UDK 621.38:620.3:681.6-33
Original scientific article/Izvirni znanstveni članek

ISSN 1580-2949
MTAEC9, 52(2)231(2018)

A SCREEN-PRINTED PIEZOELECTRIC ENERGY HARVESTER USING ZnO TETRAPOD ARRAYS

SITOTISK PIEZOELEKTRIČNEGA ZBIRALCA ENERGIJE Z UPORABO ZnO TETRAPODNIH MATRIC

Quili Dai[1,2], Peng Pan[1,2], Ruifeng Zhang[1,2], Jun Liu[2], Zhengcun Yang[2], Jun Wei[2], Qiping Yuan[1,2]

[1]Tianjin University of Technology, Tianjin Key Laboratory of Film Electronic and Communicate Devices, School of Electrical and Electronic Engineering, Tianjin 300384, China
[2]Tianjin University of Technology, Advanced Materials and Printed Electronics Center, School of Electrical and Electronic Engineering, Tianjin 300384, China
13072219279@163.com

Prejem rokopisa – received: 2017-09-25; sprejem za objavo – accepted for publication: 2017-10-20

doi:10.17222/mit.2017.157

In this work, for the first time, we combine electronic printing technology, electrostatic spraying technology and the new 3D structure of ZnO. The new structure was prepared with a simple, low-cost and massive method that was not successfully applied before. The ZnO tetrapods/PET composite device exhibits an output open-circuit voltage of ~20 V and a short-circuit current of ~300nA under repeated hand pressing. It is demonstrated that the output power from the PENG can directly drive the light-emitting diodes (LEDs) and charge a capacitor. But a typical flexible polymer produces an output voltage of 3.3 V and cannot power light-emitting diodes.

Keywords: ZnO tetrapods, wearable electronics, nanogenerator, screen-printed

V tem članku avtorji prvič opisujejo kombinacijo tehnologije elektronskega tiska, tehnologije elektrostatičnega napr-ševanja in nove 3D-strukture ZnO. Novo strukturo so pripravili z enostavno, ceneno in produktivno metodo, ki do sedaj še ni bila razvita. Kompozitna naprava sestavljena iz ZnO tetrapodov in polietilen tereftalata (PET), ima v odprtem tokokrogu napetost ~ 20 V in kratkostični tok ~ 300 nA pod vplivom ponavljajočega ročnega stiskanja. Avtorji so v raziskavi dokazali, da je izhodna moč piezoelektričnega nanogeneratorja (PENG) dovolj velika, da lahko neposredno poganja svetlobne diode (LEDs; angl.: light-emitting diodes) in polni kondenzator. Toda tipični upogljivi (fleksibilni, tisti ki se prilagajajo telesu) polimerni proizvodi imajo izhodno napetost 3,3 V in ne morejo proizvajati svetlobe z LED-icami.

Ključne besede: ZnO tetrapodi, nosljiva elektronika, nanogenerator, sitotisk

1 INTRODUCTION

In various renewable and sustainable energy environments, mechanical energy is the largest widely distributed resource[1,2] with a variety of energy types and scales, such as pressure, sound, rolling tires, vibration, and tides. In order to solve the problem of increasing energy crisis and realize the low-energy-consumption electronics of power supply, various methods have been developed to convert mechanical energy into electricity, including the piezoelectric effect, electrostatic[3,4] etc. In practice, each kind of generator has its own shortcoming, i.e., it can simultaneously mass produce, and popularize multi-type generator[5] to improve energy production harvester. In the previous work, the simulation analysis of the coupling between the piezoelectric friction and electronics have been promoting the output performance of a piezoelectric–triboelectric hybrid nanogenerator can be achieved through proper structural design and the direction of the polarization enhancement.[6]

In this work we fabricated a novel structured piezoelectric nanogenerator (PENG) based on polyethylene terephthalate (PET) and polydimethysiloxane (PDMS) film that was printing carbon paste with screen-printed and electrostatic spraying ZnO tetrapods.[7,8] In the demonstration, the ZnO tetrapods four of the three legs are arranged on the substrate below, and look like a tetrahedron. These ZnO tetrapods were synthesized through a simple, low-cost, low-temperature hydrothermal method,[9,10] which can facilitate large-scale production and preparation. The electrical output of a PENG with surface modification of the ZnO tetrapods could reach 20 V at low frequency, about 2.3 times higher than that without any ZnO materials. This clearly suggests that this kind of surface modification can dramatically enhance the electrical output of the PENG, providing a unique microstructured material for the PENG.

2 EXPERIMENTAL PART

2.1 Materials

The graphite used in the experiments was purchased from Sigma-Aldrich. Polyethylene terephthalate (PET) and polydimethysiloxane (PDMS) film, 99 % pure Zn powder, ethanol and acetone were obtained from Sigma-Aldrich.

2.2 Preparation of ZnO nanotetrapods

ZnO nanotetrapods were synthesized with 99 % pure Zn powder by a thermal evaporation and oxidation method in air. An induction-heating device with a maximum output power of 60 kW and a constant frequency of 60 kHz was used as a heating source. The induction coil made of a copper tube was processed into a shape of planar polyring. First, the Zn powder was paved evenly onto a sheet of graphite paper. The graphite paper was parallel to the induction coil and the distance between them kept at 30 mm. In order to investigate the effect of the output power on the morphologies of the ZnO nanotetrapods[11,12] the applied value of output power varied from 20 kW to 40 kW. After a few seconds from starting the induction-heating device, white cotton-like product filled the space between the induction coil and the graphite paper.

2.3 Fabrication of piezoelectric nanogenerator films

The white products of ZnO nanotetrapods were obtained in the crucibles and were collected for the next step to use. Before the in printing carbon paste, the substrates were cleaned ultrasonically in acetone, ethanol and deionized water. Then the substrates were dried at 60 °C for 0.5 h and printed with carbon paste using screen-printing technology. Finally, the ZnO tetrapods on the basis of printing carbon paste by electrostatic spraying and oven for half an hour at 110 °C in the air at atmospheric pressure.

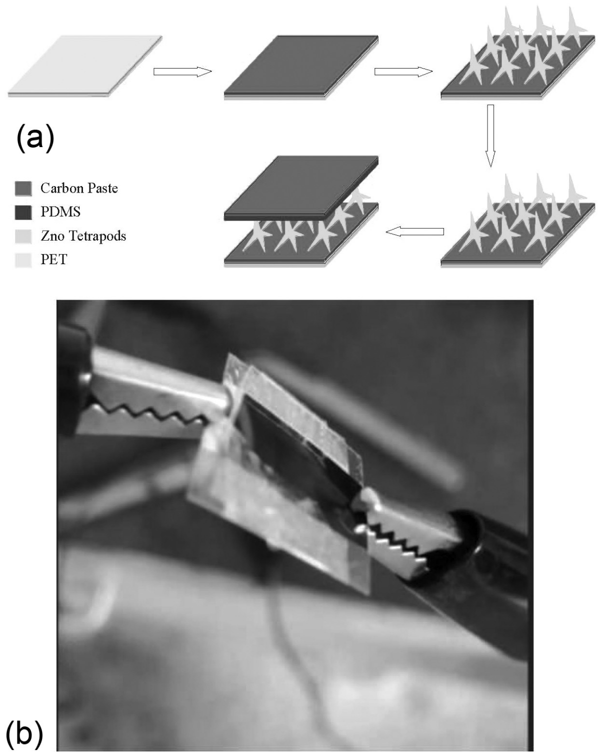

Figure 1: a) Schematic diagrams of the PENG fabricating process, b) photograph of the ZnO-PET composite film contact by wires

The schematic diagram of the PENG manufacturing process is introduced in **Figure 1a**. The PENG were manufactured by covering the top PDMS with graphite as electrodes and bottom surfaces of graphite paste on a polyethylene terephthalate (PET) substrate. The particular information about the manufacturing process is described in the Experimental details. The dimension of the fabricated PENG is the rectangular area of mm^2 show in **Figure 1b**.

3 RESULTS AND DISCUSSION

Figure 2 shows typical scanning electron microscopy (SEM) images (**Figure 2a** and **2b**) of the as-synthesized

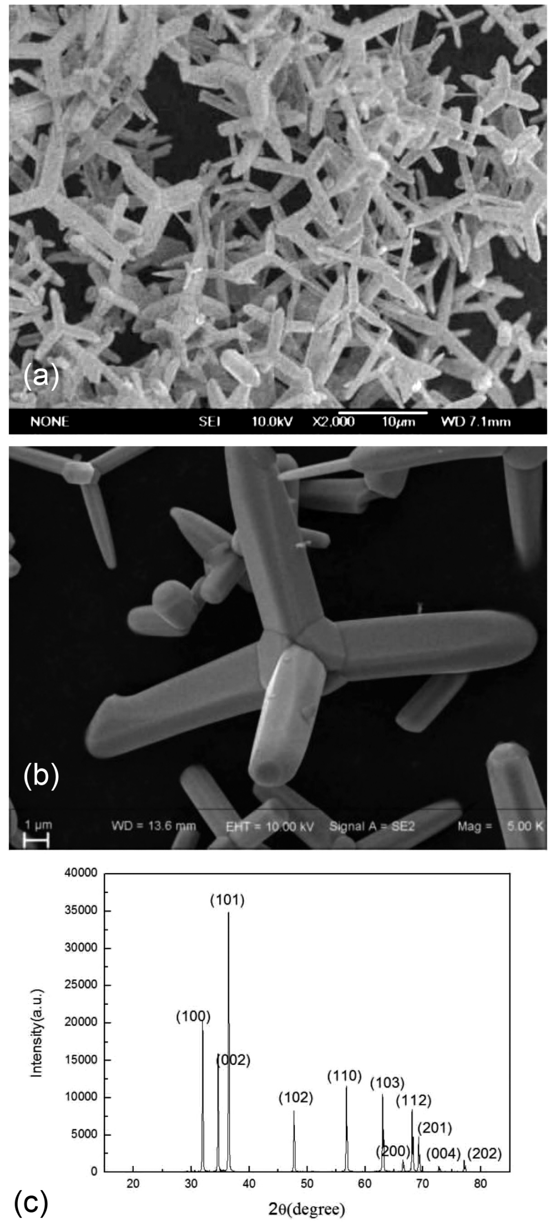

Figure 2: Representative SEM images shown up view: a) and b) of ZnO nanotetrapods, c) Rietveld-refinement analysis of XRD spectra of ZnO nanotetrapods

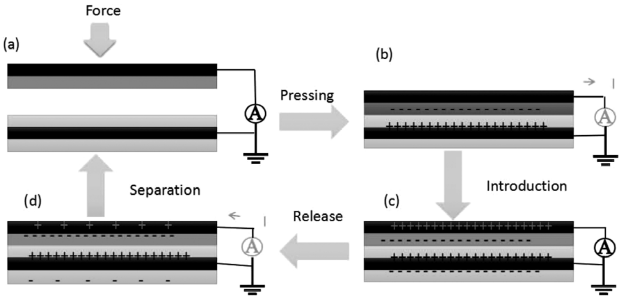

Figure 3: Working mechanism of the sandwich-shape PNG. Schematic illustration of the charge-generating process of the PNG. Pushed by the external force from the linear motor, the device will be switching back and forth between the separated state and the contacted state, and there will be an alternating flow of electrons in the external circuit driven by the induced priboelectric potential. The electric charges move for neutralization after the separation, forming a cycle.

products prepared under different magnifications. The SEM image shows that the products are composed of four-dollar ZnO crystals and extends four legs from the center. **Figure 2c** is the XRD mode of ZnO, while the sharp diffraction peak indicates good crystal quality. All the 12 diffraction peaks can be associated with ZnO crystals (JCPDS 36-1451) of hexagonal fiber structure, indicating that ZnO crystals have a high purity.[13]

The pattern is a vertical contact-separation mode of PENG. The detailed working principle is shown in **Figure 3a**. When the PENG is pressed, the two friction materials contact. According to the previous works, due to the different electron-attracting ability, is transported from one material to another electrons[14] so there will be a net negative charge of the surface layer and attract electron ability and net positive charge **Figure 3b**. Two tribo-materials, independent of the interface area of the tribo-materials earlier separation, this will lead to the interface area being saved, so the electronic connection induction electrode will drive the flow from one side to to another (creating a current cycle, **Figure 3c**). In the process, the electron lasts until the PENG is fully released, shown in **Figure 3d**. At this point, both the induces the potential difference and the transfer charge of the maximum.[15,16] As these two tribo-materials dissociate,[17,18] the potential differences will decrease and the electrons will also appear and Returns even fade away (**Figure 3a**). Therefore, the whole process will produces an optional current pulse output. When two thin films are exposed and separated, the replacement potential will drive the electrons in the external load to move back and forth. But typical flexible polymer produces an output voltage of 3.3 V and cannot power light-emitting diodes.

Open-circuit voltage (Voc) and short-circuit current (Isc) are measured to characterize the PENG electric performance when it is coated with graphite.[19] With a repeating finger imparting (pressure amplitude ~8.43 kPa, online supplementary data) the Voc is presented in **Figure 4a**. Furthermore, the AC output could be transferred to the pulse output in the same direction simply by introducing a full-wave rectifying bridge (**Figure 4d**). At an applied pressure of 8.43 kPa, the PENG is able to produce a Voc up to 20 V and the short-circuit current Isc is 300 nA. When the force decreases, (pressure amplitude ~3.45 kPa) the output voltage decreases as the Voc is 6 V (**Figure 4b**). This is attributed to the increased contact area between the electrode and the PDMS with a larger applied pressure amplitude. The PDMS shows an elastic property; the larger force PDMS can fill more vacant space, thus leading to a larger contact area. As a result, the electric output voltage and current increase until all of the vacant spacing is completely filled by the PDMS,[20] reaching an enhanced output. Different values of the resistors are connected as a PENG to further investigate the effective electric power of the PENG. As demonstrated in **Figure 4c**, the instantaneous current drops with increasing load resistance due to ohmic losses, while the voltage builds up.[21,22] Consequently, the instantaneous power output ($W = I^2_{peak}\ R$) reached the maximum at a load resistance of 0.6 MΩ. At a contacting force of 8.43 kPa, an power output of 0.4 μW was achieved (**Figure 4d**).To confirm the energy storage application of the PENG, a capacitor was connected to the NG using a full-wave bridge rectifying circuit.[23]

The electric power generated by the PENG is directly used for the opening of commercial LEDs lights.[24] In the process of finger tapping, 9 commercial LEDS are driven

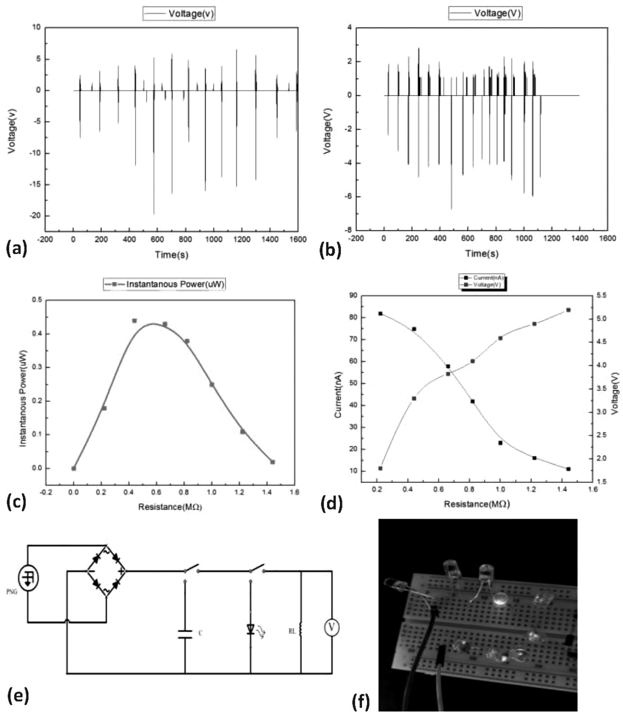

Figure 4: a) and b) under different finger pressures rectified output voltage of the PNG, c) fitted curve of the current and voltage output on different external load resistance, d) dependence of the power output on the different external load resistance, e) equivalent bridge rectifier circuit diagram for the DC voltage measurement, capacitor charging and LED lighting performance, f) the LEDs were driven successively by output generated from hand pressing

by the resulting output voltage (**Figure 4f**), which does not require external energy-storage devices (inset of **Figure 4e**).

4 CONCLUSIONS

In summary, we have successfully provided a simple, low-cost and massive production method to fabricate ZnO tetrapods electrets film based on nanogenerator, e.g., PENG. The mechanical energy-harvesting ability of the PENG is demonstrated by powering 9 light-emitting diodes (LEDs) by a simple finger touch. For the traditional energy-harvester flexible polymer produces an output voltage of 3.3 V and cannot power light-emitting diodes (LEDs). This is a simple and cost-effective method to realize the output enhancement of piezoelectric PENG. High output enables the energy harvesters to be used as power sources for light-emitting diodes or rechargeable capacitors, showing great potential in the field of self-powered systems or sensor networks.

Acknowledgments

This work was financially supported by The Recruitment Program of Global Experts and Advanced Materials and Printed Electronics Center.

5 REFERENCES

[1] S. Jana, S. Garain, S. K. Ghosh, The preparation of ¦$\tilde{A}$-crystalline non-electrically poled photoluminescant ZnO-PVDF nanocomposite film for wearable nanogenerators, Nanotechnology, 27 (**2016**), doi:10.1088/0957-4484/27/44/445403

[2] X. Chen, H. Chen, H. Zhang, A wave-shaped hybrid piezoelectric and triboelectric nanogenerator based on P(VDF-TrFE) nanofibers, Nanoscale, 9 (**2017**), 1263–1270, doi:10.1039/c6nr07 781a

[3] F. H. Alsultany, Z. Hassan, N. M. Ahmed, A high-sensitivity, fast-response, rapid-recovery UV photodetector fabricated based on catalyst-free growth of ZnO nanowire networks on glass substrate, Optical Materials, 60 (**2016**), 30–37, doi:10.10 16/j.optmat.2016.07.004

[4] N.R. Alluri, S. Selvarajan, A. Chandrasekhar, Self powered pH sensor using piezoelectric composite worm structures derived by ionotropic gelation approach, Sensors and Actuators B, 237 (**2016**), 534–544, doi:10.1016/j.snb.2016.06.134

[5] E. Modaresinezhad, S. Darbari, Realization of a room-temperature/self-powered humidity sensor, based on ZnO nanosheets, Sensors and Actuators B, 237 (**2016**), 358–366, doi:10.1016/j.snb.2016.06.097

[6] Y. Yan, C. Li, L. Zhou, B. Liu, J. Zhang, Tuning aspect ratio of hierarchical ZnO nanotetrapod, Appl. Phys. A, (**2016**) 122:1016, doi:10.1007/s00339-016-0555-0

[7] K. Baba, C. Lazzaroni, M. Nikravech, ZnO and Al doped ZnO thinfilms deposited by Spray Plasma: Effect of the growth time and Al doping on microstructural, optical and electrical properties, Thin Solid Films, 595 (**2015**) 129–135, doi:10.1016/j.tsf.20 15.10.072

[8] Q. Yang, D. Wang, M. Zhang, T. Gao, H. Xue, Lead-free (Na0.83K0.17)0.5Bi0.5TiO3 nanofibers for wearable piezoelectric nanogenerators, Journal of Alloys and Compounds 688 (**2016**), 1066–1071, doi:10.1016/j.jallcom.2016.07.131

[9] J. Yoo, S. Cho, W. Kim, J. Kwon, Effects of mechanical deformation on energy conversion effi ciency of piezoelectric nanogenerators, Nanotechnology, 26 (**2015**), 275402, doi:10.1088/0957-4484/26/27/275402

[10] Z. Q. Zheng, J. D. Yao, B. Wang, G. W. Yang, Light-controlling, flexible and transparent ethanol gas sensor based on ZnO nanoparticles for wearable devices, Scientific Reports, 35 (**2015**), doi:10.1038/srep11070

[11] X. Xue, Z. Qu,Y. Fu, B. Yu, L. Xing, Y. Zhang, Self-powered electronic-skin for detecting glucose level in body fluid basing on piezo-enzymatic-reaction coupling process, Nano Energy, 26 (**2016**), 148–156, doi:10.1016/j.nanoen.2016.05.021

[12] C. Chey, X. Liu, H. Alnoor, O. Nur, M. Willander, Fast piezoresistive sensor and UV photodetector based on Mn-doped ZnO nanorods, Phys. Status Solidi RRL, 9 (**2015**) 87–91, doi:10.1002/pssr.201409453

[13] D. Berger, A. P. de Moura, L. H. Oliveira, W. B. Bastos, F. A. La Porta, Improved photoluminescence emission and gas sensor properties of ZnO thin films, Ceramics International 42 (**2016**), 13555–13561, doi:10.1016/j.ceramint.2016.05.148

[14] S. A. Hassanzadeh-Tabrizi, M. Motlagh, S. Salahshour, Synthesis of ZnO/CuO nanocomposite immobilized on γ-Al_2O_3 andapplication for removal of methyl orange, Applied Surface Science, 384 (**2016**), 237–243, doi:10.1016/j.apsusc.20 16.04.165

[15] W. Qin, Y. Li, T. Li, J. Qiu, Xi. Ma, Microstructure-related piezoelectric properties of a ZnO film grown on a Si substrate, Ceramics International, 42 (**2016**), 16927–16934, doi:10.1016/j.ceramint.2016.07.192

[16] D. Talarico, F. Arduini, A. Constantino, D. Moscone, G. Palleschi, Carbon black as successful screen-printed electrode modifier for phenolic compound detection, Electrochemistry Communications 60 (**2015**), 78–82, doi:10.1016/j.elecom.2015.0 8.010

[17] W. Li, D. Torres, T. Wang, C.Wang, N. Sepúlveda, Flexible and biocompatible polypropylene ferroelectret nanogenerator (FENG): On the path toward wearable devices powered by human motion, Nano Energy, 30 (**2016**), 649–657, doi:10.1016/j.nanoen.2016.10.007

[18] F. Fan, W. Tang, Z. Wang, Flexible Nanogenerators for Energy Harvesting and Self-Powered Electronics, Advanced Materials, 28 (**2016**), 4283–4305, doi:10.1002/adma.201504299

[19] G. Zhao, L. Xian, S. Wu, L. Song, A. Wei, G. Wen, Ultrafast and mass production of ZnO nanotetrapods by induction-heating under air ambient, Materials Letters, 23 (**2013**), 126–129, doi:10.1016/j.matlet.2013.12.077

[20] P. S. Das, J. Y. Park, Human skin based flexible triboelectric nanogenerator using conductive elastomer and fabric films, Electronics Letters, 52 (**2016**), 1885–1887, doi:10.1049/el.2016.3174

[21] X. Yan, H. Dong, Y. Li, C. Lin, Phase transition induced strain in ZnO under high pressure, Scientific Reports, 14 (**2016**), doi:10.1038/srep24958

[22] K. Y. Shin, J. S. Lee, J. Jang, Highly sensitive, wearable and wireless pressure sensor using free-standing ZnO nanoneedle/PVDF hybrid thin film for heart rate monitoring, Nano Energy, 22 (**2016**), 95–104, doi:10.1016/j.nanoen.2016.02.012

[23] Y. Luo, Y. Zhao, L. Hu, Piezo-phototronic effect enhance d pressure sensor based on ZnO/NiO core/shell nanorods array, Nano Energy, 21 (**2016**), 106–114, doi:10.10 16/j.nanoen.2016.01.007

[24] X. Ren, H. Fan, Y. Zhao, Z. Liu, Flexible Lead-Free $BiFeO_3$/PDMS-Based Nanogenerator as Piezoelectric Energy Harvester, ACS Applied Materials & Interfaces, 8 (**2016**), 26190–26197, doi:10.1021/acsami.6b04497