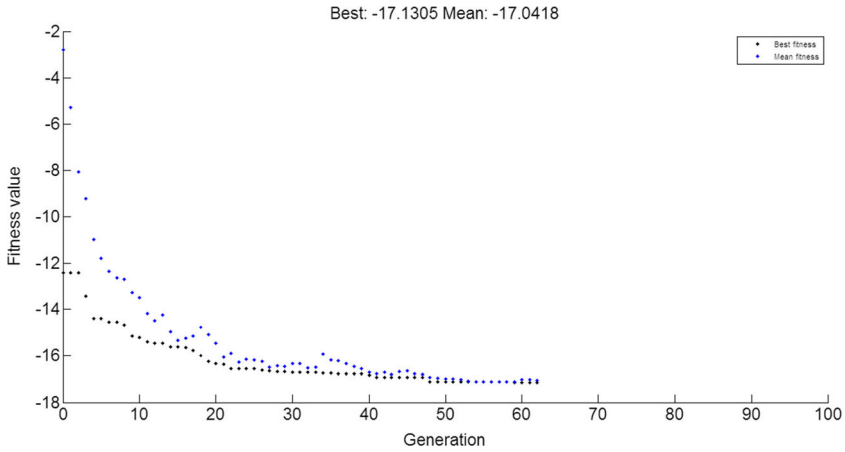

Fig. 16 The generations versus the fitness value for batch 3

The results of the GA for optimum porosity

For the porosity, the boundaries were taken as the same as that for the bulk density; the optimum input and output parameters are summarized in Table 4. It can be observed that the input parameters obtained match those obtained for the optimum density; however, the optimum porosities obtained do not match with the density results because they are not equal to zero as expected

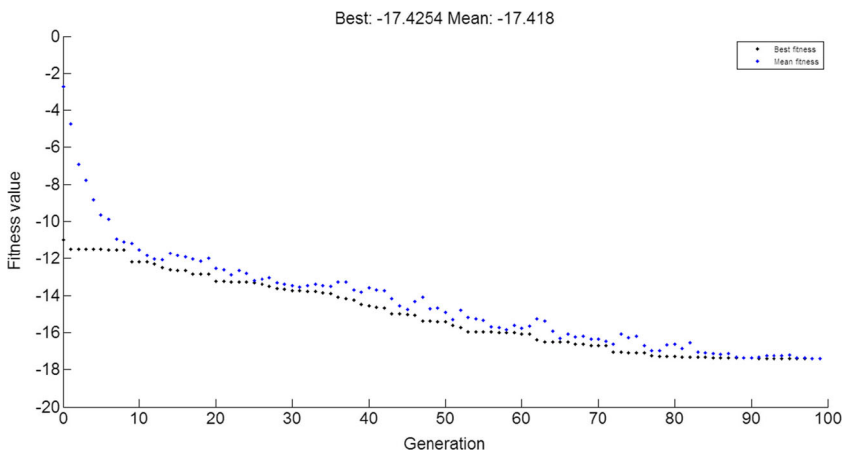

Fig. 17 The generations versus the fitness value for batch 4

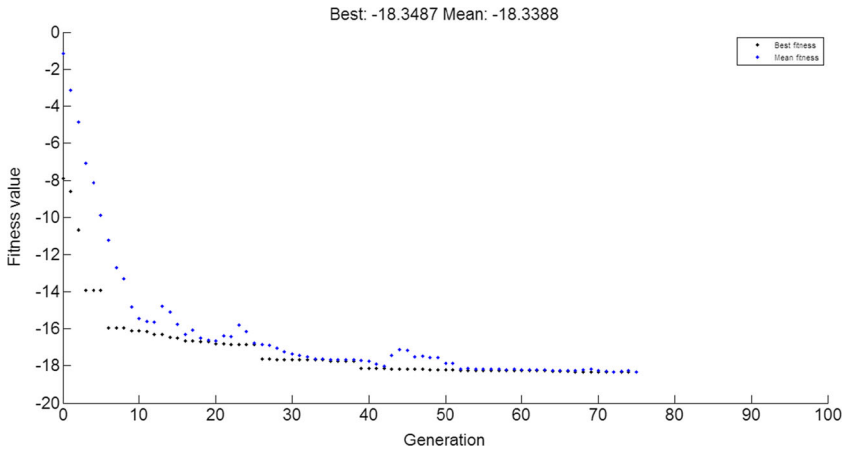

Fig. 18 The generations versus the fitness value for batch 5

when the bulk density is equal to the theoretical density. This may be due to the uncertainty in the values of the porosity, due to the nature of ASTM method, which was used to develop the model, as shown in the Figs. 4, 5, 6, 7, and 8.

The results of the GA for optimum green density

In the case of the green density, the upper and lower boundaries of the pressing pressure and pressing rate were fixed to the same values as those used for the bulk density analysis. The optimum processing parameters obtained and the optimum green density for each particle size distribution are summarized in Table 5**,** and the results are shown in the Figs. 9, 10, 11, 12, and 13.

The values obtained for the pressing pressure and pressing rate are similar to that obtained for the bulk density as given in Table 3

The results of the GA for optimum firing shrinkage

For the firing shrinkage, the results are shown in Figs. 14, 15, 16, 17, and 18, the boundaries employed were similar to that of the bulk density, the optimum processing parameters achieved and the optimum firing shrinkage for each batch are summarized in Table 6.

The correlations between the green density, firing shrinkage, and porosity are described in Table 7

The high values of R^2 indicated that there is a correlation between the predicted optimum values. This enhances the

Table 6 The optimum input and output parameters for the different batches

Batch No.	Current iteration	Press (Mpa)	Soaking time (h)	Sintering temperature (°C)	Press rate (KN/s)	Rate of sintering (°C/min)	Shrinkage %
Batch 1	57	380	6	1405	3	2.5	15.99
Batch 2	82	370	6	1433	3	2.5	16.26
Batch 3	62	380	6	1440	3	2.5	17.13
Batch 4	100	381	7.7	1458	3	2.5	17.42
Batch 5	76	385	8	1462	3	2.5	18.34

Table 7 The correlation among the output parameters

Parameters	Equations	R^2%
Porosity and green density	Porosity = −88.7 + 27.51 green density	86.18
Porosity and firing shrinkage	Porosity = 19.69–0.750 firing shrinkage	84.88
Firing shrinkage and green density	Firing shrinkage = 140.6–35.53 green density	95.23

confidence of the predicted values as the correlations obtained matched with the expected correlations between the parameters.

Conclusion

High purity perovskite barium titanate with a tetragonal phase was successfully prepared using the modified pechini method. The results showed that the particle size and the particle size distribution of BT can be controlled by adjusting the concentration of the precursor solution in the pechini method. The particle size distribution obtained was a bimodal distribution that contained two combined unimodal components. The increase in the concentration of the precursors enhances the coarser particle component of the model at the expense of that of the finer particles. Traditional ceramic technology, for the micron and submicron powders of BT, demands the use of extreme values of the processing parameters to obtain highly dense ceramics according to the results of the genetic algorithm.

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