# Electronic Supplementary Information

## Enhanced energy storage properties in lead-free $BaTiO_3$@$Na_{0.5}K_{0.5}NbO_3$ ceramics with nanodomains via a core-shell structure ultrafine-grained design

Quan Jin[1], Lili Zhao[2], Bin Cui[1,*], Jia Wang[1], Run Zhang[1], Xiaoting Zhang[1]

1. Key Laboratory of Synthetic and Natural Functional Molecule (Ministry of Education), Shaanxi Key Laboratory of Physico-Inorganic Chemistry, College of Chemistry & Materials Science, Northwest University, Xi'an, Shaanxi 710127, China

2. School of Information Science and Technology, Northwest University, Xi'an, Shaanxi 710127, China

* Corresponding author. Tel./fax: +86 29 8153-5030;

*E-mail address:* cuibin@nwu.edu.cn.

**CONTENTS** **Page Number**

## 1. SEM, TEM micrographs and particle size distribution of the BT particles

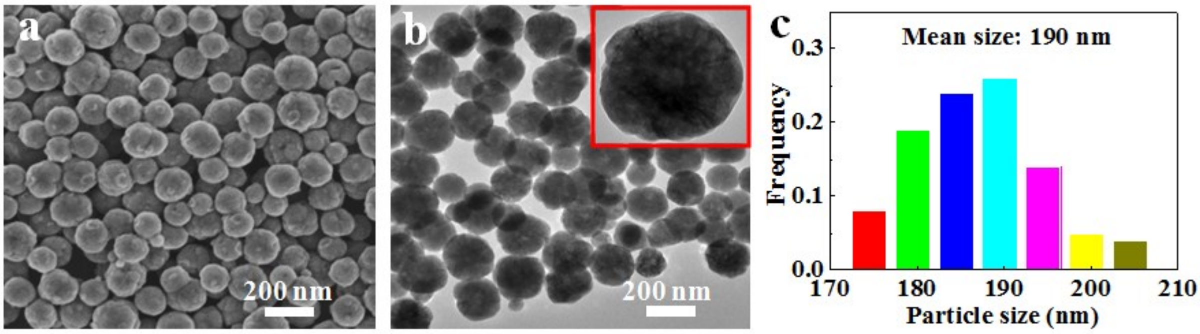


**Figure S1.** (a) SEM, (b) TEM and particle size distribution of the BT particles, respectively.

**2. EDS elemental mapping image of a single BT@KNN particle coated with 8 wt.% KNN**

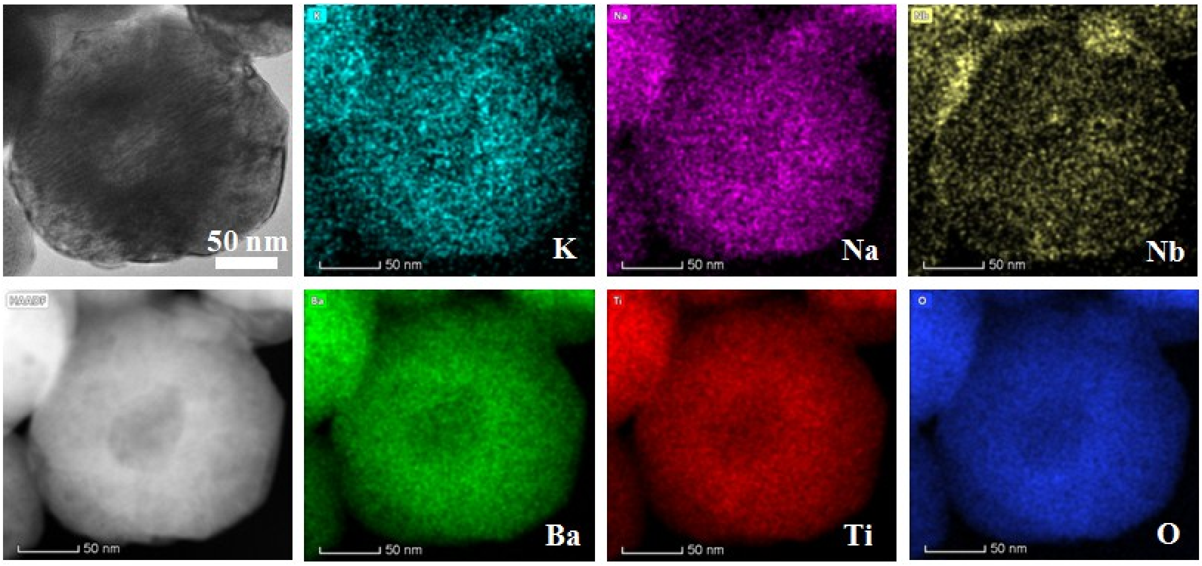


**Figure S2.** TEM, STEM images and EDS mappings of a single BT@KNN particle coated with 8 wt.% KNN.

### 3. Integrated area of Raman peaks at 718 $cm^{-1}$ of BT@KNN ceramics

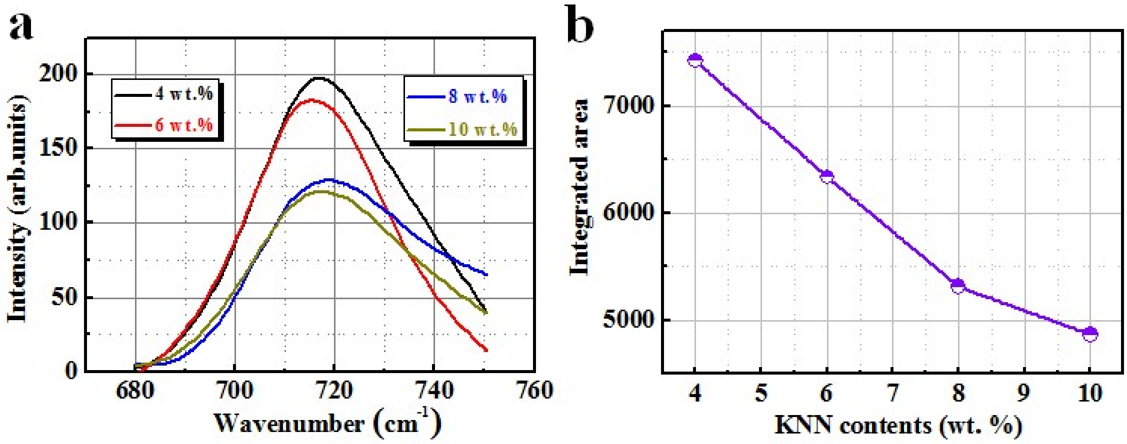


**Figure S3.** (a) Raman spectra of the BT@KNN ceramics with various amounts of KNN at the wave-number of 680 $cm^{-1}$-750 $cm^{-1}$. (b) The values of integrated area of Raman peaks at 718 $cm^{-1}$ of the BT@KNN ceramics.

**4. EDS elemental mapping image of BT@KNN ceramics with 8 wt.% KNN**

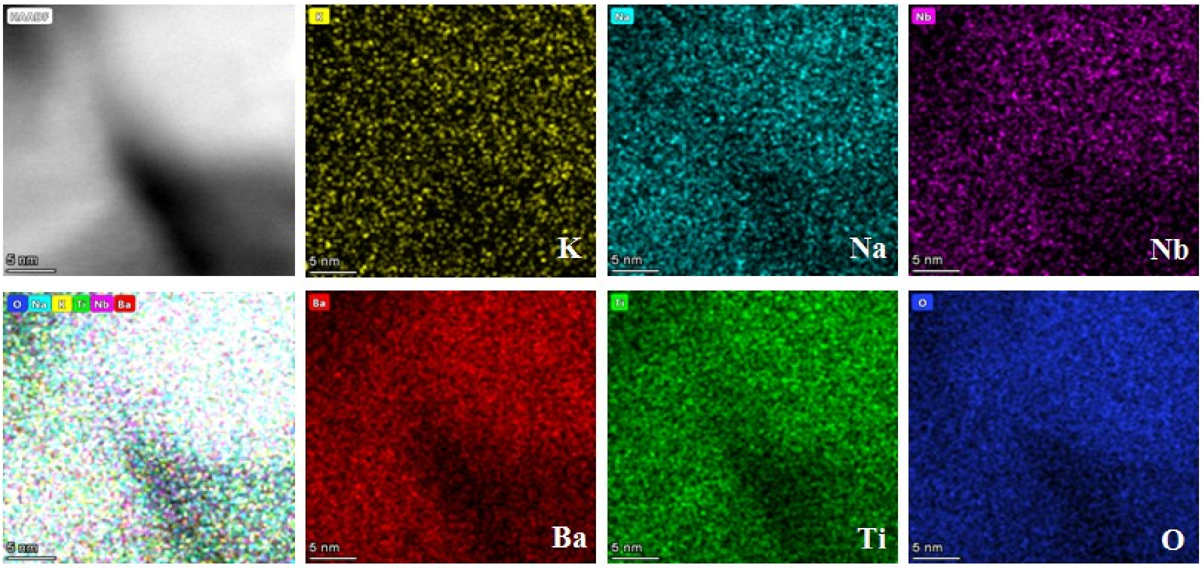


**Figure S4.** STEM images and EDS mappings of a single BT@KNN ceramics coated with 8 wt.% KNN.

**5. EDS elemental mapping image of BT@KNN ceramics with 8 wt.% KNN**

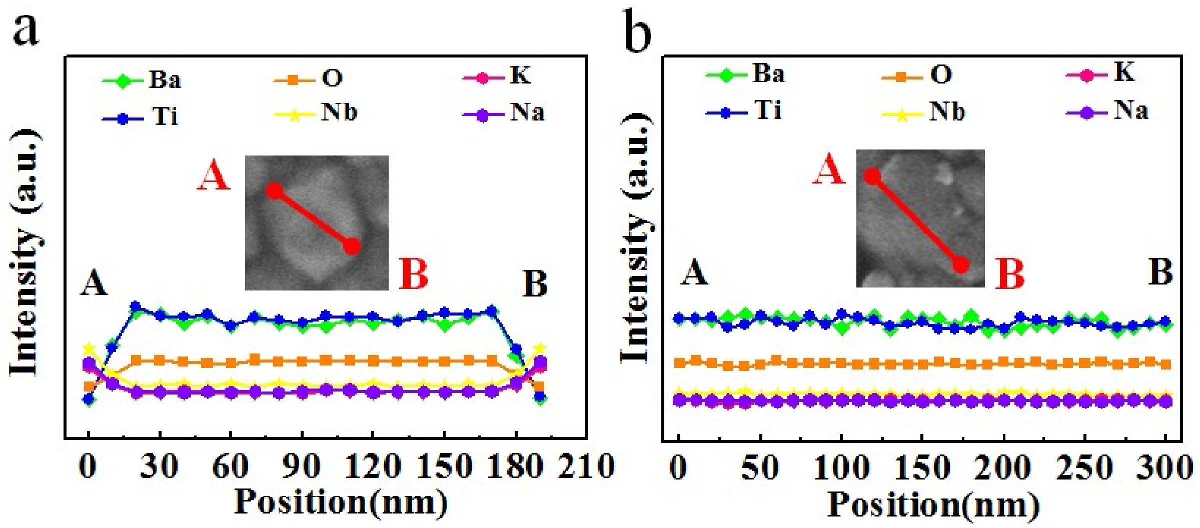


**Figure S5.** The EDS analysis results for (a) the BT@KNN ceramics with 8 wt.% KNN, and (b) BT@KNN ceramics with 10 wt.% KNN. The EDS spectra are shown along the line AB in the inset photograph.

## 6. The $\gamma$ and $T_B$ values of the BT@KNN ceramics with various contents of KNN

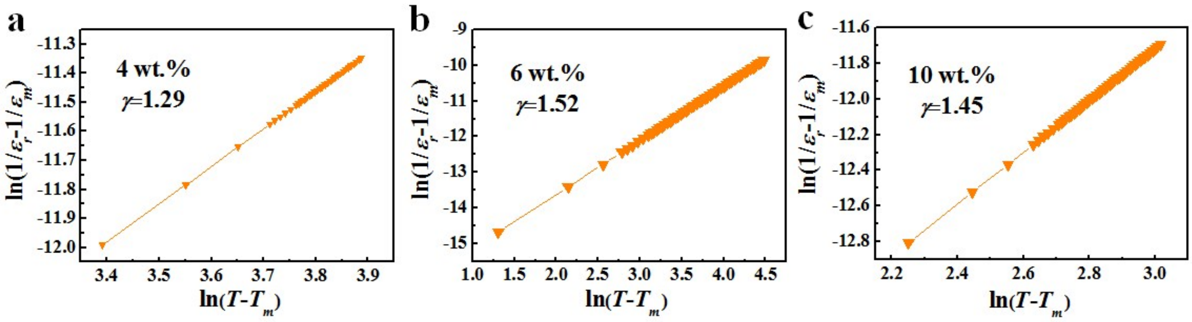


**Figure S6.** $\ln(T-T_m)$ as a function of $\ln(1/\varepsilon_r-1/\varepsilon_m)$ for BT@KNN ceramics with content of KNN (a) 4 wt.%, (b) 6 wt.% and (c) 10 wt.%.

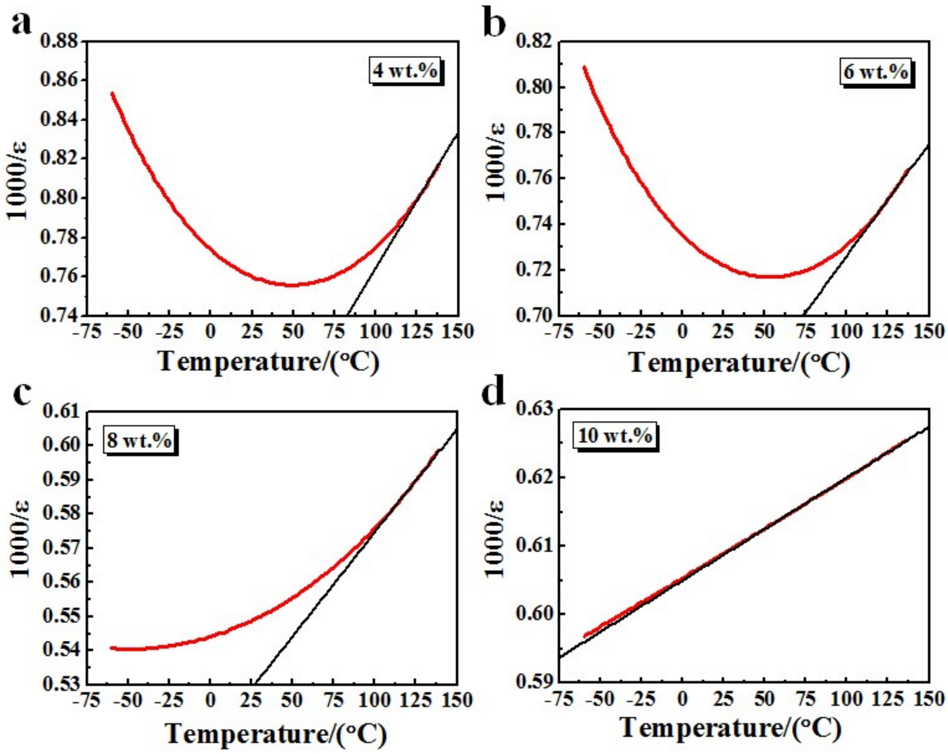


**Figure S7.** Temperature dependence of the reciprocal of dielectric permittivity to determine the $T_B$ values of BT@KNN ceramics with various contents of KNN, from which the dielectric responses start to follow Curie-Weiss law.

**7. The Weibull distributions of the BDS of BT@KNN ceramics**

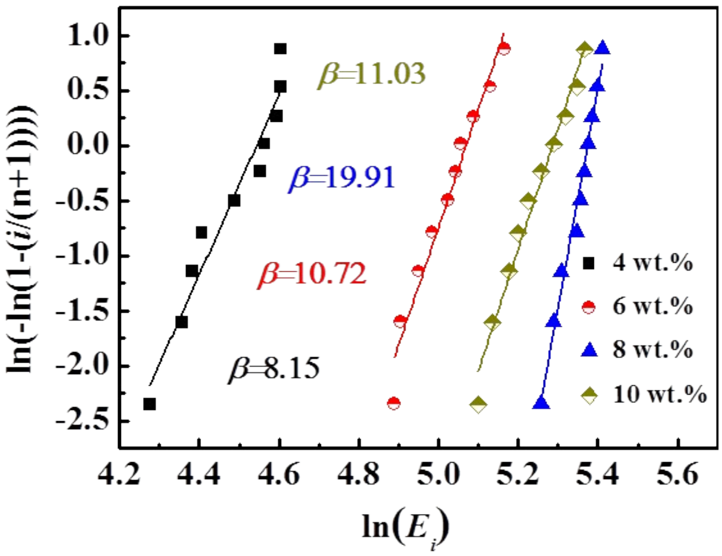


**Figure S8.** Weibull distributions of the BDS of BT@KNN ceramics

## 8. HR-TEM image of the core in BT@KNN ceramics with 8 wt.% KNN

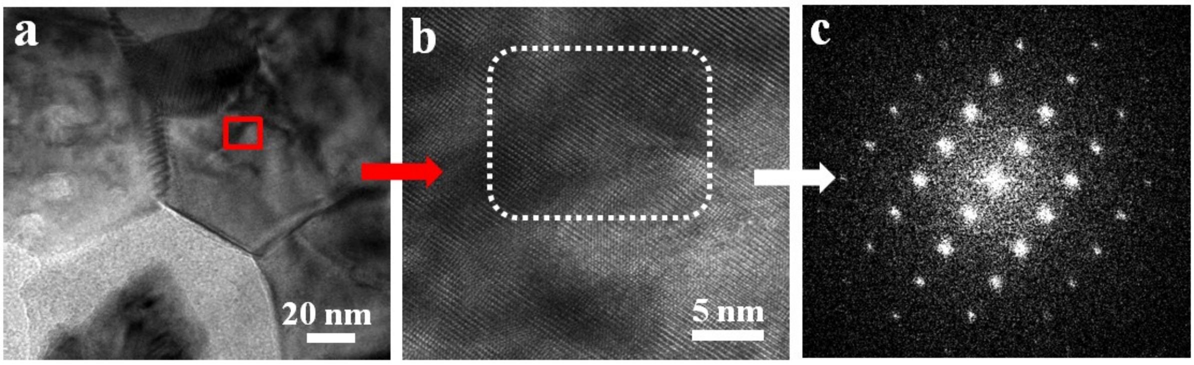


**Figure S9.** (a) TEM image of the BT@KNN ceramics coated with 8 wt.% KNN. (b) HR-TEM image of the red box area (corresponding to the core) in BT@KNN ceramics. (c) FFT patterns transformed from the HR-TEM image.

**9. The values of densities and mean grain size for the BT@KNN ceramics with various contents of KNN**

**Table S1.** The values of densities and mean grain size for the ceramics with various amounts of KNN

| Samples | Vaues of densities ($g/cm^3$) | Mean grain size (nm) |
|---|---|---|
| 4 wt.% | 5.367 | 191 |
| 6 wt.% | 5.462 | 192 |
| 8 wt.% | 5.676 | 194 |
| 10 wt.% | 5.594 | 286 |

## 10. The energy storage performance of lead-free bulk ceramics

**Table S2.** The energy storage performance of lead-free bulk ceramics

| Materials | $W_{rec}$ (J/cm$^3$) | $\eta$ (%) | Ref. |
|---|---|---|---|
| $Ba_{0.4}Sr_{0.6}TiO_3$@$SiO_2$ | 1.60 | 90.9 | 1 |
| $BaTiO_3$-based | 0.41 | 81 | 2 |
| $BaTiO_3$-$Al_2O_3$-$SiO_2$-ZnO | 0.83 | 81.6 | 3 |
| $BaTiO_3$@$SrTiO_3$ | 0.22 | 90 | 4 |
| $Ba_{0.4}Sr_{0.6}TiO_3$-ZnO-$Li_2O$ | 0.56 | 87.7 | 5 |
| $Ba_{0.970}Ce_{0.030}Ti_{0.99}Mn_{0.01}O_3$ | 0.11 | 88 | 6 |
| $BaTiO_3$@$Al_2O_3$ | 0.51 | 79.6 | 7 |
| $BaTiO_3$-MgO | 0.9 | 73.3 | 8 |
| $BaTi_{0.7}Zr_{0.3}O_3$ | 0.51 | 72.2 | 9 |
| $BaTiO_3$-$SiO_2$ | 1.43 | 53 | 10 |
| $0.92BaTiO_3$-$0.08K_{0.5}Bi_{0.5}TiO_3$ | 0.89 | 56 | 11 |
| $BaTiO_3$/$BaTiO_3$@$SiO_2$ | 1.8 | 71.5 | 12 |
| $BaTiO_3$-$Bi_{0.5}Na_{0.5}TiO_3$-$Na_{0.73}Bi_{0.09}NbO_3$ | 1.7 | 82 | 13 |
| $Ba_{0.95}Ca_{0.05}Zr_{0.2}Ti_{0.8}O_3$ | 0.41 | 72 | 14 |
| $BaTiO_3$@$BiScO_3$ | 0.68 | 81 | 15 |
| $0.62BiFeO_3$-$0.3BaTiO_3$-$0.08Nd(Zn_{0.5}Zr_{0.5})O_3$ | 2.45 | 72 | 16 |
| $0.65BiFeO_3$-$0.3BaTiO_3$-$0.05Bi(Zn_{2/3}Nb_{1/3})O_3$ | 2.06 | 53 | 17 |
| $Sr_{0.985}Ce_{0.01}TiO_3$@$SiO_2$ | 1.2 | 50 | 18 |

## 11. References


[1] Y. H. Huang, Y. J. Wu, B. Liu, T. N. Yang, J. J. Wang, J. Li, L. Q. Chen and X. M. Chen. *J. Mater. Chem. A.*, 2018, **6**, 4477-4484.

[2] C. Q. Zhu, X. H. Wang, Q. C. Zhao, Z. M. Cai, Z. Y. Cen and L. T. Li. *J. Eur. Ceram. Soc.*, 2019, **39**, 1142-1148.

[3] B. B. Liu, X. H. Wang, R. X. Zhang, L. T. Li. *J. Am. Ceram. Soc.*, 2017, **100**, 3599-3607.

[4] L. W. Wu, X. H. Wang, H. L. Gong, Y. N. Hao, Z. B. Shen and L. T. Li. *J. Mater. Chem. C.*, 2015, **3**, 750-758.

[5] B. B. Liu, X. H. Wang, R. X. Zhang and L. T. Li. *J. Alloy. Compd.*, 2017, **691**, 619-623.

[6] S. J. Liu, Q. D. Xie, L. X. Zhang, Y. Y. Zhao, X. Wang, P. Mao, J. P. Wang and X. J. Lou. *J. Eur. Ceram. Soc.*, 2018, **38**, 4664-4669.

[7] Q. C. Zhao, X. H. Wang, H. L. Gong, B. B. Liu, B. C. Luo and L. T. Li. *J. Am. Ceram. Soc.*, 2018, **101**, 1245-1254.

[8] G. Liu, L. Y. Zhang, Q. K. Wu, Z. Y. Wang, Y. Li, D. Q. Li, H. B. Liu and Y. Yan. *J. Mater. Sci-mater. El.*, 2018, **29**, 18859-18867.

[9] B. Liu, Y. Wu, Y. H. Huang, K. X. Song and Y. J. Wu. *J. Mater. Sci.*, 2019, **54**, 4511-4517.

[10] X. Lu, L. Zhang, H. Talebinezhad, Y. Tong and Z. Y. Cheng. *Ceram. Int.*, 2018, **44**, 16977-16983.

[11] J. Wang, Y. P. Pu, C. Y. Hui, C. W. Cui and Y. S. Guo. *J. Mater. Sci-mater. El.*, 2018, **29**, 6556-6563.

[12] Q. B. Yuan, J. Cui, Y. Y. Wang, R. Ma and H. Wang. *J. Eur. Ceram. Soc.*, 2017, **37**, 4645-4652.

[13] H. B. Yang, F. Yan, Y. Lin, T. Wang, F. Wang, Y. L. Wang, L. N. Guo, W. D. Tai and H. Wei. *J. Eur. Ceram. Soc.*, 2017, **37**, 3303-3311.

[14] D. Zhan, Q. Xu, D. P. Huang, H. X. Liu, W. Chen and F. Zhang. *J. Phys. Chem. Solids.*, 2018, **114**, 220-227.

[15] L. W. Wu, X. H. Wang and L. T. Li. *J. Alloy. Compd.*, 2016, **688**, 113-121.

[16] G. Wang, J. L. Li, X. Zhang, Z. M. Fan, F. Yang, A. Feteira, D. Zhou, D. C. Sinclair, T. Ma, X. L. Tan, D. W. Wang and I. M. Reaney. *Energy Environ. Sci*., 2019, **12**, 582-588.

[17] D. W. Wang, Z. M. Fan, W. B. Li, D. Zhou, A. Feteira, G. Wang, S. Murakami, S. K. Sun, Q. L. Zhao, X. L. Tan and I. M. Reaney. *ACS. Appl. Energy Mater*., 2018, **1**, 4403-4412.

[18] J. L. Qi, M. H. Cao, J. P. Heath, J. S. Dean, H. Hao, Z. H. Yao, Z. Y. Yu and H. X. Liu. *J. Mater. Chem. C.*, 2018, **6**, 9130-9139.