

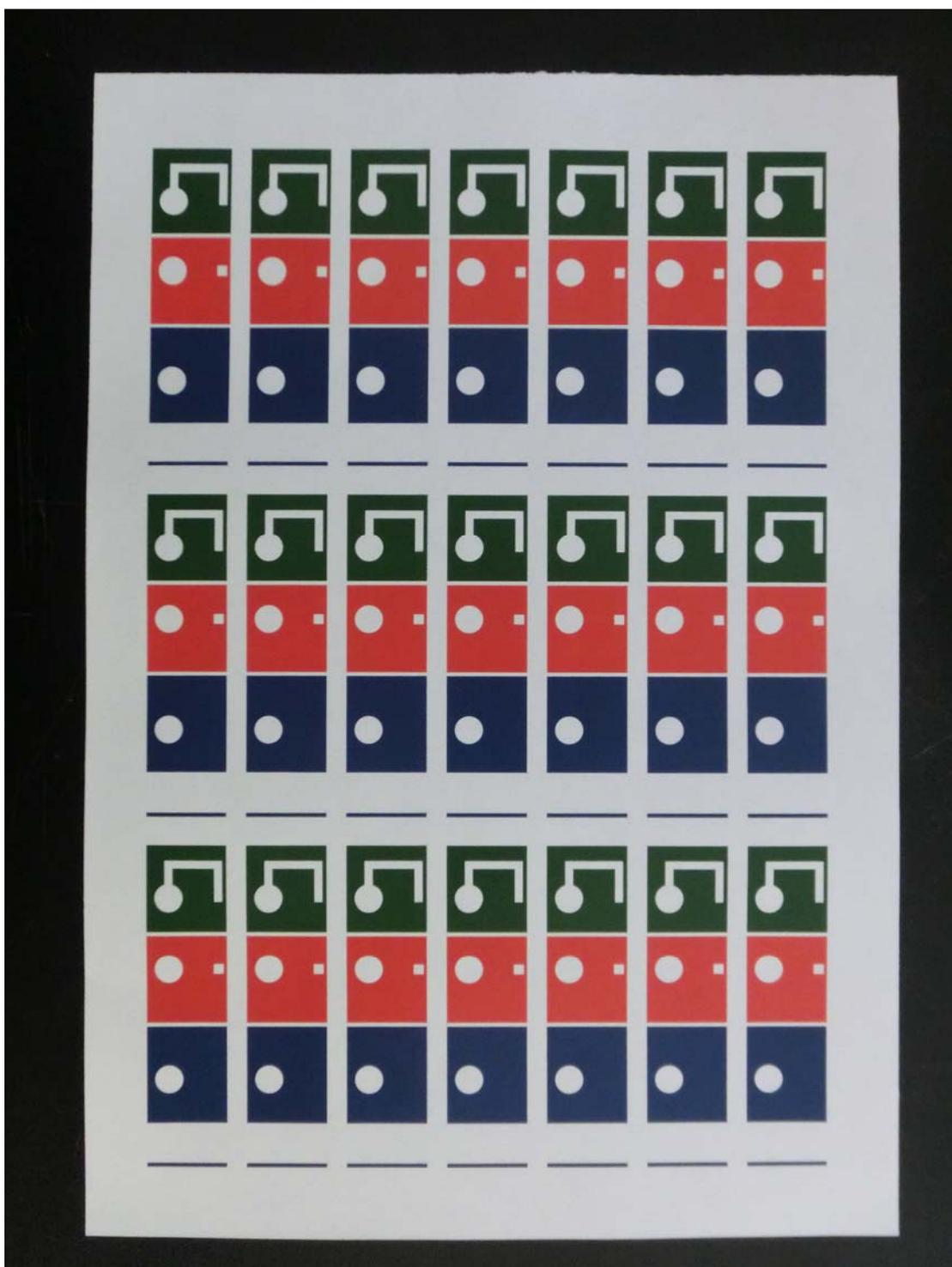
## **Electronic Supplementary Information**

### **Photoelectrochemical Lab-on-Paper Device Equipped with Porous Au-Paper Electrode and Fluidic Delay-Switch for Sensitive Detection of DNA Hybridization**

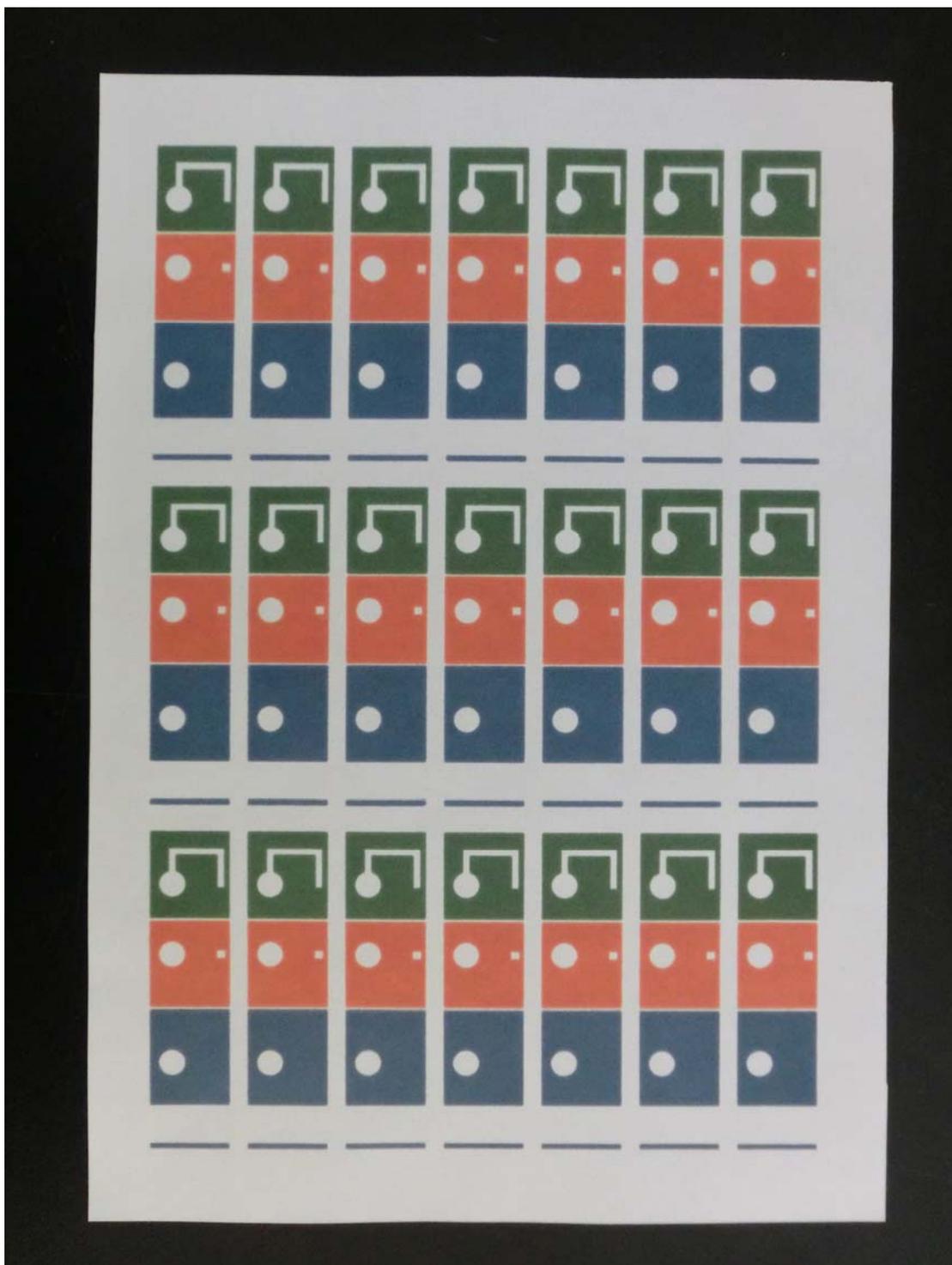
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## Abbreviations

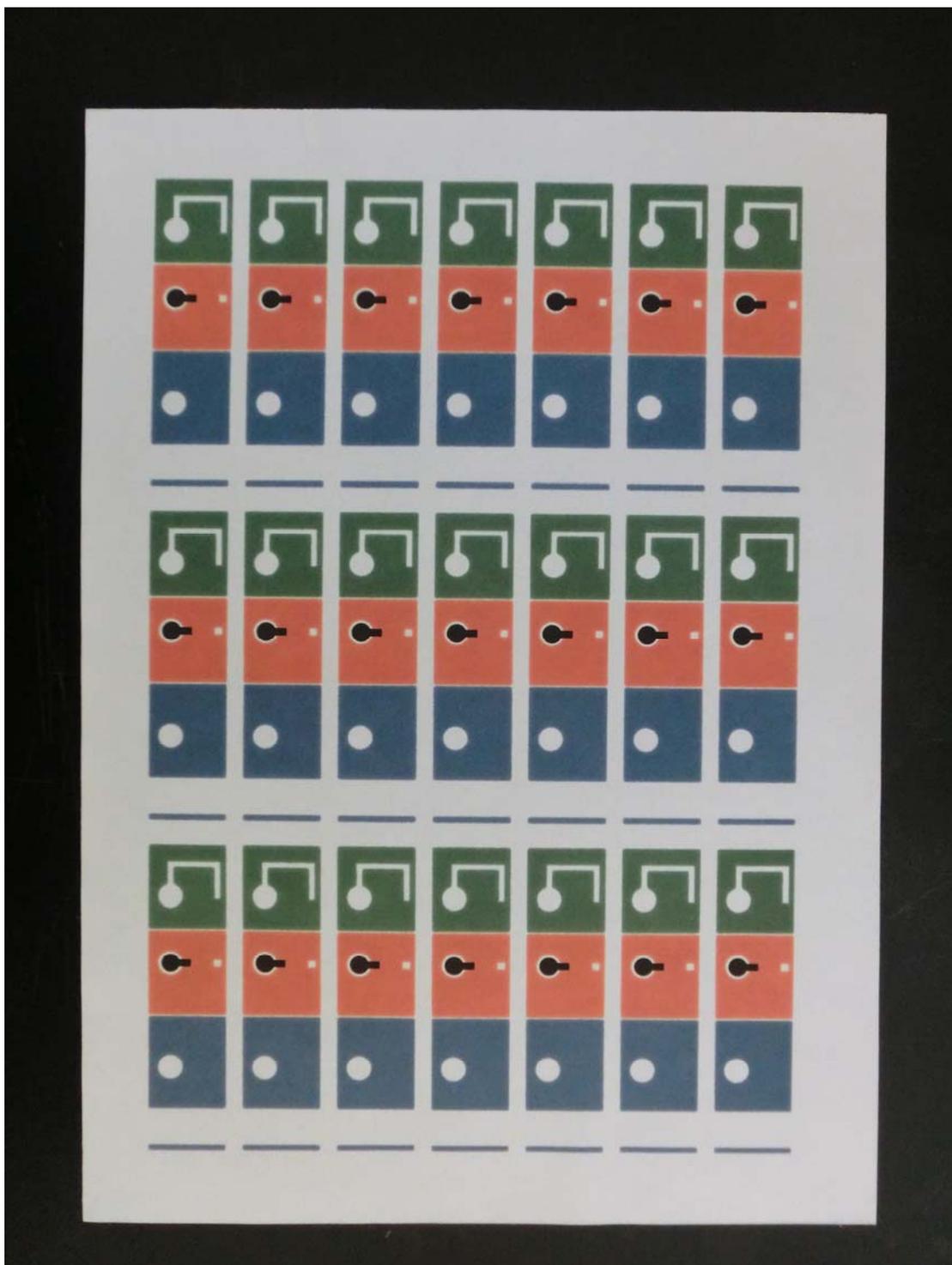
PEC	photoelectrochemical
PS	paper supercapacitor
$\mu$ -PAD	microfluidic paper-based analytical device
$\mu$ -DNA-PECPD	microfluidic DNA-based PEC paper device
DMM	digital multi-meter
FDS	fluidic delay-switch
PBS	phosphate buffer solution
Au-PWE	Au-paper working electrode
CL	chemiluminescence
ECL	electrogenerated chemiluminescence
ABEI	<i>N</i> -(aminobutyl)- <i>N</i> -(ethylisoluminol)
AuNPs	gold nanoparticles
PIP	<i>p</i> -iodophenol
LED	light-emitting diode
CdS NPs	cadmium sulfide nanoparticles
PVA	Polyvinyl alcohol
PDDA	poly(dimethyldiallylammonium chloride)
MWCNTs	multi-walled carbon nanotubes
PDDA-GR	PDDA-functionalized graphene
BSA	bovine serum albumin
NHS	<i>N</i> -Hydroxysuccinimide
EDC	1-Ethyl-3-(3-dimethylaminopropyl) carbodiimide hydrochloride
PWE	paper working electrode
SEM	scanning electron microscopy
EIS	electrochemical impedance spectroscopy
CB	conduction-band
VB	valence-band
AC	areal capacitance
HPLC	high performance liquid chromatography
RSD	relative standard deviation



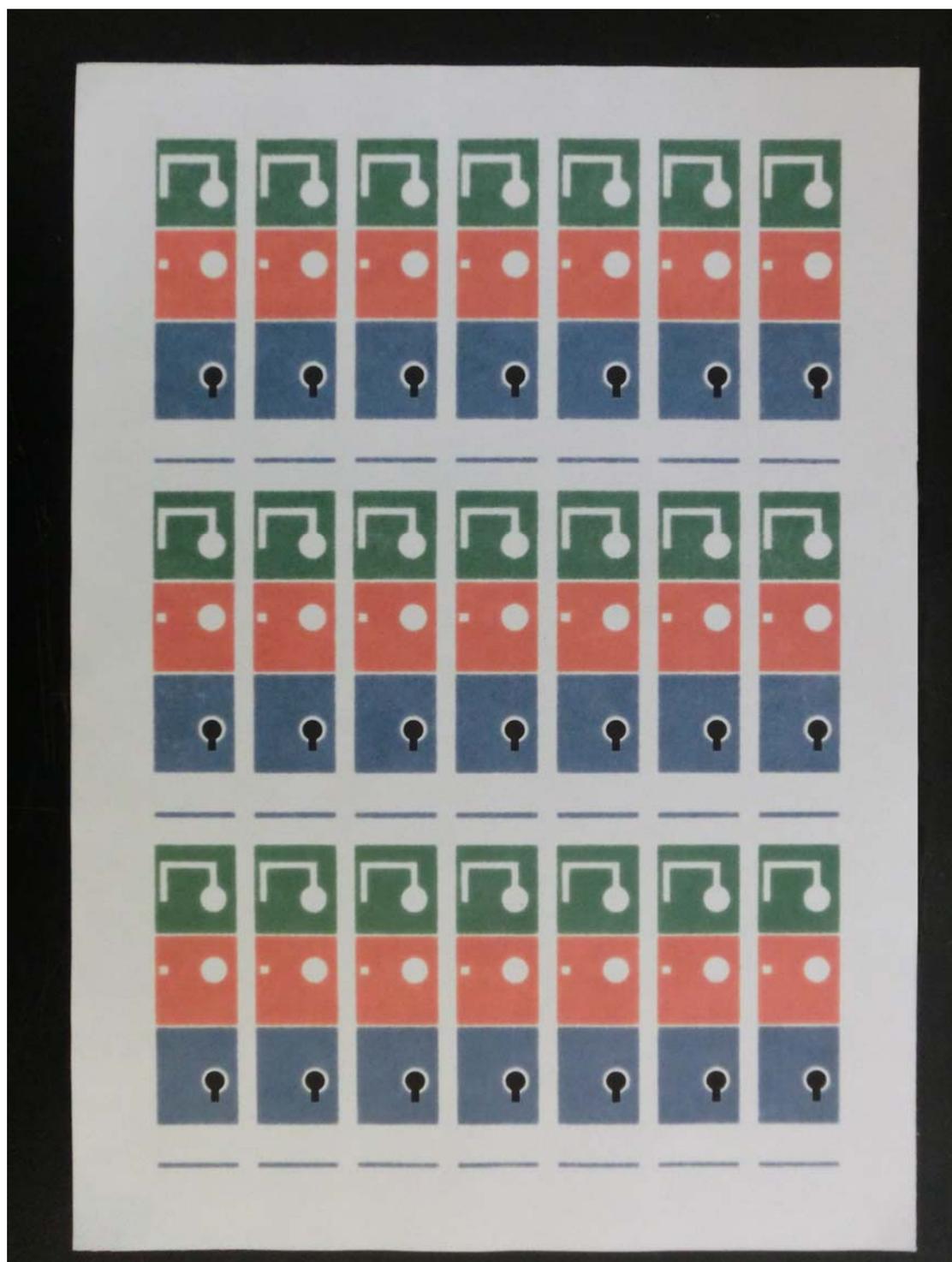
**Figure S1.** Wax-printed  $\mu$ -DNA-PECPDs on a paper sheet (A4) before baking.



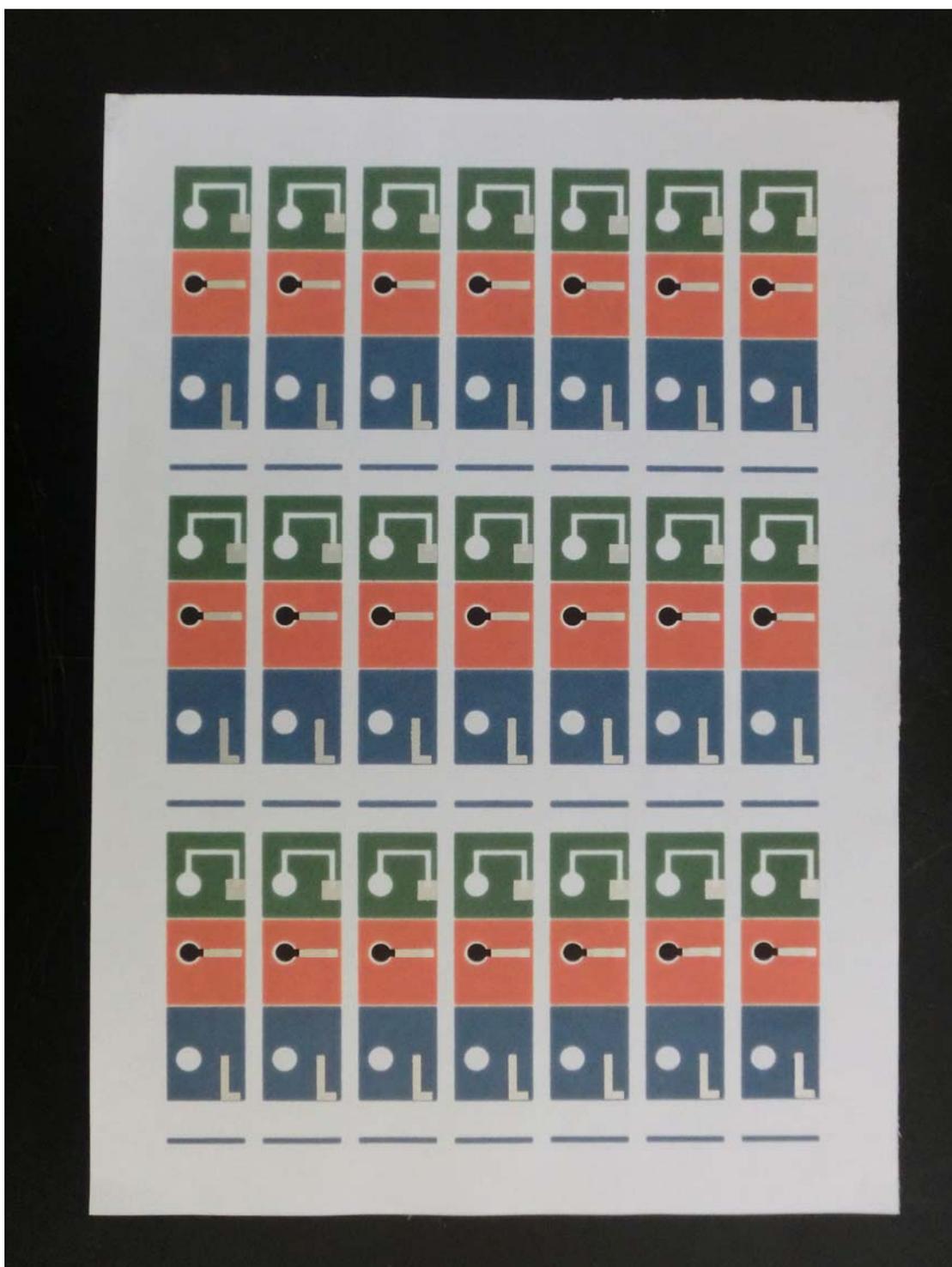
**Figure S2.** Wax-printed  $\mu$ -DNA-PECPDs on a paper sheet (A4) after baking.



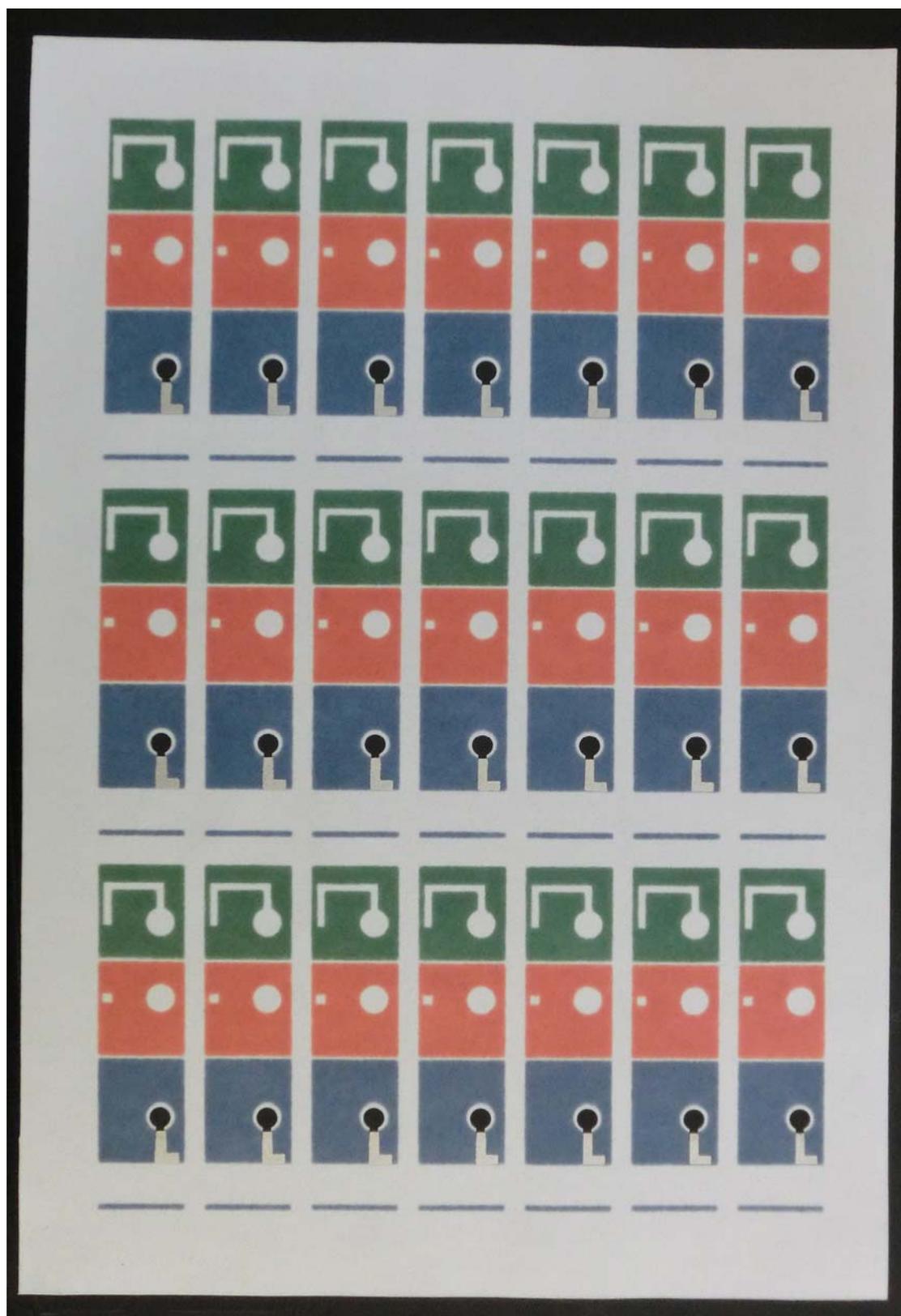
**Figure S3.**  $\mu$ -DNA-PECPDs on a paper sheet (A4) after screen-printing of carbon working electrodes.



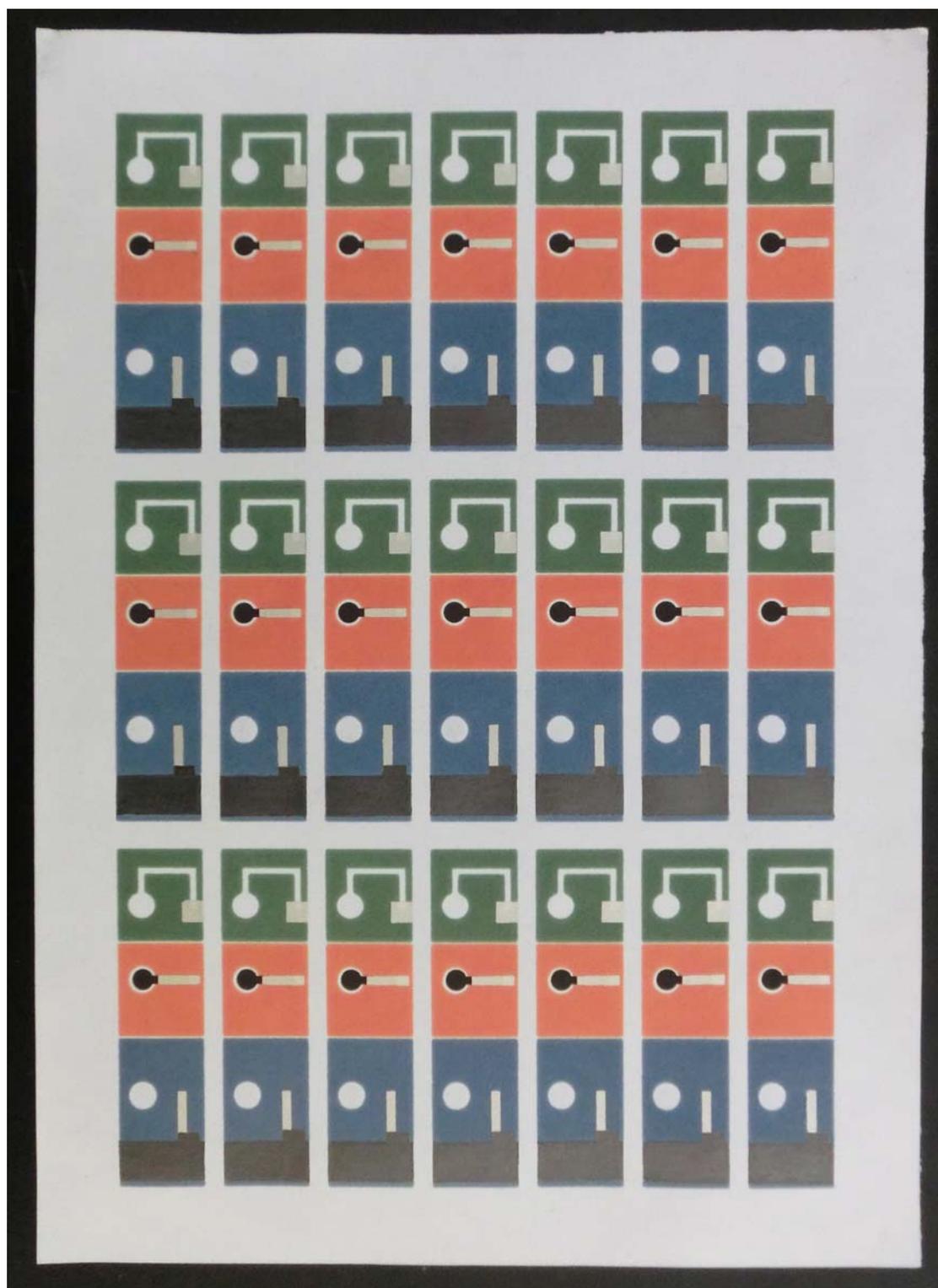
**Figure S4.**  $\mu$ -DNA-PECPDs on a paper sheet (A4) after screen-printing of carbon counter electrodes. (The reverse sides of Figure S3)



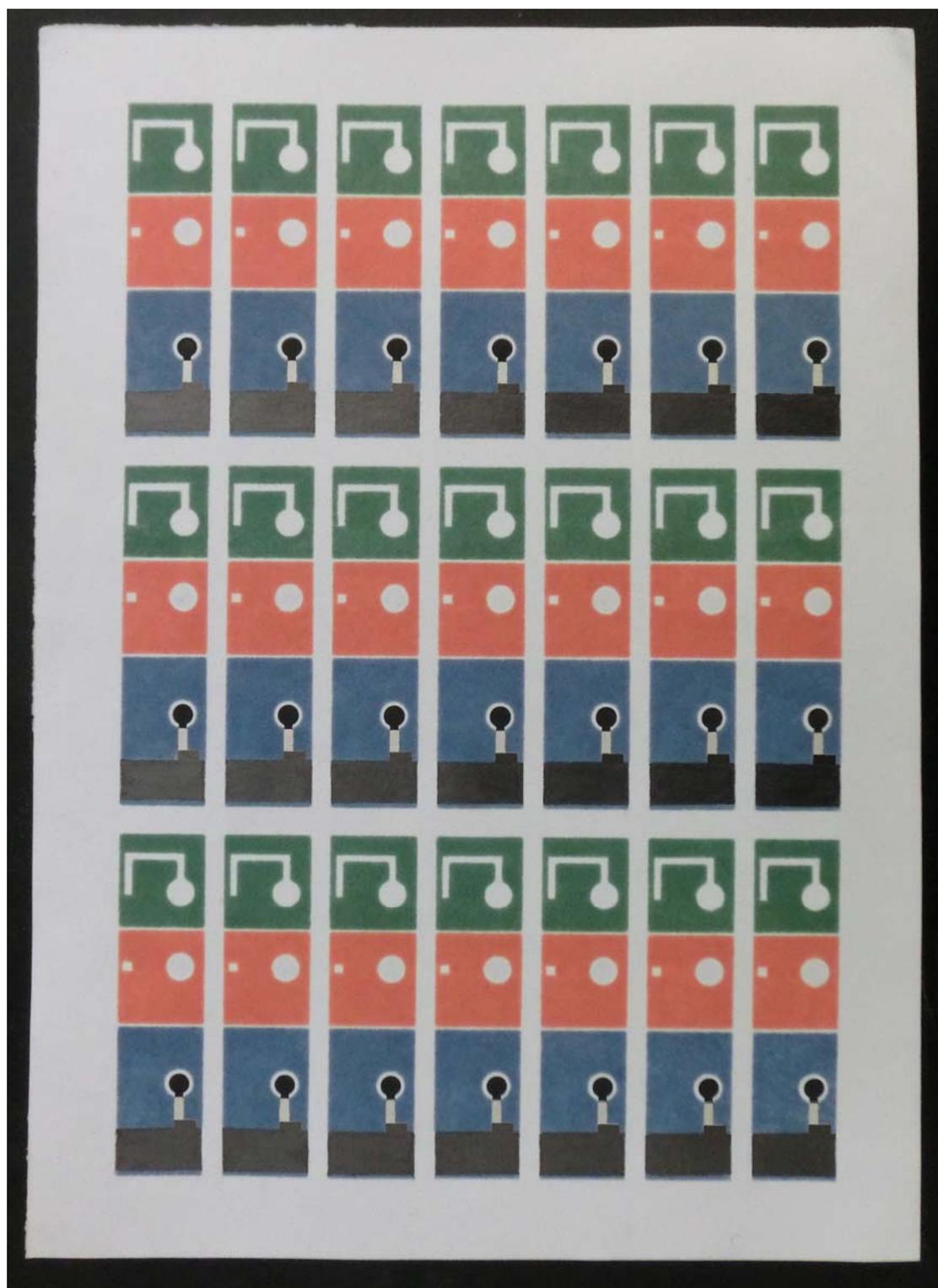
**Figure S5.**  $\mu$ -DNA-PECPDs on a paper sheet (A4) after screen-printing of silver wire and silver pad.



**Figure S6.**  $\mu$ -DNA-PECPDs on a paper sheet (A4) after screen-printing of silver wire. (The reverse sides of Figure S5)



**Figure S7.**  $\mu$ -DNA-PECPDs on a paper sheet (A4) after drawing of graphite electrodes.



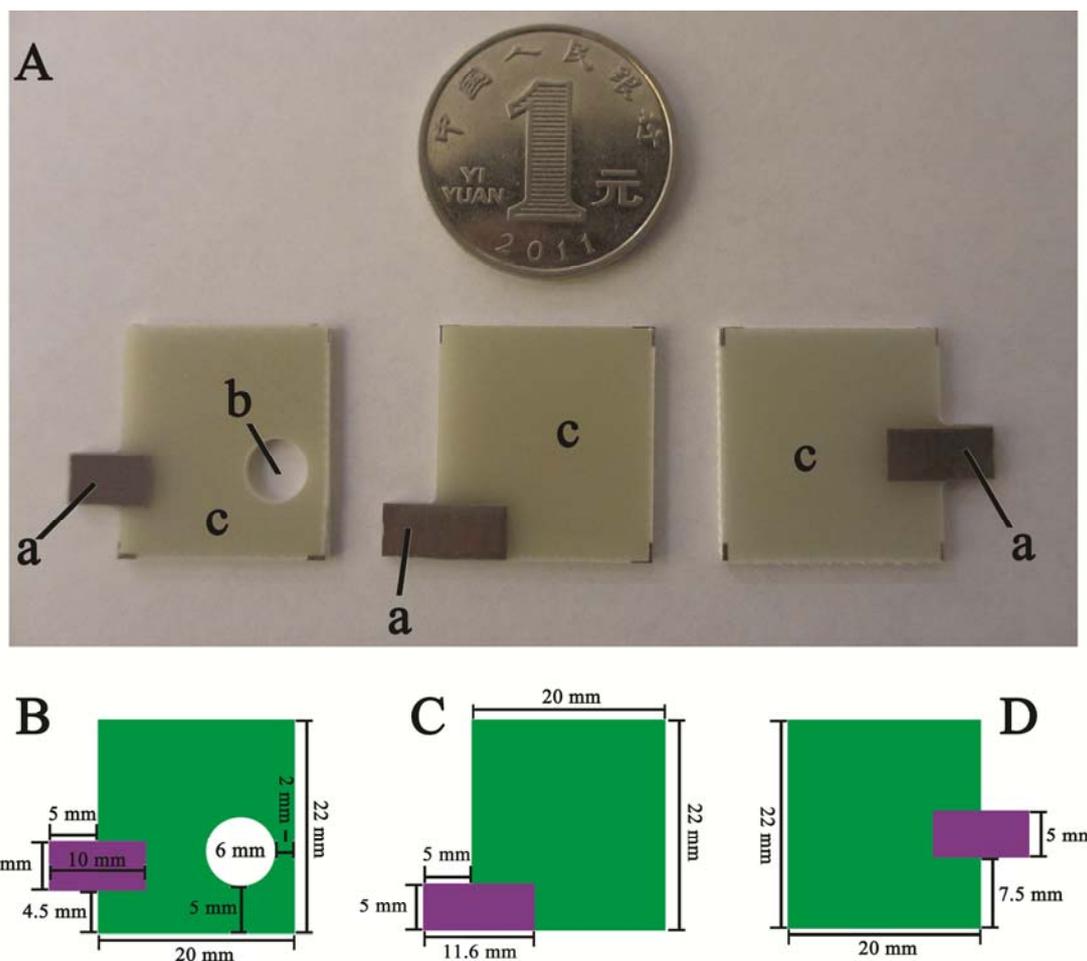
**Figure S8.**  $\mu$ -DNA-PECPDs on a paper sheet (A4) after drawing of graphite electrodes. (The reverse sides of Figure S7)

### **Preparation of AuNPs attached MWCNTs**

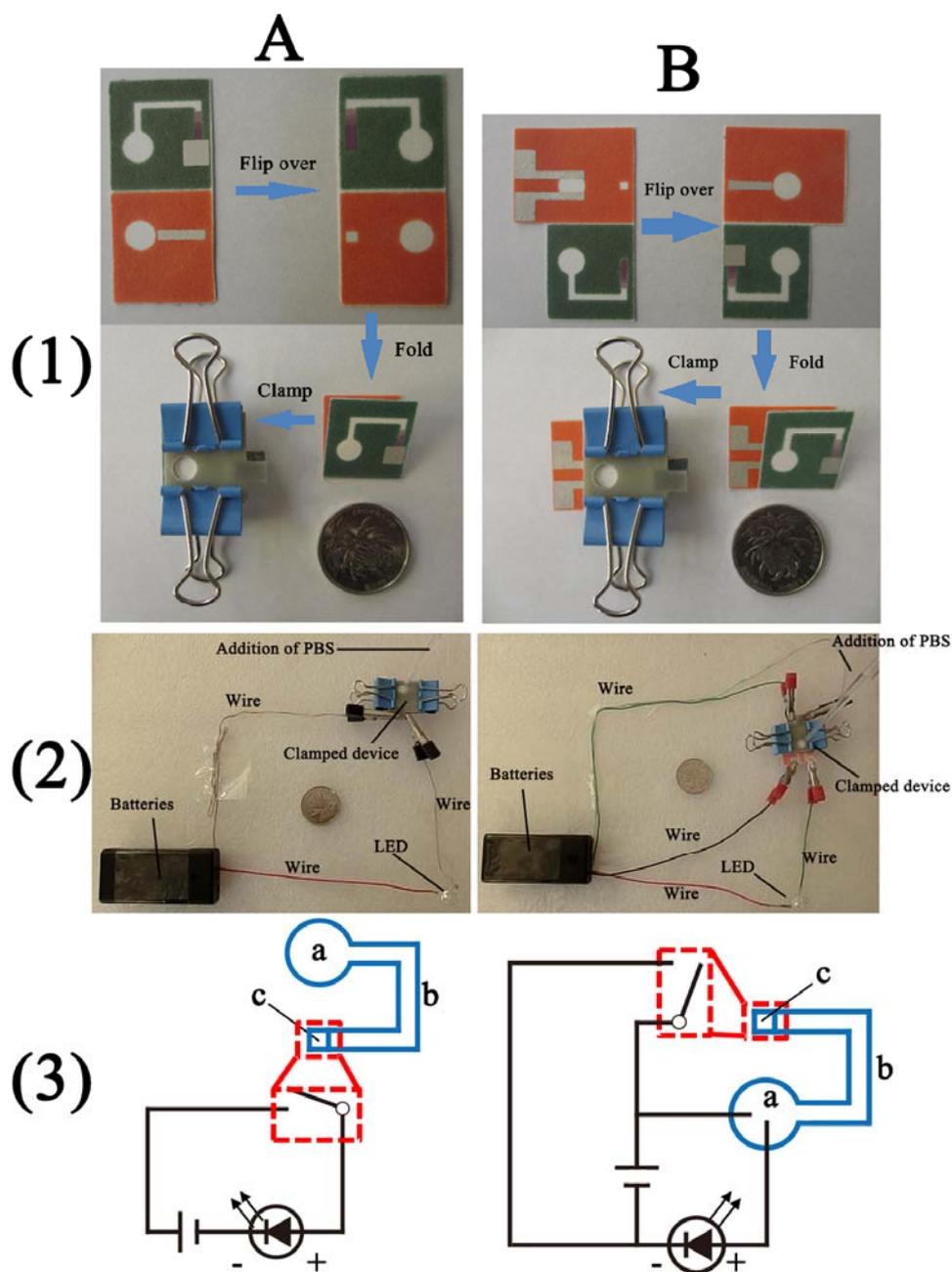
The as-received MWCNTs were first treated via sonication in 1:3 concentrated nitric-sulfuric acids at ca. 50 °C. Such a procedure could shorten the nanotubes, removed metallic and carbonaceous impurities, and generated carboxylate groups on the CNTs surface <sup>1</sup>. Then the shortened MWCNTs were functionalized with PDDA imitating previous literature <sup>2</sup>. Briefly, 5 mL shortened MWCNTs dispersion (1.0 mg/mL) was mixed with 10 mL PDDA aqueous solution (2.0 %) and sonicated for 30 min to give a homogeneous suspension. After centrifugation under 20,000 rpm, the complex was washed with water for at least three times. Then, the PDDA functionalized MWCNTs were dispersed in 10.0 mL of as-prepared colloidal AuNPs and stirred for 30 min. After centrifugation, light purple AuNPs/MWCNTs composites were obtained, which were further washed with water and redispersed in 5.0 mL of water.

### **Preparation of PDDA-GR**

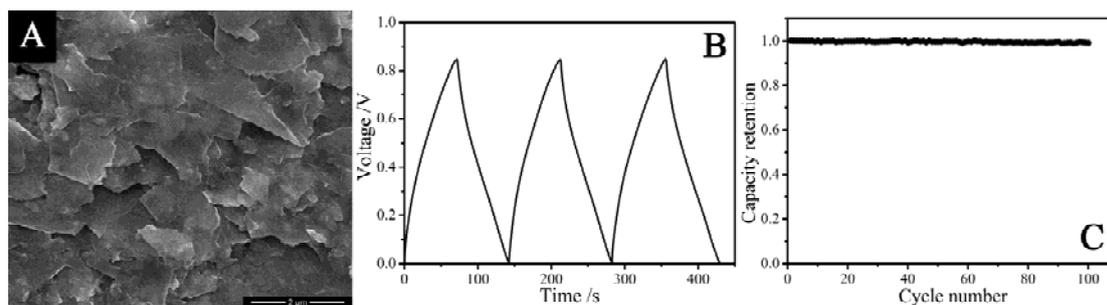
GR was functionalized with PDDA according to previous literature <sup>2</sup>. 5 mL GR dispersion (0.5 mg/mL) was mixed with 10 mL PDDA aqueous solution (1.0%) and sonicated for 30 min to give a homogeneous suspension. After centrifugation under 20,000 rpm, the complex was washed with water for at least three times and redispersed in 5 mL water with mild sonication.



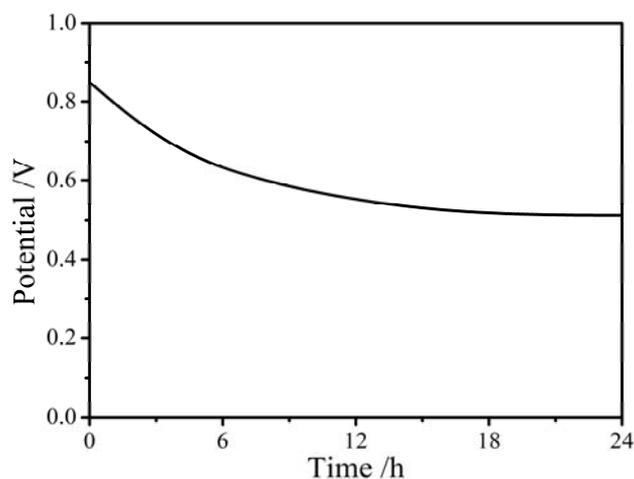
**Figure S9.** (A) The picture of the three circuit boards used in this work: (a) The conductive connector, (b) The hole for the addition of solution, (c) The insulating board. (B, C, D) The shape and size of the three circuit boards used in this work.



**Figure S10.** (A) Paper device and circuit for the demonstration of the function of fluidic switch; (B) Paper device and circuit for the determination of the delay time and delay reproducibility of the FDS; (1) the designed paper device, (2) the designed circuit, (3) circuit diagram: (a) paper initiation zone, (b) delay channel, (c) fluidic switch.



**Figure S11.** (A) SEM image of the resulted graphitic film electrode on paper; (B) Galvanostatic charge-discharge curves of this paper supercapacitor; (C) Durability test of this paper supercapacitor by measuring 100 charge-discharge cycles.



**Figure S12.** Self-discharge curve for the all-solid-state paper supercapacitor.

**Table S1** Comparison of paper-based biosensors for DNA detection

Device	Cost		Stability	Detection Limit	Reference
	Device	External equipment			
Paper-based electrochemical DNA sensor	~<\$0.1	Electrochemistry workstation (~\$7000-9000)	30 days at 4 °C	0.8 aM	9 Our previous work
Lateral flow colorimetric DNA biosensor	~<\$0.5	---	Not Mentioned	0.01 fM	10
Paper-based fluorescent DNA biosensor	~<\$0.2	Fluorescence scanner (~\$50000-70000)	Not Mentioned	80 aM	11
Paper-based colorimetric DNA biosensor	~<\$0.2	---	One month at 27-33 °C	10 fM	12
Paper-based fluorescent DNA strip	~<\$0.1	Fluorescence Imager (~\$30000-50000)	Not Mentioned	100 pM	13
Paper-based colorimetric mRNA biosensor	~<\$0.3	---	Not Mentioned	10 nM	14
Paper-based fluorescent DNA detection	~<\$0.1	Fluorescence Laser Scanner (~\$50000-80000)	Not Mentioned	8 nM	15
Paper-based chemiluminescence DNA biosensor	~<\$0.2	Luminescence analyzer (~\$5000-10000)	6 weeks at 4 °C	0.85 aM	16 Our previous work
Paper-based colorimetric DNA biosensor	~<\$0.2	Document scanner or cell phone camera (~\$200-500)	Not Mentioned	250 pg	17
Lateral flow colorimetric DNA biosensor	~<\$0.2	---	At room temperature for days	0.16 pM	18
Paper-based photoelectrochemical DNA biosensor	~<\$0.2	Digital multi-meter (~\$300-450)	2 weeks at 4 °C	15 fM	This work

## Reference

- 1 M. Zhang, Y. Yan, K. Gong, L. Mao, Z. Guo and Y. Chen, *Langmuir*, 2004, **20**, 8781-8785.
- 2 R. Cui, H. Huang, Z. Yin, D. Gao and J.-J. Zhu, *Biosens. Bioelectron.*, 2008, **23**, 1666-1673.