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S. Adel: Optimization of solid-state synthesis process of advanced ceramic materials: influence of mixing condition

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# **OPTIMIZATION OF SOLID-STATE SYNTHESIS PROCESS OF ADVANCED CERAMIC MATERIALS:** INFLUENCE OF MIXING CONDITIONS

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#### Resume

In this paper, the effect of mixing process on solid state reaction of solid oxide material mixture was studied. Lead piezoelectric ceramic specimens 0.5 Pb(Zn<sub>1/3</sub>,Sb<sub>2/3</sub>)O3-0.5 Pb<sub>0.98</sub>La<sub>0.02</sub>(Zr<sub>0.48</sub>,Ti<sub>0.52</sub>)O<sub>3</sub> prepared by different mixing procedures, were conducted under different conditions such as order, combination and mixing time. The phase formation, composition nature, structural properties of powder mixture was analyzed by X-ray diffraction. The obtained results for different mixing processes make the solid state reaction method more selective, taking into consideration the attraction forces between the reactants and the electronegativity of oxide reactants.

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# **1. Introduction**

The most widely used piezoelectric ceramics for these applications are those based on lead titanate zirconate  $x PbZrO_3 - (1-x) PbTiO_3$ solid solution, also known as PZT, because of their good properties and the wide possibilities to modify their structure and properties using various dopants [1, 2].

Different methods were developed to synthesize ceramic powders, including chemical and physical processes [3, 4]. The simplest physical method is the mechanical milling, where mechanical action is used to refine the powders [5 - 8]. In comparison, chemical process, such as solid-state reaction and various wet-chemical solution routes are the most widely employed to synthesize advanced ceramics in general [9, 10]. For solid-state reaction methods, the use of high energy milling in recent years has shown certain advantages in reducing the phase formation temperatures of ceramic materials, because transparent ceramics have

usually relatively high reaction temperatures [11, 12].

The comparison between the different synthesis methods, leads to conclude that the solid state reaction method is characterized by poor control of composition, morphology and reactivity [13, 14].

In this paper, an attempt has been made to systematically study the effect of different mixing processes for the solid state reaction method on the structural properties of PZS-PLZT ceramics. The samples were synthesized by solid state reaction method, their nature; structural properties and selectivity were investigated.

# 2. Experimental

For this study, (1-x)PZS-xPLZT system was chosen [15, 16], with 0.5Pb (Zn<sub>1/3</sub>, Sb<sub>2/3</sub>) O<sub>3</sub>-0.5Pb<sub>0.98</sub>La<sub>0.02</sub> (Zr<sub>0.48</sub>, Ti<sub>0.52</sub>) O<sub>3</sub> composition. It was synthesized from high purity oxide powders namely, PbO (99.90 % purity), ZrO2 (99.90 % purity), TiO<sub>2</sub> (99.80 % purity), ZnO (99.90 %

purity) from Sigma Aldrich, Sb<sub>2</sub>O<sub>3</sub> (99.90 % purity) and La<sub>2</sub>O<sub>3</sub> (99.90 % purity) from Alfa Aesar. The samples were prepared by the conventional ceramic process (solid-state method) using four different mixing combinations (see table 1 processes 1, 2, 3, 4, 5, 6, 7 and 8). Steechiometric amounts of metal oxides were mixed, ball-milled, and calcined at 800 °C (2 °C/min) for 120 min. The calcined powder was then ball-milled for 4 hours until very fine (small) particles are obtained. After drying, the powder was pressed and disk-like shaped then sintered at 1200 °C (heating rate = 2 °C/min) during two hours in a closed alumina crucible using a high temperature programmable furnace. It is to be noted that a lead loss is possible by evaporation of PbO which is very volatile at  $T \ge 900$  °C. To limit this effect; a PbO overloaded medium was maintained with PbZrO<sub>3</sub> powder to reduce this loss during sintering. A Bruker-axe X-ray diffraction analyzer, D8 with Cu Ka radiation  $(\lambda = 1.5406 \text{ Å})$  was used. The presence of perovskite structure and phase composition in samples were detected by Rietveld method using the crystallographic and geometric properties of resulting compounds.

# 3. Mixing combinations

In every process, each oxide should be mixed in an alcohol medium for two hours at 100 °C. After that, every two oxides were mixed and stirred in the same conditions. Finally, the resulting three mixtures (each mixture containing two oxides which were already agitated) were mixed and stirred in an alcohol medium for two hours at 100 ° C. The final mixture was milled for 30 minutes; then stirred during the same period and finally milled again for two hours. Eight mixing processes based on the choice of the addition order of oxides for the mixing operation were proposed (see Table 1 process 1, 2, 3, 4, 5, 6, 7 and 8).

# 4. Results and discussion

The purity and crystallinity of the synthesized samples were further examined

by the powder XRD technique. The XRD patterns of the 0.5Pb (Zn<sub>1/3</sub>, Sb<sub>2/3</sub>) O3-0.5Pb<sub>0.98</sub>La<sub>0.02</sub> (Zr<sub>0.48</sub>, Ti<sub>0.52</sub>) O<sub>3</sub> specimens sintered at 1200 °C after using different mixing procedure are shown in Fig. 1 (process 1, 2, 3, 4, 5, 6, 7 and 8). The different mixing procedures obviously affected the nature and the microstructure properties of sintered samples.

The obtained compounds (phases) with quantification percentage are shown in Table 2. Varying the mixing procedure affects chemical kinetic formation of resulting compounds. This may be due to the attraction forces between different solid powder reactants [17 - 19].

Fig. 2 shows the quantitative percentage of PZT and PLZT depending on the mixing procedure. The results reveal that the mixing procedure influenced the nature of formed compound. The mixing processes (1, 3, 5, 6, 7 and 8) led to produce the PZT compounds, while (2) and (4) processes were found to lead to PLZT compounds. It can be seen that the mixing process can provide greater selectivity in the reaction system.

Obviously, the mixing procedures influence not only the nature of the phase and microstructure, but also Zr/Ti composition. As for the mixing procedures (1, 2, 5, 6, 7 and 8), the percentage of zirconium is greater than that of titanium in the formed phases. But for the third and fourth processes (3, 4), the Zr percentage was found less than that of Ti (see Table 3).

The total electronegativity of the oxide  $(E_{ox})$  was determined by Portier using the following equation (1) [20, 21]:

$$\mathbf{E}_{ox} = ((X_M)^m * (X_0)^n)^{\binom{1}{(m+n)}}$$
(1)

Where  $X_M$  and  $X_O$  are the electronegativities of the cation and oxygen anion, respectively, and *m* is the stoichometery of the cation in the oxide while *n* is the stoichiometry of the oxygen anion in the oxide. So the Table 4 shows the approximate values of  $(E_{ox})$  (oxide reactant electronegativity) and mass composition of the samples.

	Process 01	Process 02	Process 03	Process 04	Process 05	Process 06		Process 07	Process 08
Step 01: Agitation at 100°C in alcohol medium (2hours), for each oxide	PbO;TiO <sub>2</sub> ZrO <sub>2</sub> ;Sb <sub>2</sub> O <sub>3</sub> ZnO;La <sub>2</sub> O <sub>3</sub>	PbO;TiO <sub>2</sub> ZrO <sub>2</sub> ;Sb <sub>2</sub> O <sub>3</sub> ZnO;La <sub>2</sub> O <sub>3</sub>	PbO;TiO <sub>2</sub> ZrO2;Sb2O3 ZnO;La <sub>2</sub> O <sub>3</sub>	PbO;TiO <sub>2</sub> ZrO2;Sb2O3 ZnO;La <sub>2</sub> O <sub>3</sub>	PbO;TiO <sub>2</sub> ZrO <sub>2</sub> ;Sb <sub>2</sub> O <sub>3</sub> ZnO;La <sub>2</sub> O <sub>3</sub>	PbO;TiO <sub>2</sub> ZrO <sub>2</sub> ;Sb <sub>2</sub> O <sub>3</sub> ZnO;La <sub>2</sub> O <sub>3</sub>		PbO;TiO <sub>2</sub> ZrO <sub>2</sub> ;Sb <sub>2</sub> O <sub>3</sub> ZnO;La <sub>2</sub> O <sub>3</sub>	PbO;TiO <sub>2</sub> ZrO <sub>2</sub> ;Sb <sub>2</sub> O <sub>3</sub> ZnO;La <sub>2</sub> O <sub>3</sub>
Step 02: Mixing at 100°C in alcohol medium	(PbO+TiO <sub>2</sub> ); (ZrO <sub>2</sub> +Sb <sub>2</sub> O <sub>3</sub> ); (ZnO+La <sub>2</sub> O <sub>3</sub> )	(PbO+La <sub>2</sub> O <sub>3</sub> ); (ZrO <sub>2</sub> +Sb <sub>2</sub> O <sub>3</sub> ); (ZnO+ TiO <sub>2</sub> )	(PbO+Sb <sub>2</sub> O <sub>3</sub> ); (ZrO <sub>2</sub> +La <sub>2</sub> O <sub>3</sub> ); (ZnO+ TiO <sub>2</sub> )	(PbO+La <sub>2</sub> O <sub>3</sub> ); (ZrO <sub>2</sub> +TiO <sub>2</sub> ); (ZnO+Sb <sub>2</sub> O <sub>3</sub> )	(La <sub>2</sub> O <sub>3</sub> +Sb <sub>2</sub> O <sub>3</sub> ); (ZnO+ TiO <sub>2</sub> ); (ZrO <sub>2</sub> + PbO)	(ZrO <sub>2</sub> +La <sub>2</sub> O <sub>3</sub> ); (PbO+Sb <sub>2</sub> O <sub>3</sub> )	alcohol ing(2hrs)	$(ZrO_2+La_2O_3)$ $(ZrO_2+La_2O_3+TiO_2)$	(PbO+Sb <sub>2</sub> O <sub>3</sub> ) (PbO+Sb <sub>2</sub> O <sub>3</sub> +ZnO)
Step 03:	(PbO+TiO <sub>2</sub> ); (ZrO2+Sb2O3):	$(PbO+La_2O_3);$ $(ZrO_2+Sb_2O_3):$	$(PbO+Sb_2O_3);$ $(ZrO_2+La_2O_3):$	(PbO+La <sub>2</sub> O <sub>3</sub> ); (ZrO <sub>2</sub> +TiO <sub>2</sub> ):	$(La_2O_3+Sb_2O_3);$ $(ZnO+TiO_2):$	(ZrO <sub>2</sub> +La <sub>2</sub> O <sub>3</sub> );	ng at 100°C in m (2hrs) /Mill	(ZrO <sub>2</sub> +La <sub>2</sub> O <sub>3</sub> + TiO <sub>2</sub> +ZnO)	(PbO+Sb <sub>2</sub> O <sub>3</sub> +ZnO+ TiO <sub>2</sub> )
(2hours)	$(ZnO+La_2O_3)$	$(ZnO+TiO_2)$	(ZnO+ TiO <sub>2</sub> )	$(ZnO+Sb_2O_3)$	(ZrO <sub>2</sub> + PbO)	(PbO+Sb <sub>2</sub> O <sub>3</sub> )	Mixi mediu	$(ZrO_2+La_2O_3+TiO_2+ZnO+Sb_2O_3)$	(PbO+Sb <sub>2</sub> O <sub>3</sub> +ZnO+ TiO <sub>2</sub> +ZrO <sub>2</sub> )
<b>Step 04:</b> Mixing at 100°C in alcohol medium (2hours)	$\begin{array}{l} (PbO+TiO_2+\\ ZrO_2+Sb_2O_3+\\ ZnO+La_2O_3) \end{array}$	$\begin{array}{l} (PbO+La_2O_3+\\ ZrO_2+Sb_2O_3+\\ ZnO+TiO_2) \end{array}$	$\begin{array}{l} (PbO+Sb_2O_3+\\ ZrO_2+La_2O_3+\\ ZnO+TiO_2) \end{array}$	$\begin{array}{l} (PbO+La_2O_3+\\ ZrO_2+TiO_2+\\ ZnO+Sb_2O_3) \end{array}$	(La2O3+Sb2O3+ ZnO+ TiO2+ ZrO2+ PbO)	(ZrO <sub>2</sub> +La <sub>2</sub> O <sub>3</sub> +TiO <sub>2</sub> ); (ZnO+PbO+Sb <sub>2</sub> O <sub>3</sub> )	(Z TiO <sub>2</sub> +Z	ZrO <sub>2</sub> +La <sub>2</sub> O <sub>3</sub> + ZnO+ Sb <sub>2</sub> O <sub>3</sub> +PbO)	(PbO+Sb <sub>2</sub> O <sub>3</sub> +ZnO+ TiO <sub>2</sub> +ZrO <sub>2</sub> + La <sub>2</sub> O <sub>3</sub> )
Step 05: Step 06: Step 07: Step 08:			Millin Mixi Millin	g in alcohol medium ing in alcohol mediun g in alcohol mediun Calcination at 80	for (0.5hour) : (PbO- um (2hours) : (PbO+T n for (2hours) : (PbO+ $0^{\circ}C$ : (PbO+TiO <sub>2</sub> +Zr	+TiO <sub>2</sub> +ZrO <sub>2</sub> +Sb <sub>2</sub> O <sub>3</sub> +ZnO TiO <sub>2</sub> +ZrO <sub>2</sub> +Sb <sub>2</sub> O <sub>3</sub> +ZnO+I -TiO <sub>2</sub> +ZrO <sub>2</sub> +Sb <sub>2</sub> O <sub>3</sub> +ZnO- O <sub>2</sub> +Sb <sub>2</sub> O <sub>3</sub> +ZnO+I, $a_2O_3$ )	+La <sub>2</sub> O <sub>3</sub> ) La <sub>2</sub> O <sub>3</sub> ) +La <sub>2</sub> O <sub>3</sub> )		

### Different mixing process.

Table 1



c) mixing procedure 3 Fig. 1. XRD patterns of sintered samples for different mixing procedures 1, 2, 3, 4, 5, 6, 7, and 8. (full colour version available online)



f) mixing procedure 6 Continuing of Fig. 1. XRD patterns of sintered samples for different mixing procedures 1, 2, 3, 4, 5, 6, 7, and 8. (full colour version available online)



Continuing of Fig. 1. XRD patterns of sintered samples for different mixing procedures 1, 2, 3, 4, 5, 6, 7, and 8. (full colour version available online)



Fig. 2. Quantitative percentage of PLZT and PZT as a function of mixing process.

# Table 2

	quantitative percentage								
Resulting compounds	Process 1	Process 2	Process 3	Process 4	Process 5	Process 6	Process 7	Process 8	
$Pb(Zr_{0.9}Ti_{0.1})O_3$	38.6%	14.5%	6.3%	19%	0%	12.7%	0%	20.9%	
$Pbt(Zr_{0.58}Ti_{0.42})O_3$	2.6%	0%	3.5%	0%	26.4%	49.1%	53.9%	26.7%	
$(Pb_{0.9}La_{0.1})(Zr_{0.3}Ti_{0.7})O_3$	1.3%	49.5%	0%	42.9%	0%	7.3%	7.6%	13.3%	
Pb ( $Zr_{0.97}Ti_{0.03}$ ) $O_3$	17%	0%	75.6%	0%	0%	20%	2.6%	0%	
$Pb(Zr_{0.75}Ti_{0.25})O_3$	2.7%	8%	0%	0%	24.8%	0%	0%	0%	
Pb ( $Zr_{0.7}Ti_{0.3}$ ) $O_3$	37.7%	0%	0%	0%	0%	0%	0%	0%	
$(Pb_{0.92}La_{0.08})(Ti_{0.392}Zr_{0.588})O_{3}$	0%	5.2%	3.5%	11.3%	0.6%	10.9%	25.5%	16.3%	
Pb ( $Zr_{0.44}Ti_{0.56}$ ) $O_3$	0%	22.8%	11.1%	11.4%	48.2%	0%	0%	22.7%	
Pb ( $Zr_{0.838}Ti_{0.162}$ ) $O_3$	0%	0%	0%	15.5%	0%	0%	0%	0%	
Pb ( $Zr_{0.601}Ti_{0.399}$ ) $O_3$	0%	0%	0%	0%	0%	0%	0%	0%	
Pb ( $Zr_{0.796}Ti_{0.204}$ ) $O_3$	0%	0%	0%	0%	0%	0%	10.4%	0%	
Total	≈100% (99.9%)	≈100%	≈100%	≈100% (100.1%)	≈100%	≈100%	≈100%	≈100% (99.9%)	

Relative quantitative percentage of PZT and PLZT formed phases (relative to all the obtained PZT based phases) as a function of mixing process.

Table 3

Percentage of Zr and Ti in the PZT and PLZT phases formed as a function of mixing process.										
$\mathbf{Q}_{\%\mathbf{Z}\mathbf{r}>\%\mathbf{T}\mathbf{i}} = \left[\sum_{w} \%Pb\left(Zr_{x}, Ti_{y}\right) + \sum_{w} \%Pb La\left(Zr_{x'}, Ti_{y'}\right)\right] with: x > y and x' > y'$										
$\mathbf{Q}_{\%\mathbf{T}_1 > \%\mathbf{Z}\mathbf{r}} = \left[\sum \%Pb(Zr_x, Ti_y) + \sum \%Pb La(Zr_{x'}, Ti_{y'})\right] with: y > x and y' > x'$										
	Process 1 Process 2 Process 3 Process 4 Process 5 Process 6 Process 7 Process 8									
<b>Q%Zr &gt; %</b> Ti	98.7%	27.7%	88.9%	45.7%	51.8%	92.7%	92.4%	64%		
Q%Ti > %Zr	1.3%	72.3%	11.1%	54.3%	48.2%	7.3%	7.6%	36%		

Table 4

Approximate values of (oxide reactant electronegativity) and mass.									
	mass of oxide used for each sample (g)	<i>n</i> (mole)	Oxide Electronegativity (ev or 1.602E-19 J) for 1mole of oxide (based on Pauling scale)	(E <sub>ox</sub> ) (ev or 1.602E-19 J) (Oxide Electronegativity*n)					
PbO	6.5959	0.0295	2.831	0.08351					
TiO <sub>2</sub>	0.6264	0.0078	2.631	0.02052					
ZrO <sub>2</sub>	0.8828	0.0071	2.506	0.01779					
Sb <sub>2</sub> O <sub>3</sub>	1.4415	0.0049	2.796	0.01370					
ZnO	0.4048	0.0049	2.382	0.01167					
La <sub>2</sub> O <sub>3</sub>	0.0483	0.000148	2.18	0.00032					

#### Table 5

Absolute values of $\Delta(E_{ox})$ (difference between oxide reactant electronegativity).									
Absolute $\Delta(E_{ox})$ (ev)	PbO	TiO <sub>2</sub>	ZrO <sub>2</sub>	Sb <sub>2</sub> O <sub>3</sub>	ZnO	La <sub>2</sub> O <sub>3</sub>			
PbO	-	0.06299	0.06572	0.06981	0.07184	0.08319			
TiO <sub>2</sub>	0.06299	-	0.00273	0.00682	0.00885	0.01988			
ZrO <sub>2</sub>	0.06572	0.00273	-	0.00409	0.00612	0.01747			
Sb <sub>2</sub> O <sub>3</sub>	0.06981	0.00682	0.00409	-	0.00203	0.01338			
ZnO	0.07184	0.00885	0.00612	0.00203	-	0.01135			
$La_2O_3$	0.08319	0.01988	0.01747	0.01338	0.01135	-			

Table 6

Evolution of attraction level (between oxide reagents) with the absolute values of  $\Delta(\mathbf{E}_{ox})$  (difference between oxide reagents electronegativity).

Absolute values of ∆Eox	Attractions level	Process 1	Process 2	Process 3	Process 4	Process 5	Process 6	Process 7	Process 8
0.08319 0.07184 0.06981 0.06572 0.06299	strong attractions between oxides reagents	(PbO; TiO <sub>2</sub> )	(PbO; La <sub>2</sub> O <sub>3</sub> )	(PbO; Sb <sub>2</sub> O <sub>3</sub> )	(PbO; La <sub>2</sub> O <sub>3</sub> )	(PbO; ZrO <sub>2</sub> )	(PbO; Sb <sub>2</sub> O <sub>3</sub> ) - (PbO;ZnO)	(PbO; Sb <sub>2</sub> O <sub>3</sub> ) - (PbO;ZnO)	(PbO; Sb <sub>2</sub> O <sub>3</sub> ) - (PbO;TiO <sub>2</sub> ) - (PbO; ZrO <sub>2</sub> ) - , (PbO; La <sub>2</sub> O <sub>3</sub> )
0.01988 0.01747 0.01338 0.01135	medium attractions between oxides reagents	(La <sub>2</sub> O <sub>3</sub> ; ZnO)		(La <sub>2</sub> O <sub>3</sub> ; ZrO <sub>2</sub> )		(La <sub>2</sub> O <sub>3</sub> ; Sb <sub>2</sub> O <sub>3</sub> )	(La <sub>2</sub> O <sub>3</sub> ; ZrO <sub>2</sub> ) - (La <sub>2</sub> O <sub>3</sub> ; TiO <sub>2</sub> )	(La <sub>2</sub> O <sub>3</sub> ; ZrO <sub>2</sub> ) - (La <sub>2</sub> O <sub>3</sub> ; TiO <sub>2</sub> )	(La <sub>2</sub> O <sub>3</sub> ; TiO <sub>2</sub> )
0.00885 0.00682 0.00612 0.00409 0.00273 0.00203	Low attractions between oxides reagents	$(ZrO_2; Sb_2O_3)$	(TiO <sub>2</sub> ;ZnO) – (ZrO <sub>2</sub> ;Sb <sub>2</sub> O <sub>3</sub> )	(TiO <sub>2</sub> ;ZnO)	(TiO <sub>2</sub> ; ZrO <sub>2</sub> ) – (ZnO;Sb <sub>2</sub> O <sub>3</sub> )	(TiO <sub>2</sub> ;ZnO)			(TiO <sub>2</sub> ;ZnO)

A difference between the values of electronegativity can cause attraction or repulsion [22] between different oxides reagents, which can influence on kinetics solid reaction. Table 5 summarizes the absolute values of difference in electronegativity  $\Delta(E_{ox})$ between oxide reagents.

Therefore, based on the absolute values obtained of  $\Delta(E_{ox})$ , attractions between oxides can be classified into three categories: strong, medium and low attractions as shown in Table 6.

According to the results in Table 5, 6 and the order of reagents (oxides) addition

to the mixture for agitation, it is remarkable that there is a transfer or attraction between the entities of some oxides and others one. Arguably, (ZrO<sub>2</sub> was transferred to PbO and TiO<sub>2</sub>), (ZrO<sub>2</sub>, TiO<sub>2</sub> to PbO and Sb<sub>2</sub>O<sub>3</sub>) and (TiO<sub>2</sub> to PbO and ZrO<sub>2</sub>) for processes 1, 3 and 5 respectively. This leads to produce a maximum amount of PZT phase products. Moreover, for the processes 2 and 4, an attraction can be seen between entities of TiO<sub>2</sub>, ZrO<sub>2</sub> oxides and those of PbO and La<sub>2</sub>O<sub>3</sub> producing PLZT phase products at high rate. It has also been noticed that the choice of the mixing order between the various reactant oxides plays an important role, because it directly affects the electronegativity difference between the oxide and consequently the attraction between them. Therefore, other oxides added to the mixture will attract the oxides that are less attracted by each other.

About the different percentage ratios obtained for (PZT / PLZT) products ( $\approx 82.5\%$  / 17.5%; 67% / 33%; 70% / 30%) in processes 6, 7 and 8 respectively, the results may be due to the existence of some oxides that have a greater electronegativity than others, which probably caused a spatial barrier preventing reaction between the reagents involved.

These results suggest that the selectivity in the solid-state reaction may be related to the electronegativity and the different attraction forces between reactants. Therefore, the knowledge of phase formation mechanism and kinetics is essential for the optimization of the synthesis processes [23 - 26].

# 6. Conclusion

The present article describes the solid state synthesis of piezoelectric ceramic material. It was found that the mixing methods have a significant impact on conversion, and make selective synthesis process. It can be concluded that the most important factors governing this selectivity are electronegativity and interaction forces. The use of (2) and (4) processes leads to produce PLZT with higher quantities. However, the other processes (1, 3, 5, 6, 7 and 8) can produce a major amount of PZT.

An optimum mass fraction of selected product can be obtained by taking into consideration the reactant physical properties such as electronegativity, ionic potential and attraction forces.

The present article suggests estimate the products distribution in the final synthesis mixture of advanced ceramics materials by the solid state synthesis process with suitable selectivity. Developing this study would improve the industrial efficiency of the synthesis method by solid route.

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