# Supporting Information

**Highly Sensitive Interlocked Piezoresistive Sensors Based on Ultrathin Ordered Nanocone Array Films and Their Sensitivity Simulation**

Yawen Lu, [†, ‡, #] Yin He, [†, ‡, #] Jutao Qiao, [†, ‡] Xin Niu, [†, ‡] Xiaojiu Li, [§] Hao Liu, [†, ‡, *] and Li Liu [∥, *]

[†]School of Textiles Science and Engineering, Tiangong University, Tianjin 300387, China.

[‡]Institute of Smart Wearable Electronic Textiles, Tiangong University, Tianjin 300387, China.

[§]Key Laboratory of Advanced Textile Composite Materials, Ministry of Education, Tiangong University, Tianjin 300387, China.

[∥] Beijing Institute of Fashion Technology, Beijing 100029, China.

*Corresponding authors: liuhao_0760@163.com, fzyll@bift.edu.cn

#These authors contributed equally to this work.

## Section 1 Calculation of theoretical resistance

Piezoresistive mainly on the contact resistance ($R^c$) and the intrinsic resistance ($R^i$) between the two neighboring nanocones of IOCA.

$$R = R^c + R^i \quad (1)$$

The $R^c$ at the contact surface between the interlocked nanocones can be defined by Holm's equation [1]:

$$R^c = \frac{1}{2\mathrm{k}\sqrt{\mathrm{A}/\pi}} \quad (2)$$

where A is the contact area between interlocked nanocones, k is the equivalent conductivity of the two contacting nanocones.

The $R^i$ of PPy nanocones in the composite film can be defined as:

$$R^i = \rho \frac{\mathrm{L}}{\mathrm{s}} \quad (3)$$

where ρ is resistivity of PPy nanocones, L is the height of the interlocked nanocones, s is the equivalent cross-sectional area of the interlocked nanocones.

Since the influence on the resistivity ρ and conductivity k are small under the small load, the change of ρ and k are not considered during the loading process. Longitudinal compression has a great influence on the variation of the longitudinal length (L) of the conductor, while the variation on the lateral cross-sectional area (s) is negligibly small.

Therefore, the relative resistance change can be derived as follows:

$$R_0 = R_0^c + R_0^i \quad (4)$$

$$\frac{\Delta R^c}{R_0^c} = 1 - \sqrt{\frac{A_0}{A}} \quad (5)$$

$$\frac{\Delta R^i}{R_0^i} = \frac{\Delta \mathrm{L}}{L_0} \quad (6)$$

$$\frac{\Delta R}{R_0} = \frac{R_0^c}{R_0} \times \frac{\Delta R^c}{R_0^c} + \frac{R_0^i}{R_0} \times \frac{\Delta R^i}{R_0^i} = \frac{R_0^c}{R_0} \times \left(1 - \sqrt{\frac{A_0}{A}}\right) + \frac{R_0^i}{R_0} \times \frac{\Delta \mathrm{L}}{L_0} \quad (7)$$

$$\frac{R_0^c}{R_0} + \frac{R_0^i}{R_0} = 1 \quad (8)$$

$$\frac{\Delta R}{R_0} = \left(1 - \frac{R_0^i}{R_0}\right) \times \left(1 - \sqrt{\frac{A_0}{A}}\right) + \frac{R_0^i}{R_0} \times \frac{\Delta \mathrm{L}}{L_0} \quad (9)$$

$$\mathrm{S} = \frac{\Delta R/R_0}{P} = \left(1 - \frac{R_0^i}{R_0}\right) \times \frac{\left(1 - \sqrt{A_0/A}\right)}{P} + \frac{R_0^i}{R_0} \times \frac{\Delta \mathrm{L}/L_0}{P} \quad (10)$$

The $R_0$ is the initial resistance of the IOCA, and the $R_0^i$ is equal to the sum of interlocked OCA sheet resistances, which are constant and can be measured by U3402A digital multimeters (Agilent Technologies, USA) (in Table. S1). The resistance change ($\Delta R/R_0$) are affected by contact area (A) and interlocked layers height changes (L). The calculation of theoretical formula were shown in Table. S2.

**Table S1.** Summary of the resistance parameters of different pressure sensors based on planar, 1460 nm, 750 nm and 460 nm sized OCA.

| Structure | | Corresponding Resistance | | | |
|---|---|---|---|---|---|
| | | $r^i$ | $R_0^i$ | $R_0$ | $R_0^i/R_0$ |
| Interlocked | 1460 nm | 2219.092 | 4438.185 | 26996.259 | 0.1644 |
| | 750 nm | 2032.172 | 4064.343 | 14117.204 | 0.2879 |
| | 460 nm | 1703.195 | 3406.389 | 7108.492 | 0.4792 |
| | planar | 1000.000 | 2000.000 | 2043.945 | 0.9785 |
| Single | single | - | 3219.092 | 13497.243 | 0.2385 |

**Table S2.** Theoretical Formula of IOCA based on planar, 1460 nm, 750 nm and 460 nm sized OCA.

| Structure | Theoretical Formula |
|---|---|
| 1460 | $\Delta R/R_0 = 0.8356\left(1-\sqrt{A_0/A}\right)+0.1644\,\Delta L/L_0$ |
| 750 | $\Delta R/R_0 = 0.7121\left(1-\sqrt{A_0/A}\right)+0.2879\,\Delta L/L_0$ |
| 460 | $\Delta R/R_0 = 0.5208\left(1-\sqrt{A_0/A}\right)+0.4792\,\Delta L/L_0$ |
| Planar | $\Delta R/R_0 = 0.0215\left(1-\sqrt{A_0/A}\right)+0.9785\,\Delta L/L_0$ |
| Single | $\Delta R/R_0 = 0.7615\left(1-\sqrt{A_0/A}\right)+0.2385\,\Delta L/L_0$ |

**Section 2 Experiment Supplement**

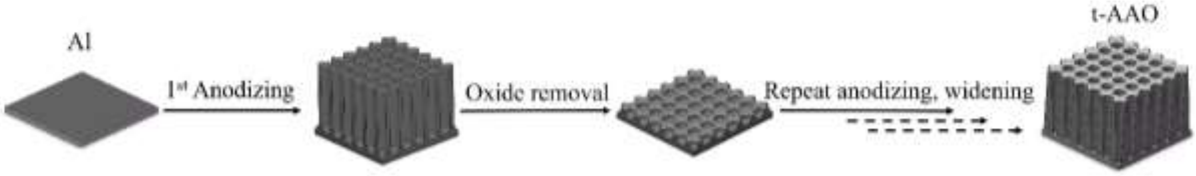



**Figure S1.** Schematic illustration of the preparation of t-AAO.

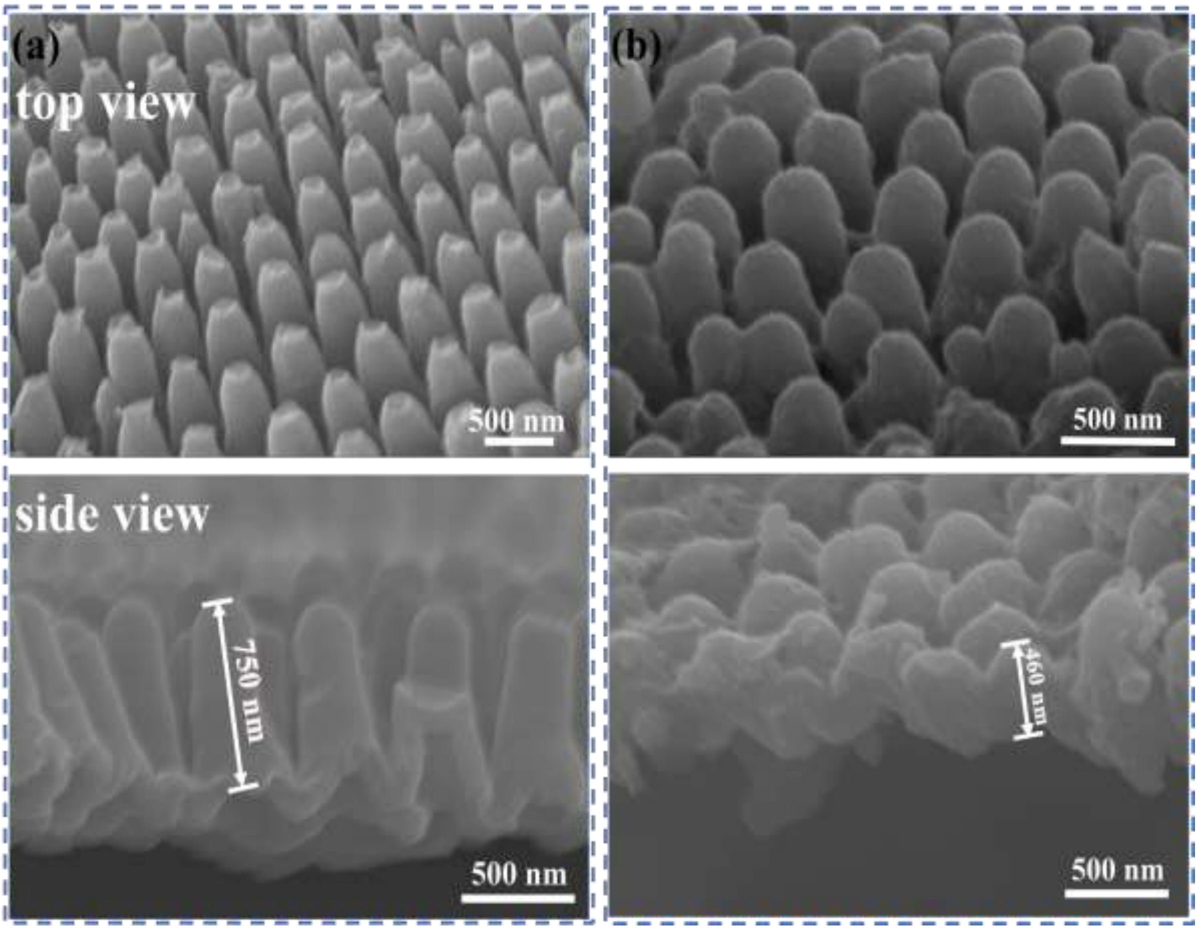
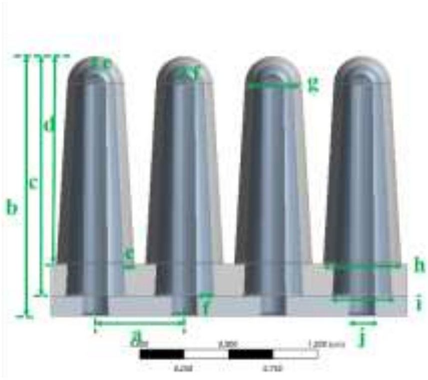
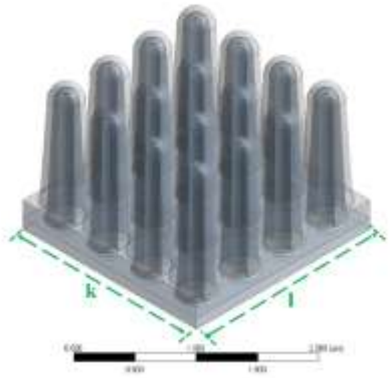
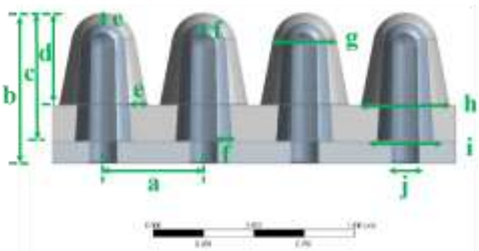
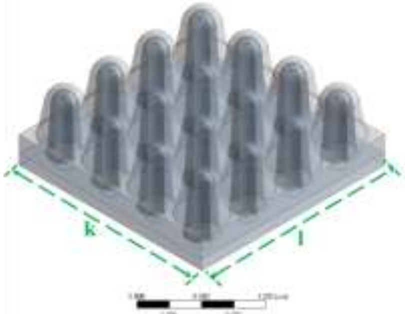
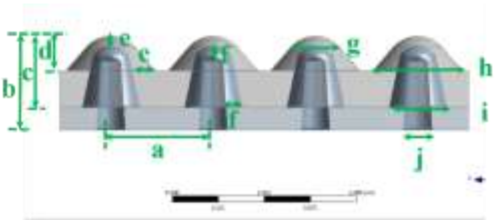
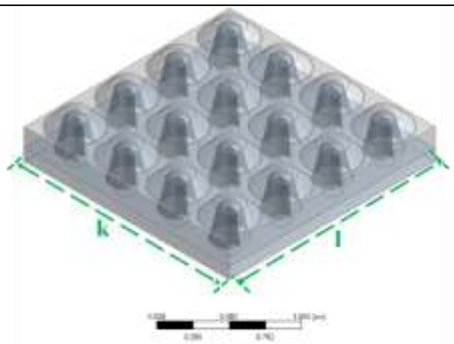
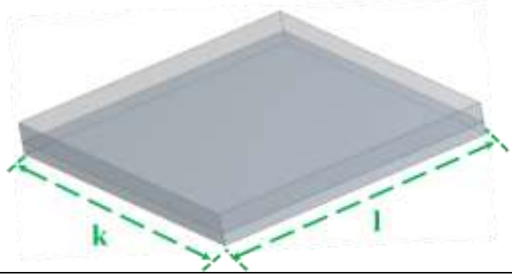


**Figure S2.** Top view and side view FESEM images of OCA with the length of (a) 750 nm, (b) 460 nm.

**Table S3.** Dimensions of four different finite elements geometry structure of the OCA in side view and in 45℃ angle view.

| | Side View | 45℃ Angle View |
|---|---|---|
| OCA-1460 |  |  |
| | a=500nm, b=1460nm, c=1350nm, d=1170nm, e=80nm, f=70nm, g=320nm, h=440nm, i=280nm, j=140nm, k=1cm, l=1cm | |
| OCA-750 |  |  |
| | a=500nm, b=750nm, c=640nm, d=460nm, e=80nm, f=70nm, g=320nm, h=440nm, i=280nm, j=140nm, k=1cm, l=1cm | |
| OCA-460 |  |  |
| | a=500nm, b=460nm, c=350nm, d=170nm, e=80nm, f=70nm, g=320nm, h=440nm, i=280nm, j=140nm, k=1cm, l=1cm | |
| CPF |  |  |
| | m=180nm, n=110nm, k=1cm, l=1cm | |

Note: m is the thickness of PPy, which is equivalent to (c-d); n is the thickness of PMMA, which is equivalent to (b-c).

**Table S4.** Summary of the key parameters of recently reported micro-nano structure sensors.

| Materials | Electrical signal | Pressure sensitivity [kPa$^{-1}$] | Pressure range [kPa] | Minimum pressure detection [Pa] | Ref. in Main |
|---|---|---|---|---|---|
| PVDF/rGO interlocked microdome | Resistance | 31.9 | < 0.02 | 0.6 | [26] |
| ZnO Nanowire/PDMS micropillar | Resistance | 6.8 | < 0.3 | 0.6 | [28] |
| CNT/PDMS | Resistance | 15.1 | < 0.5 | 0.2 | [29] |
| PANI/Au/PDMS micropillars | Current | 2.0 | < 0.22 | 15 | [33] |
| Ag Nanowire/ rose petals | Capacitance | 1.54 | < 1 | 0.6 | [34] |
| Ag Nanowire/PVDF | Capacitance | 54.31 | < 0.5 | 0.1 | [35] |
| Ag Nanowire/PDMS | Capacitance | 1.2 | < 2 | 0.8 | [38] |
| Microstructured Au film/PDMS | Current | 50.17 | < 0.07 | 10.4 | [42] |
| Au Nanowire/ tissue paper | Current | 1.14 | < 5 | 13 | [58] |
| SCNT/PDMS | Current | 1.07 | 20 | - | [59] |
| Polypyrrole hollow-sphere PPy hydrogels | Resistance | 133.1 | < 0.03 | 0.8 | [60] |

**Table S5.** Comparison of the key parameters of recently reported interlocked microstructured sensors.

| Microstructure surface | Size (diameter/ height) | Electrical signal | Sensitivity [kPa$^{-1}$] | Pressure range [kPa] | Ref. in Main |
|---|---|---|---|---|---|
| Microcapsule | 28μm /- | Current | 24.63 | 0.01-0.2 | [11] |
| Nanowire | 50 nm / 1μm | Resistance | 5 | 0.5 | [20] |
| Microdome1 | 10μm / 4μm | Resistance | 31.9 | < 0.02 | [26] |
| Micropillar &Nanowire | 10μm / 10μm | Resistance | 6.8 | < 0.3 | [28] |
| Hemisphere | 3.5μm / 6μm | Resistance | 15.1 | < 0.5 | [29] |
| Microdomain | 18.4 μm / 16.1μm | Current | 50.17 | < 0.07 | [42] |
| Microdome2 | 45μm / 30μm | Resistance | 1.24 | 0.15 | [61] |
| Microdome3 | 2.5μm / 1.5μm | Current | 196 | 10 | [62] |
| Microridge1 | 100μm / 120μm | Triboelectricity | 0.55 | - | [63] |
| Microridge2 | 35μm /30μm | Current | 3.3-10 | 1 | [64] |
| Nanocone | 260-440nm/ 1460 nm | Resistance | 268.36 | 0.2 | this work |

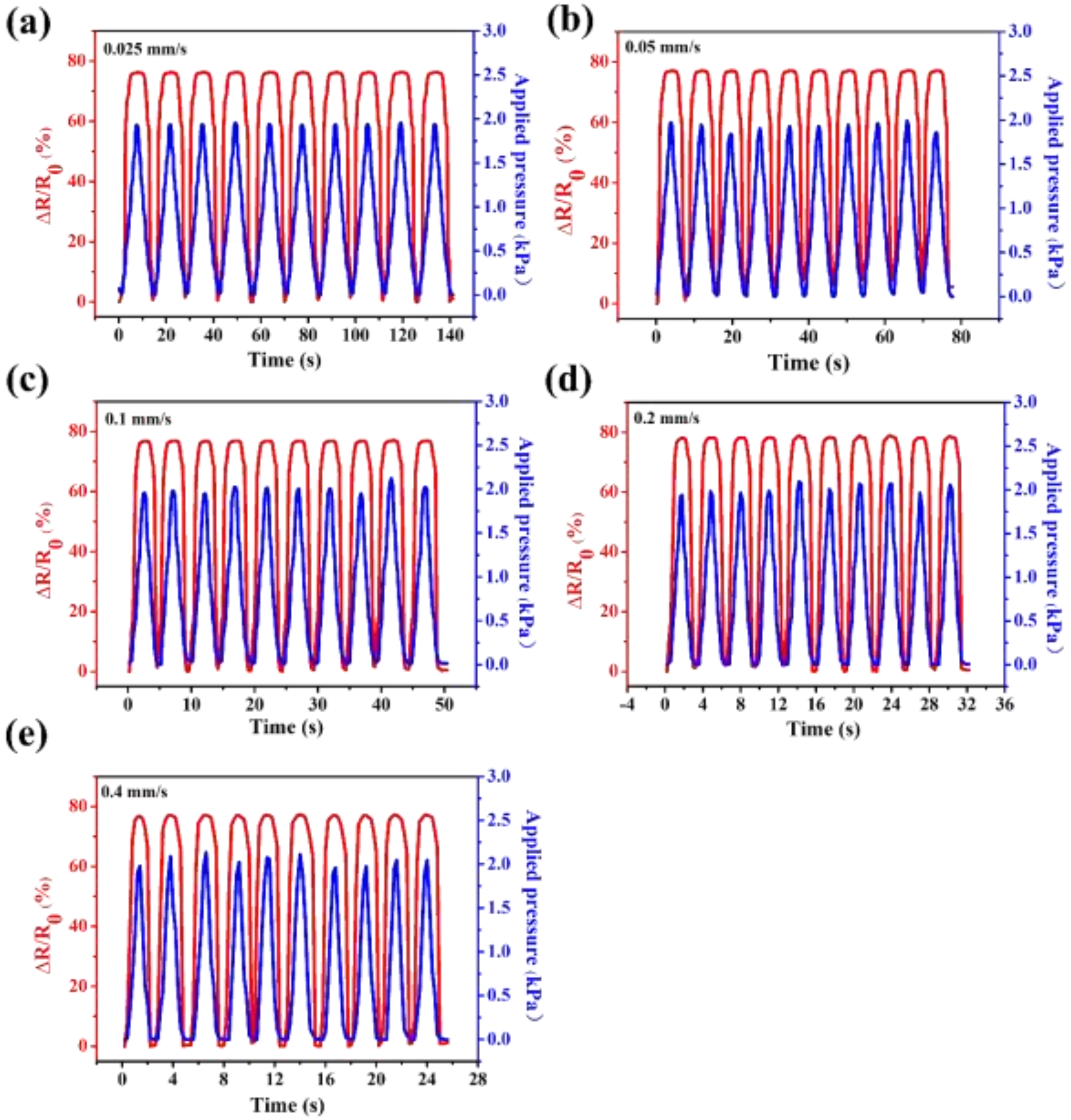



**Figure S3.** Resistance variations at a pressure of 2 kPa with applying speeds of (a) 0.025 mm/s, (b) 0.05 mm/s, (c) 0.1 mm/s, (d) 0.2 mm/s, and (e) 0.4 mm/s.

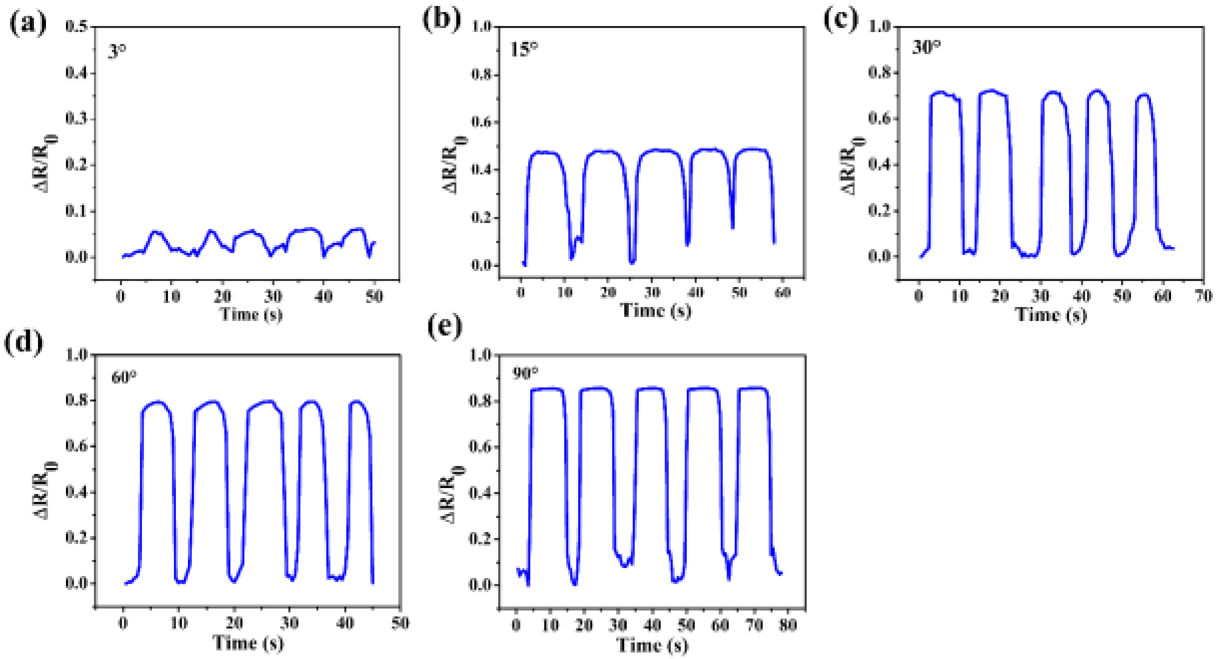



**Figure S4.** Resistance variations of the IOCA with angles of (a) 3°, (b) 15°, (c) 30°, (d) 60°, and (e) 90°.

## References


(1) Wu, Z.; Wang, S.; Zhang, L.; Hu, J. An analytical model and parametric study of electrical contact resistance in proton exchange membrane fuel cells. Journal of Power Sources, **2009**, *189(2)*,1066-1073.