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From core-shell $Ba_{0.4}Sr_{0.6}TiO_3$@SiO_2 particles to dense ceramics with high energy storage performance by spark plasma sintering

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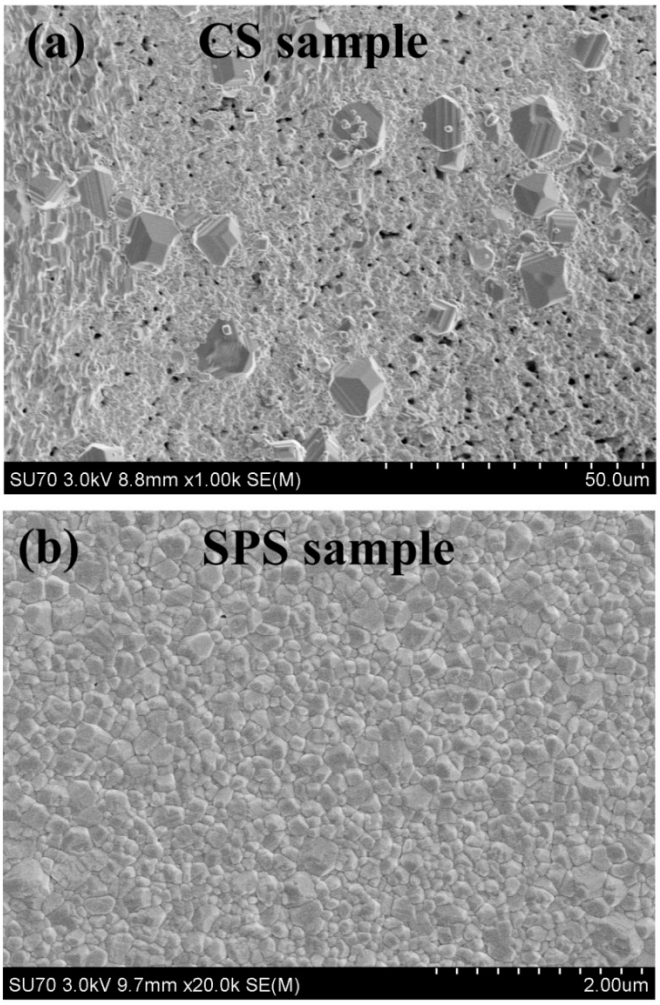

Fig. S1. SEM photos of BST ceramics prepared by (a) conventional sintering and (b) spark plasma sintering.

Table S1(a) Current research in Ba-based ceramics for energy storage application.

Composition	Breakdown strength (kV/cm)	Discharge energy density (J/cm^3)	year	Ref.
BST@SiO_2-8	**400**	**1.60**	**in this work**	
$BaZr_{0.1}Ti_{0.9}O_3$	30	0.5	2017	[1]
$Ba_{0.4}Sr_{0.6}TiO_3$+Al_2O_3-SiO_2	198.8	0.564	2017	[2]
$BaTiO_3$+La_2O_3+SiO_2	136	0.54	2017	[3]
$(Ba_{0.85}Ca_{0.15})(Ti_{0.9}Zr_{0.1})O_3$	30	0.52	2016	[4]
$Ba_{0.9995}La_{0.0005}TiO_3$	300	0.564	2016	[5]
$Ba_{0.95}Ca_{0.05}Zr_{0.3}Ti_{0.7}O_3$	160	0.59	2016	[6]
$BaTiO_3$-$BiScO_3$	120	0.68	2016	[7]
$Ba_{0.4}Sr_{0.6}TiO_3$-0.5wt%SiO_2	134	0.86	2016	[8]
$BaTiO_3$+2.0wt%SiO_2	200	1.2	2015	[9]
$BaTiO_3$+Al_2O_3-SiO_2	190	0.725	2015	[10]
$0.88BaTiO_3$-$0.12Bi(Mg_{1/2}Ti_{1/2})O_3$	250	1.81	2015	[11]
$Ba_{0.4}Sr_{0.6}TiO_3$	197	1.3	2015	[12]
$Ba_{0.4}Sr_{0.6}TiO_3$	90	0.32	2015	[13]
$(0.65BiFeO_3$-$0.35BaTiO_3)$-3mol%Nb_2O_5	90	0.71	2014	[14]
$BaTiO_3$-$SrTiO_3$	47	0.22	2014	[15]
$Ba_{0.4}Sr_{0.6}TiO_3$	243	1.28	2014	[16]
$0.9BaTiO_3$-$0.1Bi(Mg_{2/3}Nb_{1/3})O_3$	145	1.13	2014	[17]
$0.85Ba(Zr_{0.2}Ti_{0.8})O_3$-$0.15(Ba_{0.7}Ca_{0.3})TiO_3$	170	0.94	2013	[18]
$Ba_{0.4}Sr_{0.6}TiO_3$+2.0wt% glass	84	0.44	2012	[19]
$BaTiO_3$+7.0wt% glass	90	0.32	2012	[20]
$BaTiO_3$-$SrTiO_3$	157	1.16	2011	[21]
$Ba_{0.4}Sr_{0.6}TiO_3$-30wt% MgO	331	1.14	2010	[22]
$Ba_{0.4}Sr_{0.6}TiO_3$-5vol%$(BaO$-SiO_2-$B_2O_3)$	200	0.89	2009	[23]

Table S1(b) Current research in Bi-based ceramics for energy storage application.

Composition	Breakdown strength (kV/cm)	Discharge energy density (J/cm^3)	year	Ref.
$0.95(0.93Bi_{0.5}Na_{0.5}TiO_3\text{-}0.07BaTiO_3)\text{-}0.05KNbO_3$	168	1.72	2017	[24]
$0.4(Bi_{0.5}Na_{0.5}TiO_3)\text{-}0.225BaTiO_3\text{-}0.375BiFeO_3$	80	1.4	2017	[25]
$Bi_{0.5}Na_{0.5}TiO_3\text{-}SrTiO_3\text{-}NaNbO_3$	70	0.73	2017	[26]
$Bi_{0.5}Na_{0.5}TiO_3\text{-}Ba_{0.85}Ca_{0.15}Ti_{0.85}Zr_{0.1}Sn_{0.05}O_3$+ MgO	189.7	1.62	2017	[27]
$0.80Bi_{0.5}Na_{0.5}TiO_3\text{-}0.20Bi_{0.5}K_{0.5}TiO_3+BaZrO_3$	70	0.73	2016	[28]
$[(Bi_{0.5}Na_{0.5})_{0.93}Ba_{0.07}]_{1-x}La_xTi_{1-y}Zr_yO_3$	100	1.21	2016	[29]
$(1\text{-}x)(Na_{0.5}Bi_{0.5})TiO_3\text{-}xSrTiO_3$	65	0.65	2016	[30]
$0.92(Na_{1/2}Bi_{1/2})TiO_3\text{-}0.08BaTiO_3+Bi(Mg_{1/2}Ti_{1/2})O_3$	135	2.0	2016	[31]
$Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3$	105	1.41	2016	[32]
$0.55Bi_{0.5}Na_{0.5}TiO_3\text{-}0.45Ba_{0.85}Ca_{0.15}Ti_{0.9-x}Zr_{0.1}Sn_xO_3$	130.2	1.21	2016	[33]
$0.96[Bi_{0.5}(Na_{0.84}K_{0.16})_{0.5}Ti_{1-x}Ta_xO_3]\text{-}0.04SrTiO_3$	50	0.65	2016	[34]
$0.94Bi_{0.5}Na_{0.5}TiO_3\text{-}0.06BaTiO_3+CaZrO_3$	70	0.7	2016	[35]
$Bi_{0.487}Na_{0.427}K_{0.06}Ba_{0.026}TiO_3\text{-}xCeO_2$	75	0.94	2016	[36]
$Bi_{0.5}Na_{0.5}TiO_3\text{-}Ba_{0.85}Ca_{0.15}Ti_{0.9}Zr_{0.1}O_3+MgO$	156.7	1.04	2016	[37]
$(Bi_{0.5}Na_{0.5})_{0.94}Ba_{0.06}Ti_{1-x}(Al_{0.5}Nb_{0.5})_xO_3$	70	0.7	2016	[38]
$0.85[Bi_{0.5}Na_{0.5}TiO_3\text{-}BaTiO_3]\text{-}0.15Na_{0.73}Bi_{0.09}NbO_3$	142	1.4	2016	[39]
$(Bi_{0.5}Na_{0.5})TiO_3\text{-}(Bi_{0.5}K_{0.5})TiO_3]\text{-}(K_{0.5}Na_{0.5})NbO_3$	100	1.2	2015	[40]
$Bi_{0.5}Na_{0.5}TiO_3+NaNbO_3$	70	0.71	2015	[41]
$0.93Bi_{0.5}Na_{0.5}TiO_3\text{-}0.06BaTiO_3\text{-}0.01K_{0.5}Na_{0.5}NbO_3$	50	0.598	2015	[42]
$0.89Bi_{0.5}Na_{0.5}TiO_3\text{-}0.06BaTiO_3\text{-}0.05K_{0.5}Na_{0.5}NbO_3$	99	0.9	2014	[43]
$(1\text{-}x)Bi_{0.47}Na_{0.47}Ba_{0.06}TiO_3\text{-}xKNbO_3$	100	0.89	2014	[44]
$[(Bi_{1/2}Na_{1/2})_{0.94}Ba_{0.06}]La_{(1-x)}Zr_xTiO_3$	83.4	1.58	2014	[45]
$0.89Bi_{0.5}Na_{0.5}TiO_3\text{-}0.06BaTiO_3\text{-}0.05K_{0.5}Na_{0.5}NbO_3$	56	0.59	2011	[46]

Table S1(c) Current research in Pb-based ceramics for energy storage application.

Composition	Breakdown strength (kV/cm)	Discharge energy density (J/cm^3)	year	Ref.
$Pb_{0.97}La_{0.02}(Zr_xSn_{0.925-x}Ti_{0.075})O_3$	89.1	2.35	2017	[47]
$(Pb_{0.94-x}Ba_xLa_{0.04})[(Zr_{0.7}Sn_{0.3})_{0.88}Ti_{0.12}]$	59	0.9	2017	[48]
$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$	70	0.47	2016	[49]
$Pb_{0.90}La_{0.04}Ba_{0.04}[(Zr_{0.7}Sn0.3)0.88Ti_{0.12}]O_3$	66.67	0.74	2016	[50]
$(Pb_{0.97}La_{0.02})$ $(Zr_{0.5}Sn_{0.5-x}Ti_x)O_3$	158	4.2	2016	[51]
$Pb_{0.99}Nb_{0.02}[(Zr_{0.60}Sn_{0.40})_{0.95}Ti0.05]_{0.98}O_3$	62	0.73	2016	[52]
$(Pb_{0.87}Ba_{0.1}La_{0.02})(Zr_{0.65}Sn_{0.3}Ti_{0.05})O_3$ +0.75mol%Y	130	2.75	2016	[53]
$La_xPb_{1-3x/2}(Lu_{1/2}Nb_{1/2})O_3$	681	3.85	2015	[54]
$(Pb_{0.87}Ba_{0.1}La_{0.02})(Zr_{0.68}Sn_{0.24}Ti_{0.08})O_3$	180	3.2	2015	[55]
$(Pb_{0.858}Ba_{0.1}La_{0.02}Y_{0.008})(Zr_{0.65}Sn_{0.3}Ti_{0.05})O_3$ -$(Pb_{0.97}La_{0.02})(Zr_{0.9}Sn_{0.05}Ti_{0.05})O_3$	306	6.4	2015	[56]
$(Pb_{0.858}Ba_{0.1}La_{0.02}Y_{0.008})(Zr_{0.65}Sn_{0.3}Ti_{0.05})O_3$ -$(Pb_{0.97}La_{0.02})(Zr_{0.9}Sn_{0.05}Ti_{0.05})O_3$	200	4.65	2015	[57]
$(Pb_{0.925-x}La_{0.05}Ba_x)(Zr_{0.52}Sn_{0.39}Ti_{0.09})O_3$	80	0.7	2015	[58]
$Pb_{0.97}La_{0.02}(Zr_{0.58}Sn_{0.335}Ti_{0.085})O_3$	86	1.37	2015	[59]

Reference

1. Y. Zhang, Y. Li, H. Zhu, Z. Fu, and Q. Zhang. *J. Mater. Sci. Mater. Electron*., 2017, **28**,514.
2. B. Liu, X. Wang, R. Zhang, and L. Li. *J. Alloy. Compd*., 2017, **691**, 619.
3. R. Ma, B. Cui, M. Shangguan, S. Wang, Y. Wang, Z. Chang, and Y. Wang. *J. Alloy. Compd*., 2016, **690**, 438.
4. Z. Wang, J. Wang, X. Chao, L. Wei, B. Yang, D. Wang, and Z. Yang. J. Mater. Sci. Mater. Electron., 2016, 27, 5047.

5. V. S. Puli, D. K. Pradhan, S. Adireddy, M. Kothakonda, R. S. Katiyar, and D. B. Chrisey. *J. Alloy. Compd*., 2016, **688**, 721.
6. D. Zhan, Q. Xu, D. P. Huang, H. X. Liu, W. Chen, and F. Zhang. *J. Alloy. Compd*., 2016, **682**, 594.
7. L. Wu, X. Wang and L. Li. *J. Alloy. Compd*., 2016, **688**, 113.
8. C. Diao, H. Liu, H. Hao, M. Cao, and Z. Yao. *Ceram. Int*., 2016, **42**, 12639.
9. Y. Zhang, M. Cao, Z. Yao, Z. Wang, Z. Song, A. Ullah, H. Hao, and H. Liu. *Mater. Res. Bull*., 2015, **67**, 70.
10. B. Liu, X. Wang, Q. Zhao, and L. Li. *J. Am. Ceram. Soc*., 2015, **98**, 2641.
11. Q. Hu, L. Jin, T. Wang, C. Li, Z. Xing, and X. Wei. *J. Alloy. Compd*., 2015, **640**, 416.
12. Z. Song, H. Liu, H. Hao, S. Zhang, M. Cao, Z. Yao, Z. Wang, W. Hu, Y. Shi, and B. Hu. *IEEE. T. Ultrason. Ferr*., 2015, **62**, 609.
13. Y. Wang, Z. Y. Shen, Y. M. Li, Z. M. Wang, W. Q. Luo, and Y. Hong. *Ceram. Int*., 2015, **41**, 8252.
14. T. Wang, L. Jin, Y. Tian, L. Shu, Q. Hu, and X. Wei. *Mater. Lett*., 2014, **137**, 79.
15. L. Wu, X. Wang, H. Gong, Y. Hao, Z. Shen, and L. Li. *J. Mater. Chem. C*., 2015, **3**, 750.
16. Z. Song, H. Liu, S. Zhang, Z. Wang, Y. Shi, H. Hao, M. Cao, Z. Yao, and Z. Yu. *J. Eur. Ceram. Soc*., 2014, **34**, 1209.
17. T. Wang, L. Jin, C. Li, Q. Hu, and X. Wei. *J. Am. Ceram. Soc*., 2015, **98**, 559.
18. V. S. Puli, D. K. Pradhan, D. B. Chrisey, M. Tomozawa, G. L. Sharma, J. F. Scott, and R. S. Katiyar. *J. Mater. Sci*., 2013, **48**, 2151.
19. K. Chen, Y. Pu, N. Xu, and X. Luo. *J. Mater. Sci. Mater. Electron*., 2012, **23**, 1599.
20. X. Wang, Y. Zhang, X. Song, Z. Yuan, T. Ma, Q. Zhang, C. Deng, and T. Liang. *J. Eur. Ceram. Soc*., 2012, **32**, 559.
21. C. Sheng and F. Dogan. *Mater. Lett*., 2011, **65**, 978.
22. Q. Zhang, L. Wang, J. Luo, Q. Tang, and D. Jun. *Int. J. Appl. Ceram. Tec*., 2010, **7**, E124.
23. Q. Zhang, L. Wang, J. Luo, Q. Tang, and J. Du. *J. Am. Ceram. Soc*., 2010, **92**, 1871.
24. Q. Xu, J. Xie, Z. He, L. Zhang, M. Cao, X. Huang, M. T. Lanagan, H. Hao, Z. Yao, and H. Liu. *J. Eur. Ceram. Soc*., 2017, **37**, 99.
25. A. Mishra, B. Majumdar and R. Ranjan. *J. Eur. Ceram. Soc*., 2017, **37**, 2379.
26. L. F, Z. J, B. Shen, X. Liu, K. Yang, Y. Zhang, P. Li, B. Liu, and H. Zeng. *J. Appl. Phys*., 2017, **5**, 54103.
27. Y. Pu, M. Yao, L. Zhang, and M. Chen. *J. Alloy. Compd*., 2017,**702** ,171 .
28. Q. Li, J. Wang, Z. Liu, G. Dong, and H. Fan. *J. Mater. Sci*., 2016, **51**, 1153.
29. X. Lu, J. Xu, L. Yang, C. Zhou, Y. Y. Zhao, C. Yuan, Q. Li, G. Chen, and H. Wang. *Journal of Materiomics*, 2016, **2**, 87.
30. W. P. Cao, W. L. Li, X. F. Dai, T. D. Zhang, J. Sheng, Y. F. Hou, and W. D. Fei. *J. Eur. Ceram. Soc*., 2015, **36**, 593.
31. P. Chen and B. Chu. *J. Eur. Ceram. Soc*., 2016, **36**, 81.

32. Y. Zhao, J. Xu, L. Yang, C. Zhou, X. Lu, C. Yuan, Q. Li, G. Chen, and H. Wang. *J. Alloy. Compd*., 2016, **666**, 209.
33. Y. Pu, M. Yao, L. Zhang, and P. Jing. *J. Alloy. Compd*., 2016, **687**, 689.
34. R. A. Malik, A. Hussain, A. Maqbool, A. Zaman, T. K. Song, W. J. Kim, and M. H. Kim. Giant Strain, *J. Alloy. Compd*., 2016, **682**, 302.
35. Q. Li, J. Wang, Y. Ma, L. Ma, G. Dong, and H. Fan. *J. Alloy. Compd*., 2016, **663**, 701.
36. G. Liu, H. Fan, G. Dong, J. Shi, and Q. Chang. *J. Alloy. Compd*., 2016, **664**, 632.
37. M. Yao, Y. Pu, H. Zheng, L. Zhang, M. Chen, and Y. Cui. *Ceram. Int*., 2016, **42**, 8974.
38. Y. Zhao, J. Xu, C. Zhou, C. Yuan, Q. Li, G. Chen, H. Wang, and L. Yang. *Ceram. Int*., 2016, **42**,2221.
39. Q. Xu, M. T. Lanagan, X. Huang, J. Xie, L. Zhang, H. Hao, and H. Liu. *Ceram. Int*., 2016, **42**, 9728.
40. J. Hao, Z. Xu, R. Chu, W. Li, J. Du, and F. Peng. *Solid State Commun*., 2015, **204**, 19.
41. Q. Xu, T. Li, H. Hao, S. Zhang, Z. Wang, M. Cao, Z. Yao, and H. Liu. *J. Eur. Ceram. Soc*., 2015, **35**, 545.
42. M. Chandrasekhar and P. Kumar. *Ceram. Int*., 2015, **41**, 5574.
43. J. Ding, Y. Liu, Y. Lu, H. Qian, H. Gao, H. Chen, and C. Ma. *Mater. Lett*., 2014, **114**, 107.
44. B. Wang, L. Luo, X. Jiang, W. Li, and H. Chen. *J. Alloy. Compd*., 2014, **585**, 14.
45. Y. Wang, Z. Lv, H. Xie, and J. Cao. *Ceram. Int*., 2014, **40**, 4323.
46. F. Gao, X. Dong, C. Mao, W. Liu, H. Zhang, L. Yang, F. Cao, and G. Wang. *J. Am. Ceram. Soc*., 2011, **94**, 4382.
47. Z. Liu, Y. Bai, X. Chen, X. Dong, H. Nie, F. Cao, and G. Wang. *J. Alloy. Compd*., 2017, **691**, 721.
48. R. Xu, Q. Zhu, J. Tian, Y. Feng, and Z. Xu. *Ceram. Int*., 2016, **43**,2481.
49. T. F. Zhang, X. G. Tang, Q. X. Liu, Y. P. Jiang, X. X. Huang, and Q. F. Zhou. *J. Phys. D. Appl. Phys*., 2016, **49**, 95302.
50. R. Xu, Z. Xu, Y. Feng, H. He, J. Tian, and D. Huang. *J. Am. Ceram. Soc*., 2016, **99**, 2984.
51. X. Wang, J. Shen, T. Yang, Y. Dong, and Y. Liu. *J. Alloy. Compd*., 2016, **655**, 309.
52. X. Chen, Z. Liu, C. Xu, F. Cao, G. Wang, and X. Dong. *AIP Adv*., 2016, **6**, 593.
53. L. Zhang, S. Jiang, Y. Zeng, M. Fu, K. Han, Q. Li, Q. Wang, and G. Zhang. *Ceram. Int*., 2014, **40**, 5455.
54. X. Yang, Y. Liu, C. He, H. Tailor, and X. Long. *J. Eur. Ceram. Soc*., 2015, **35**, 4173.
55. G. Zhang, D. Zhu, X. Zhang, L. Zhang, J. Yi, B. Xie, Y. Zeng, Q. Li, Q. Wang, and S. Jiang. *J. Am. Ceram. Soc*., 2015, **98**, 1175.
56. L. Zhang, S. Jiang, B. Fan, and G. Zhang. *J. Alloy. Compd*., 2015, **622**, 162.
57. L. Zhang, S. Jiang, B. Fan, and G. Zhang. *J. Alloy. Compd*., 2015, **41**, 1139.
58. Q. Zhang, X. Liu, Y. Zhang, X. Song, J. Zhu, I. Baturin, and J. Chen. *Ceram. Int*.,

2015, **41**, 3030.
59. Z. Liu, X. Chen, W. Peng, C. Xu, X. Dong, F. Cao, and G. Wang. *Appl. Phys. Lett*., 2015, **106**, 15.