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## APPENDICES

Appendix A XRD raw data of PZT via Sol-Gel Process and Microwave Technique at 150 °C for 25 h and calcined at 900 °C for 1.5 h, using Pb, Ti and Zr Glycolate precursors

2 Theta	h	k	l	d-observe	Intensity
21.520	0	0	1	4.1259	1710
21.620	1	0	0	4.1070	1927
30.740	1	0	1	2.9062	10831
31.140	1	1	0	2.8692	1489
37.880	1	1	1	2.3732	1563
44.000	0	0	2	2.0562	2863
44.560	2	0	0	2.0317	1250
49.500	0	1	2	1.8399	962
	2	1	0		
54.620	1	1	2	1.6789	3649

Using the 001 and 100 reflection were chosen to determine the lattice parameter of tetragonal and rhombohedral structures .

1. For tetragonal structure was calculated from Eq 4.1

$$\frac{1}{d_{(hkl)}^2} = \frac{h^2 + k^2}{a^2} + \frac{l^2}{c^2} \quad (4.1)$$

1.1 (hkl) = (001),  $d_{001} = 4.1259$

$$\frac{1}{(4.1259)^2} = \frac{0^2+0^2}{a^2} + \frac{1^2}{c^2}$$

$$\frac{1}{(4.1259)^2} = \frac{1}{c^2}$$

$$c = 4.1259$$

1.2 (hkl) = (100),  $d_{100} = 4.1070$

$$\frac{1}{(4.1070)^2} = \frac{1^2+0^2}{a^2} + \frac{0^2}{c^2}$$

$$\frac{1}{(4.1070)^2} = \frac{1}{a^2}$$

$$a = 4.1070$$

Input  $a = 4.1070$  and  $c = 4.1259$  in Eq 4.1

$$\frac{1}{d_{(hkl)}^2} = \frac{h^2+k^2}{(4.1070)^2} + \frac{l^2}{(4.1259)^2} \quad (4.3)$$

The following equation (Eq 4.3) was used to determine the spacing  $d$  parameter of tetragonal structure to compare  $d$  (observed) with  $d$  (calculated)

1.3 (hkl) = (101) or (011)

$$\frac{1}{d_{(101)}^2} = \frac{1^2+0^2}{(4.1070)^2} + \frac{1^2}{(4.1259)^2}$$

$$d^2_{(101)} = 8.4710$$

$$d^2_{(101)} = 2.9106$$

1.4 (hkl) = (110)

$$\frac{1}{d^2_{(110)}} = \frac{1^2+1^2}{(4.1070)^2} + \frac{0^2}{(4.1259)^2}$$

$$\frac{1}{d^2_{(110)}} = \frac{2}{(4.1070)^2}$$

$$d^2_{(110)} = 8.4337$$

$$d_{(110)} = 2.9041$$

1.5 (hkl) = (111)

$$\frac{1}{d^2_{(111)}} = \frac{1^2+1^2}{(4.1070)^2} + \frac{1^2}{(4.1259)^2}$$

$$\frac{1}{d^2_{(111)}} = 0.1773$$

$$d^2_{(111)} = 5.6397$$

$$d_{(111)} = 2.3748$$

1.6 (hkl) = (002)

$$\frac{1}{d^2_{(002)}} = \frac{0^2+0^2}{(4.1070)^2} + \frac{2^2}{(4.1259)^2}$$

$$\frac{1}{d_{(002)}^2} = \frac{4}{(4.1259)^2}$$

$$d_{(002)}^2 = 4.2558$$

$$d_{(002)} = 2.0629$$

1.7 (hkl) = (200) or (020)

$$\frac{1}{d_{(200)}^2} = \frac{2^2+0^2}{(4.1070)^2} + \frac{0^2}{(4.1259)^2}$$

$$\frac{1}{d_{(200)}^2} = \frac{4}{(4.1070)^2}$$

$$d_{(200)}^2 = 4.1269$$

$$d_{(200)} = 2.0535$$

1.8 (hkl) = (012) or (102)

$$\frac{1}{d_{(012)}^2} = \frac{0^2+1^2}{(4.1070)^2} + \frac{2^2}{(4.1259)^2}$$

$$\frac{1}{d_{(012)}^2} = 0.2943$$

$$d^2_{(012)} = 3.3980$$

$$d_{(012)} = 1.8434$$

**1.9 (hkl) = (201)**

$$\frac{1}{d^2_{(201)}} = \frac{2^2+0^2}{(4.1070)^2} + \frac{1^2}{(4.1259)^2}$$

$$\frac{1}{d^2_{(201)}} = 0.2959$$

$$d^2_{(201)} = 3.3795$$

$$d_{(201)} = 1.8384$$

**1.10 (hkl) = (112)**

$$\frac{1}{d^2_{(112)}} = \frac{1^2+1^2}{(4.1070)^2} + \frac{2^2}{(4.1259)^2}$$

$$\frac{1}{d^2_{(112)}} = 0.3536$$

$$d^2_{(112)} = 2.8285$$

$$d_{(112)} = 1.6818$$

2. For rhombohedral structure was calculated from Eq 4.2.

$$\frac{1}{d_{(hkl)}^2} = \frac{(h^2+k^2+l^2)\sin^2\alpha + 2(hk+kl+lh)(\cos^2\alpha-\cos\alpha)}{a^2(1+2\cos^3\alpha-3\cos^2\alpha)} \quad (4.2)$$

Where  $\alpha = 89.772^\circ$  [32] and input d (observed) of (hkl) = (001) is 4.1259 in Eq 4.2.

$$\frac{1}{d_{(001)}^2} = \frac{(0^2+0^2+l^2)\sin^289.772 + 2(0+0+0)(\cos^289.772-\cos89.772)}{a^2(1+2\cos^389.772-3\cos^289.772)}$$

$$\frac{1}{(4.1259)^2} = \frac{\sin^289.772}{a^2(1+2\cos^389.772-3\cos^289.772)}$$

$$(4.1259)^2 = a^2(1.000063627)$$

$$a^2 = 17.0220$$

$$a = 4.1259$$

Input a = 4.1259 in the Eq 4.2.

$$\frac{1}{d_{(hkl)}^2} = \frac{(h^2+k^2+l^2)\sin^2\alpha + 2(hk+kl+lh)(\cos^2\alpha-\cos\alpha)}{(4.1259)^2(1+2\cos^3\alpha-3\cos^2\alpha)} \quad (4.4)$$

The following equation (Eq 4.4) was used to determine the spacing d parameter of rhombohedral structure to compare d (observed) with d (calculated)

2.1 (hkl) = (100) or (010)

$$\frac{1}{d_{(100)}^2} = \frac{(1^2+0^2+0^2)\sin^2 89.772 + 2(0+0+0)(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$\frac{1}{d_{(100)}^2} = \frac{\sin^2 89.772}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(100)} = 4.1259$$

2.2 (hkl) = (101), (011) or (110)

$$\frac{1}{d_{(110)}^2} = \frac{(1^2+1^2+0^2)\sin^2 89.772 + 2(1+0+0)(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$\frac{1}{d_{(110)}^2} = \frac{2\sin^2 89.772 + 2(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(110)}^2 = 8.5451$$

$$d_{(110)} = 2.9232$$

2.3 (hkl) = (111)

$$\frac{1}{d_{(111)}^2} = \frac{(1^2+1^2+1^2)\sin^2 89.772 + 2(1+1+1)(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$\frac{1}{d_{(111)}^2} = \frac{3\sin^2 89.772 + 6(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(111)}^2 = 5.7198$$

$$d_{(111)} = 2.3916$$

2.4 (hkl) = (200), (020) or (002)

$$\frac{1}{d_{(200)}^2} = \frac{(2^2+0^2+0^2)\sin^2 89.772 + 2(0+0+0)(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$\frac{1}{d_{(200)}^2} = \frac{4\sin^2 89.772}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(200)}^2 = 4.2556$$

$$d_{(200)} = 2.0629$$

2.5 (hkl) = (201), (102) or (012)

$$\frac{1}{d_{(201)}^2} = \frac{(2^2+0^2+1^2)\sin^2 89.772 + 2(0+0+2)(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$\frac{1}{d_{(201)}^2} = \frac{5\sin^2 89.772 + 4(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(201)}^2 = 3.4147$$

$$d_{(201)} = 1.8479$$

2.6 (hkl) = (112)

$$\frac{1}{d_{(112)}^2} = \frac{(1^2+1^2+2^2)\sin^2 89.772 + 2(1+2+2)(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$\frac{1}{d_{(112)}^2} = \frac{6\sin^2 89.772 + 10(\cos^2 89.772 - \cos 89.772)}{(4.1259)^2(1 + 2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(112)}^2 = 2.8443$$

$$d_{(112)} = 1.6865$$

Appendix B XRD raw data of PZT via Sol-Gel Process and calcined at 900 °C for 1.5 h, using Ti, Zr Glycolate and Lead Acetate Trihydrate precursors

2 Theta	h	k	l	d-observe	Intensity
21.780	0	0	1	4.0772	2132
30.940	1	1	0	2.8878	8473
38.180	1	1	1	2.3552	1459
44.260	2	0	0	2.0448	2688
49.920	2	0	1	1.8254	970
54.940	1	1	2	1.6699	2964

Using the 001 reflection were chosen to determine the lattice parameter of rhombohedral structures from Eq 4.2.

$$\frac{1}{d_{(hkl)}^2} = \frac{(h^2+k^2+l^2)\sin^2\alpha + 2(hk+kl+lh)(\cos^2\alpha-\cos\alpha)}{a^2(1+2\cos^3\alpha-3\cos^2\alpha)} \quad (4.2)$$

Where  $\alpha = 89.772^\circ$  [32] and input d (observed) of (hkl) = (001) is 4.0772 in Eq 4.2.

$$\frac{1}{d_{(001)}^2} = \frac{(0^2+0^2+l^2)\sin^289.772 + 2(0+0+0)(\cos^289.772-\cos89.772)}{a^2(1+2\cos^389.772-3\cos^289.772)}$$

$$\frac{1}{(4.0772)^2} = \frac{\sin^289.772}{a^2(1+2\cos^389.772-3\cos^289.772)}$$

$$(4.0772)^2 = a^2(1.000063627)$$

$$a = 4.0772$$

Input  $a = 4.0772$  in Eq 4.2

$$\frac{1}{d_{(hkl)}^2} = \frac{(h^2+k^2+l^2)\sin^2\alpha + 2(hk+kl+lh)(\cos^2\alpha-\cos\alpha)}{(4.0772)^2(1+2\cos^3\alpha-3\cos^2\alpha)} \quad (4.5)$$

3.1 (hkl) = (100), (010) or (001)

$$\begin{aligned} \frac{1}{d_{(100)}^2} &= \frac{(1^2+0^2+0^2)\sin^289.772 + 2(0+0+0)(\cos^289.772-\cos89.772)}{(4.0772)^2(1+2\cos^389.772-3\cos^289.772)} \\ \frac{1}{d_{(100)}^2} &= \frac{\sin^289.772}{(4.0772)^2(1+2\cos^389.772-3\cos^289.772)} \\ d_{(100)} &= 4.0771 \end{aligned}$$

3.2 (hkl) = (110), (101) or (011)

$$\begin{aligned} \frac{1}{d_{(110)}^2} &= \frac{(1^2+1^2+0^2)\sin^289.772 + 2(1+0+0)(\cos^289.772-\cos89.772)}{(4.0772)^2(1+2\cos^389.772-3\cos^289.772)} \\ \frac{1}{d_{(110)}^2} &= \frac{2\sin^289.772 + 2(\cos^289.772-\cos89.772)}{(4.0772)^2(1+2\cos^389.772-3\cos^289.772)} \\ d_{(110)}^2 &= 8.3440 \\ d_{(110)} &= 2.8886 \end{aligned}$$

3.3 (hkl) = (111)

$$\frac{1}{d_{(111)}^2} = \frac{(1^2+1^2+1^2)\sin^289.772 + 2(1+1+1)(\cos^289.772-\cos89.772)}{(4.0772)^2(1+2\cos^389.772-3\cos^289.772)}$$

$$\frac{1}{d_{(111)}^2} = \frac{3\sin^2 89.772 + 6(\cos^2 89.772 - \cos 89.772)}{(4.0772)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(111)}^2 = 5.5852$$

$$d_{(111)} = 2.3633$$

3.4 (hkl) = (200), (020) or (002)

$$\frac{1}{d_{(200)}^2} = \frac{(2^2 + 0^2 + 0^2)\sin^2 89.772 + 2(2+0+0)(\cos^2 89.772 - \cos 89.772)}{(4.0772)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$\frac{1}{d_{(200)}^2} = \frac{4\sin^2 89.772 + 4(\cos^2 89.772 - \cos 89.772)}{(4.0772)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(200)}^2 = 4.1812$$

$$d_{(200)} = 2.0448$$

3.5 (hkl) = (201), (012) or (102)

$$\frac{1}{d_{(201)}^2} = \frac{(2^2 + 0^2 + 1^2)\sin^2 89.772 + 2(0+0+2)(\cos^2 89.772 - \cos 89.772)}{(4.0772)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$\frac{1}{d_{(201)}^2} = \frac{5\sin^2 89.772 + 4(\cos^2 89.772 - \cos 89.772)}{(4.0772)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(201)}^2 = 3.3321$$

$$d_{(201)} = 1.8254$$

3.6 (hkl) = (112)

$$\frac{1}{d_{(201)}^2} = \frac{(1^2+1^2+2^2)\sin^2 89.772 + 2(1+2+2)(\cos^2 89.772 - \cos 89.772)}{(4.0772)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$\frac{1}{d_{(201)}^2} = \frac{6\sin^2 89.772 + 10(\cos^2 89.772 - \cos 89.772)}{(4.0772)^2(1+2\cos^3 89.772 - 3\cos^2 89.772)}$$

$$d_{(201)}^2 = 2.7889$$

$$d_{(201)} = 1.6700$$

All of these spacing d parameter calculation were shown in Table 4.10 and 4.14.

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