Supplementary Information

Preferential Perovskite Surface-termination Induced High Piezoresponse in Lead-free *in-situ* Fabricated $Cs_3Bi_2Br_9$-PVDF Nanocomposites Promotes Biomechanical Energy Harvesting

Aditi Sahoo[1], Tufan Paul[1], Ankan Nath[2], Soumen Maiti[3], Prabhat Kumar[1], Prasenjit Ghosh[2] and Rupak Banerjee[1, 4]*

[1]*Department of Physics, Indian Institute of Technology Gandhinagar, Palaj, Gandhinagar 382355, India*

[2]*Department of Physics, Indian Institute of Science Education and Research, Dr. Homi Bhabha Road, Pune 411008, India*

[3] *St. Thomas Colleges of Engineering & Technology, Kolkata 700023, India*

[4]*K C Patel Centre for Sustainable Development, Indian Institute of Technology Gandhinagar, Palaj, Gandhinagar 382355, India*

*Corresponding author email: rupakb@iitgn.ac.in

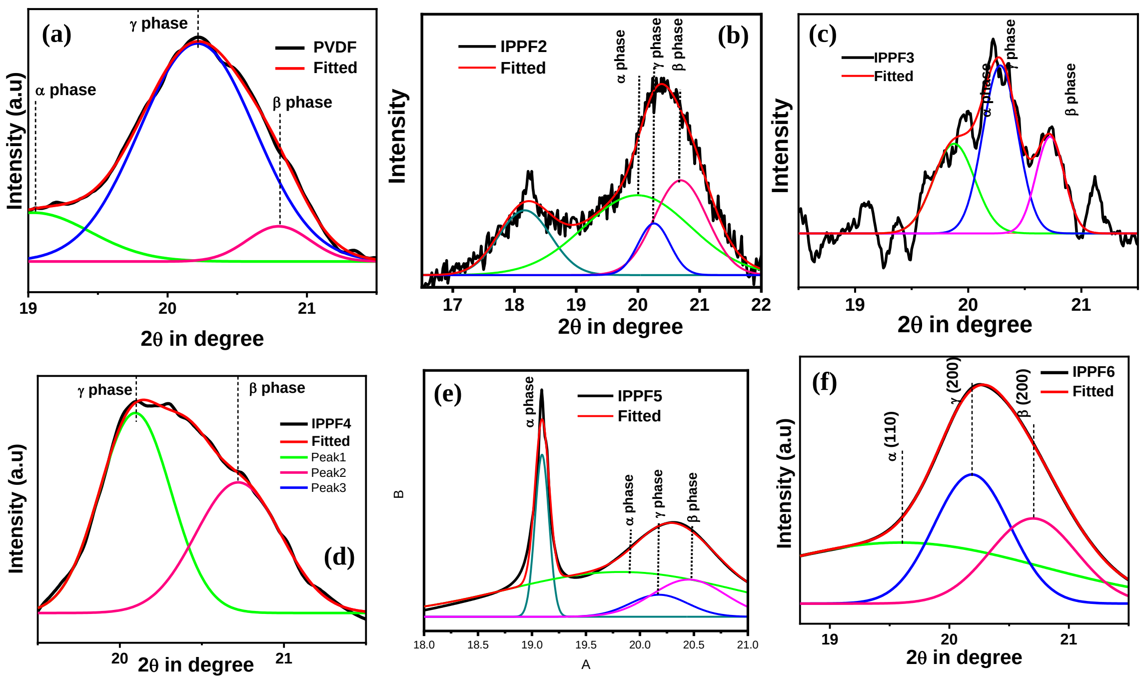


Figure S1: (a)-(f) *The deconvoluted XRD peak profiles are showing the peaks corresponding to* α, β *and,* γ *phases in PVDF and all composite films.*

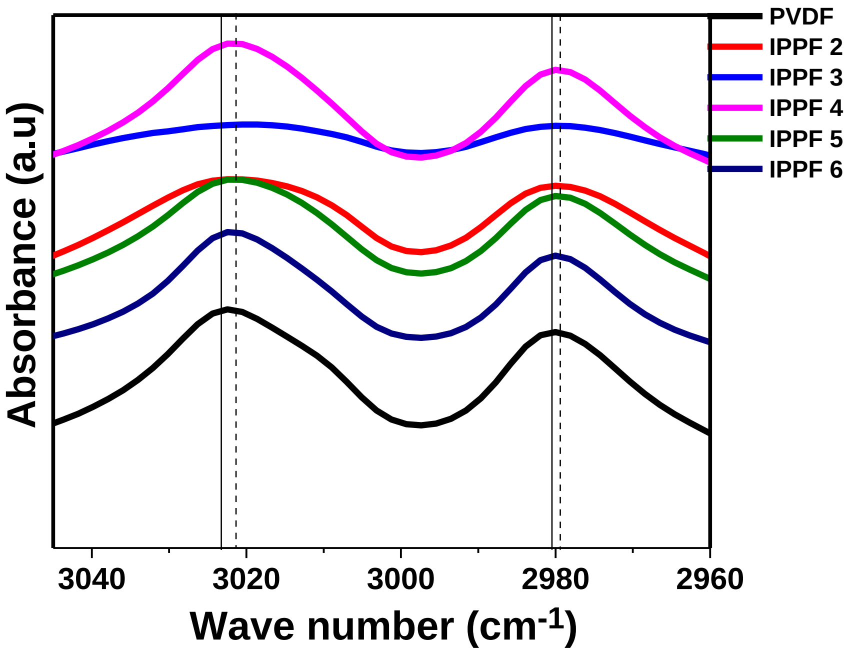


Figure S2: *Shift in FTIR stretching vibration of PVDF and all composite films.*

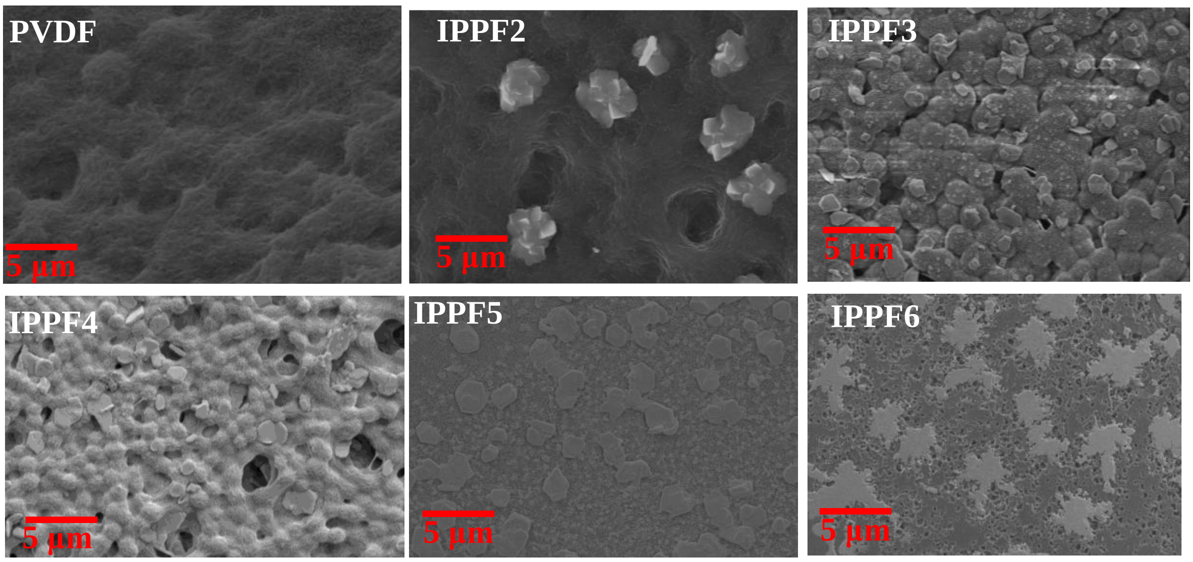


Figure S3: *FESEM image of (a) pristine PVDF and (b) IPPF2, (c) IPPF3, (d) IPPF4, (e) IPPF5, (f) IPPF6.*

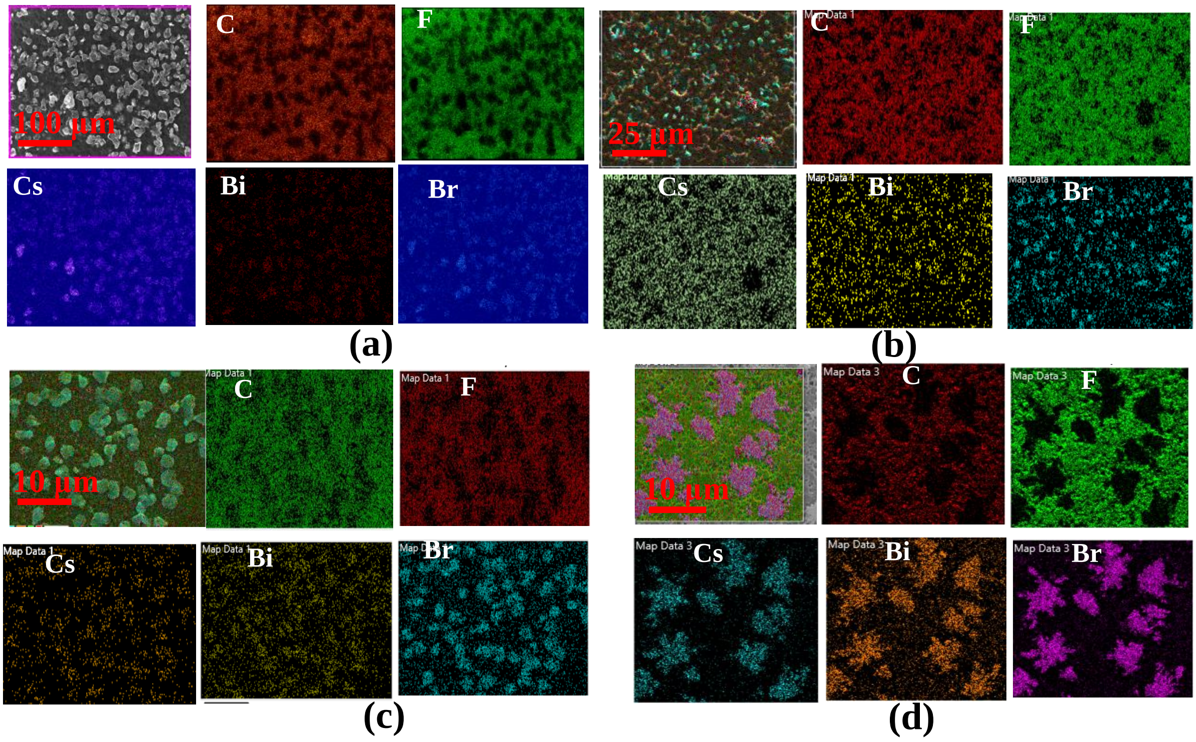


Figure S4: *EDX mapping of the composite films (a) IPPF2, (b) IPPF3, (c) IPPF5, (d)IPPF6.*

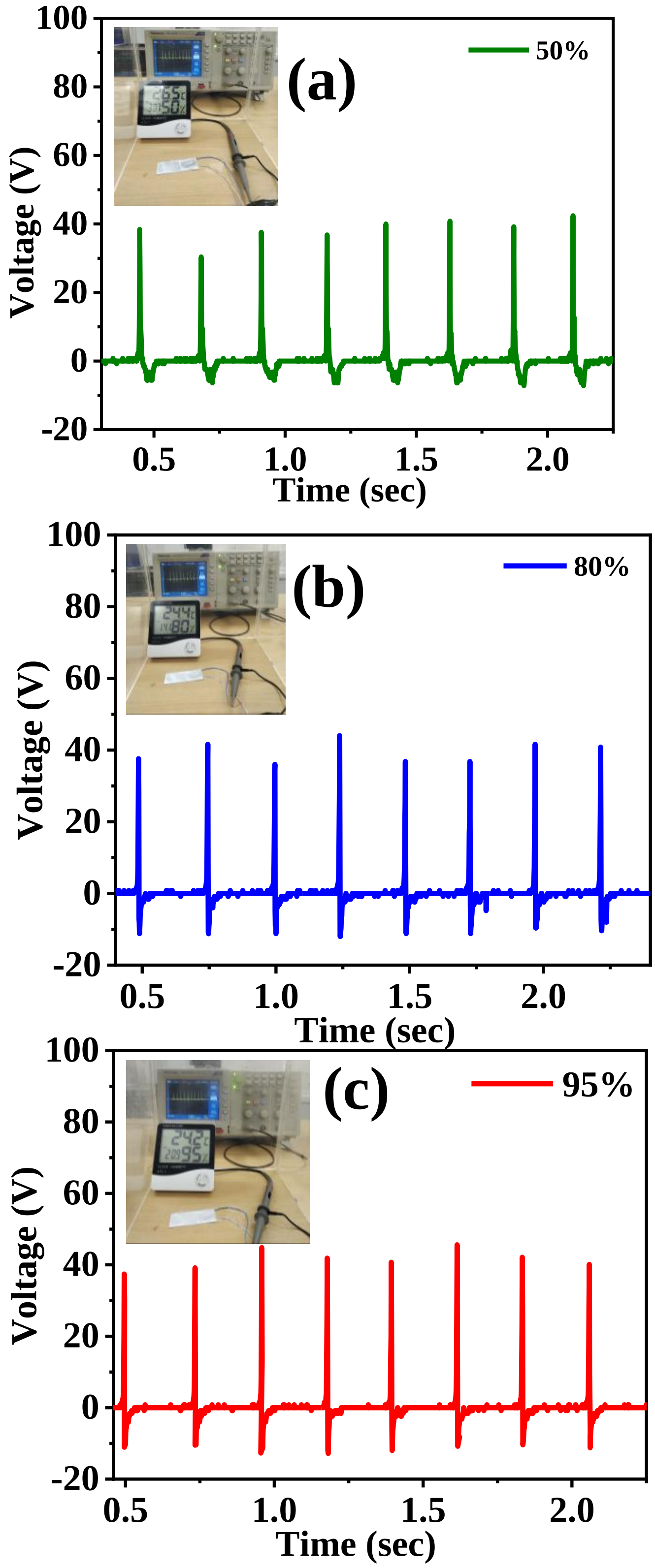
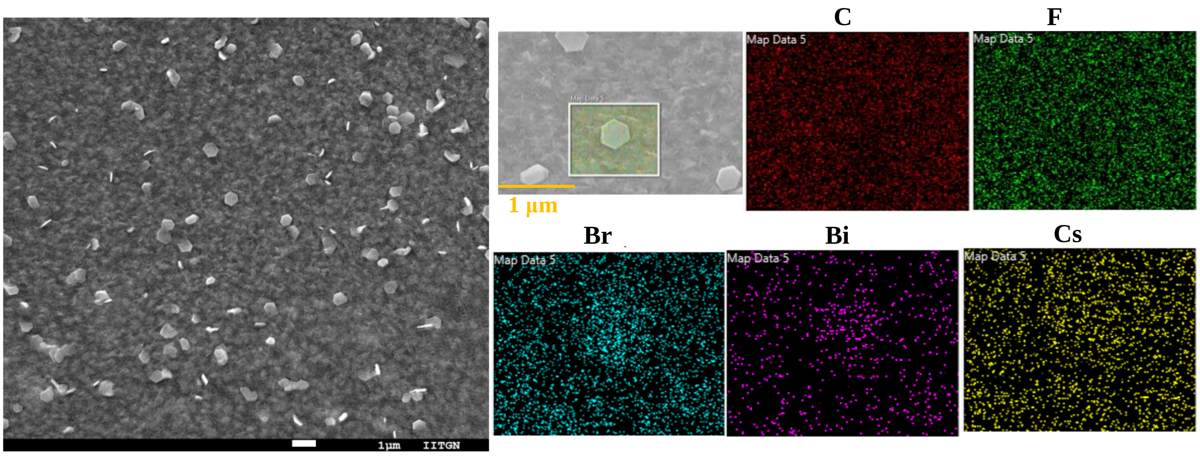


Figure S5:. *The performance of the nanogenerator with the humidity of the environment from (a) 50%   to (b) 80%, and finally to (c) 95%. Inset shows real-time images of the operation of the nanogenerator device in controlled humidity conditions.*



(a) (b)

Figure S6: *(a) FESEM image and (b) EDX mapping of IPPF4 after long cycle operation.*

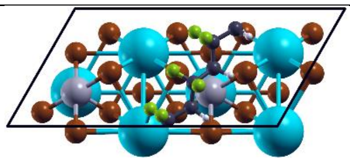
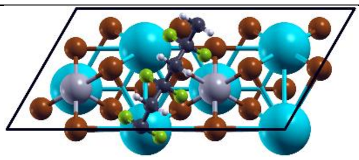
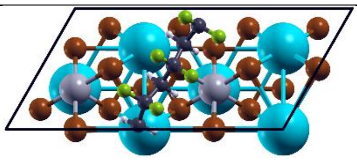
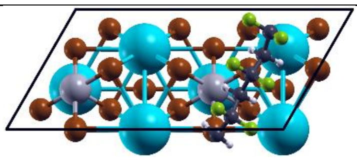
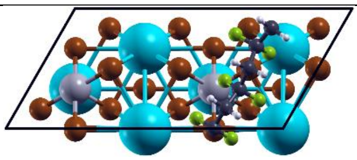
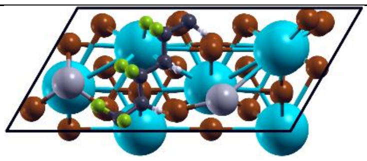
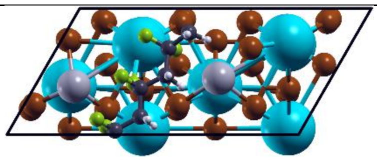
Table S1: Piezoelectric co-efficient comparison

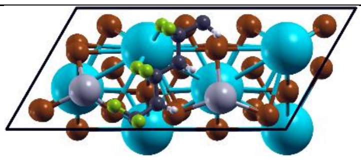
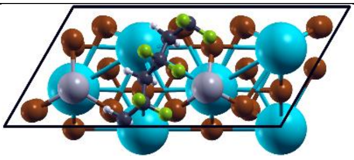
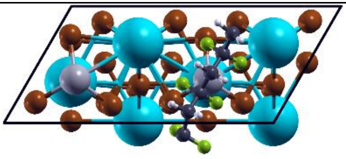
Material	**Piezoelectric constant (d_{33}) (pC/N)**	**References**
$CH_3NH_3PbI_3$	5.12	1
ZnO	3	2
GaN	3.1	3
PZT	290	4
$BaTiO_3$	190	5
$LiNbO_3$	11	6
$K_{0.8}Na_{0.2}NbO_3$	110	7
FAPbBr3	25	8
MDABCO-NH_4I_3	14	9
(4-aminotetrahydropyran)2$PbBr_4$	76	10
Insitu PVDF@$Cs_3Bi_2Br_9$	**10**	**This work**

Table S2: Nanogenerator performance comparison

Polymer	Filler/ Device structure	Pressure/ Force	Output voltage	Current / Current density	Application	Stability / Cycle	Ref.
PVDF	$FAPbBr_3$	Finger imparting	26.2 V	2.1 μA	LCD screen, speaker etc.	4 weeks	11
PVDF	$BaTi_{(1-x)}Zr_xO_3$ (BTZO)		11.9 V	1.35 μA			12
PVDF - TrFE)	$CsPbBr_3$ QDs	0.6 MPa	11.5 V		Powering LEDs	1200 cycles	13
PEDOT - coated PVDF		8.3 kPa	48 V	6 μA	Weight Measurement Mapping, Vibration Sensor	21000 cycles	14
PDMS	$ZnSnO_3$	Repeating finger	40 V	0.4 μA	Glowing of LEDs		15
PVDF	DNA	13 kPa	6 V	0.088 μA	Glowing of LEDs		16
PDMS	PZT	Bending	10 V	1.3 μA	Glowing of LEDs	3 days / 6000 cycle	17
PVDF	$MAPbBr_3$	Finger touch	5 V	60 nA	Responses observed from NG as different letters	3600 cycles	18
PDMS	$FAPbBr_3$		8.5 V	3.8 $\mu A/cm^2$			19
PVDF	**$Cs_3Bi_2Br_9$**	**Biomechanical Motions**	**40 V**	**4.1 μA**	**LED glowing**	**1000 cycles/ 6 months**	**This work**

Table S3: Different configurations of a single strand of β-PVDF and the perovskite interface for the Bi and $CsBr_3$ terminations of the (0001) surface of $Cs_3Bi_2Br_9$.

Interface Configuration	Binding energy (eV/monomer)
Bi-termination	
	**1.13**
	**-0.70**
	**-0.66**
	**-0.56**
	**-0.53**
$CsBr_3$ termination	
	**-2.13**
	**-1.81**

	**-1.59**
	**-1.12**
	**-0.45**

References:

1. Y. J. Kim, T. V. Dang, H. J. Choi, B. J. Park, J. H. Eom, H. A. Song, D. Seol, Y. Kim, S. H. Shin, J. Nah, *J. Mater. Chem. A*, 4 (2016) 756.
2. N. H. Langton, D. Matthews, Br. *J. Appl. Phys.*, 9 (1958) 453.
3. C. M. Lueng, H. L. W. Chan, C. Surya, C. L. Choy, *J. Appl. Phys.*, 88 (2000) 5360.
4. B. Jaffe, *J. Am. Ceram. Soc.*, 41 (1958) 494.
5. A. G. Chynoweth, *J. Appl. Phys.* 27 (1956) 78.
6. L. A. Reznitchenko, A. V. Turik, E. M. Kuznetsova, V. P. Sakhnenko, *J. Phys.: Condens. Matter*, 13 (2001) 3875.
7. H. Tian, C. Hu, X. Meng, Z. Zhou, G. Shi, *J. Mater. Chem. C*, 3 (2015) 9609.
8. A. A. Zhumekenov, M. I. Saidaminov, M. A. Haque, E. Alarousu, S. P. Sarmah, B. Murali, I. Dursun, X.-H. Miao, A. L. Abdelhady, T. Wu, O. F. Mohammed, O. M. Bakr, *ACS Energy Lett.*, 1 (2016) 32.
9. J. Young, J. M. Rondinelli, *Phys. Rev. Mater.*, 2 (2018) 065406.

10. X.-G. Chen, X.-J. Song, Z.-X. Zhang, P.-F. Li, J.-Z. Ge, Y.-Y. Tang, J.-X. Gao, W.-Y. Zhang, D.-W. Fu, Y.-M. You, *J. Am. Chem. Soc.*, 142 (2019) 1077.
11. S. K. Si, S. Paria, S. K. Karan, S. Ojha, A. K. Das, A. Maitra, A. Bera, L. Halder, A. De, and B. B. Khatua, *Nanoscale*, 12 (2020) 7214.
12. N. R. Alluri, B. Saravanakumar and S. J. Kim, *ACS Appl. Mater. Interfaces*, 7 (2015) 9831.
13. J. Nie, L. Zhu, W. Zhai, A. Berbille, L. Li and Z.L. Wang, *ACS Applied Electronic Materials*, 3 (2021), 2136.
14. K. Maity, and D. Mandal, *ACS Appl. Mater. Interfaces*, 10 (2018) 18257
15. M.M. Alam, S.K. Ghosh, A. Sultana, and D. Mandal, *Nanotechnology*, 26 (2015) 165403.
16. A. Tamang, S.K. Ghosh, S. Garain, M.M. Alam, J. Haeberle, K. Henkel, D. Schmeisser, and D. Mandal, *ACS Appl. Mater. Interfaces*, 7 (2015) 16143.
17. K.I. Park, C.K. Jeong, J. Ryu, G.T. Hwang, and K. J. Lee, *Advanced Energy Materials*, 3 (2013) 1539.
18. A. Sultana, M.M. Alam, P. Sadhukhan, U.K. Ghorai, S. Das, T.R. Middya, and D. Mandal, *Nano Energy*, 49 (2018) 380.
19. R. Ding, H. Liu, X. Zhang, J. Xiao, R. Kishor, H. Sun, B. Zhu, G. Chen, F. Gao, X. Feng and J. Chen, *Adv. Funct. Mater.*, 26 (2016) 7708.