Supporting Information for

**Tailoring the Meso-Structure of Gold Nanoparticles in Keratins-Based Activated Carbon toward High-Performance Flexible Sensor**

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**S1 Materials and Methods**

**S1.1 Chemical Reagents and Apparatus**

In exception to wool fibers, all other chemicals are of analytical grade and used as received without further purification. Merino 64’s wool fibers were bought from Tongxiang Dushi Woolen Material Co., Ltd. Chloroauric acid tetrahydrate (HAuCl\(_4\)·4H\(_2\)O, ≥47.8%), sodium bicarbonate (NaHCO\(_3\), ≥99.5%), and ethanol (C\(_2\)H\(_6\)O, ≥99.7%) were from Sinopharm Chemical Reagent. Sodium dodecylsulfate (SDS, ≥99%), sodium sulfide nonahydrate (Na\(_2\)S·9H\(_2\)O, ≥98.0%), urea (CH\(_4\)N\(_2\)O, ≥99.0%), citric acid monohydrate (C\(_6\)H\(_8\)O\(_7\)·H\(_2\)O, ≥99.5%), disodium hydrogen phosphate dodecahydrate (Na\(_2\)HPO\(_4\)·12H\(_2\)O, ≥99.0%), sodium chloride (NaCl, ≥99.5%) and sodium hydroxide (NaOH, ≥96.0%) were obtained from Xilong Chemical Industry. Uric acid (C\(_4\)H\(_4\)N\(_4\)O\(_3\), 99%) was obtained from Alfa Aesar. D- (+)-Glucose (C\(_6\)H\(_12\)O\(_6\), batch no. C10100819, 99%), L-(+)-lactic acid (C\(_3\)H\(_6\)O\(_3\), batch no. C10101039, 90%) and Poly(vinyl butyral) (PVB) (M.W90000-120000) was obtained from Shanghai Macklin Biochemical Co. Ltd. Nafion PFSA polymer (Dispersion D520 (5%)) was obtained from OU point Ltd. The water used in all experiments was ultrapure (the resistivity >18 MΩ cm\(^{-1}\)).

**S1.2 Preparation of Regenerated WK Solution**

The raw wool fibers were treated to extract WK via reduction method\(^1\). Accurately weighted 12.5 g pre-treated wool fibers were added in a 250 mL aqueous solution containing urea (4 M),
Na₂S (0.1 M), and SDS (0.02 M). The above mixture was stirred at 50 °C for 12 h with subsequent filtration. The filtrated stock was dialyzed against ultrapure water (18 MΩ) using a dialysis bag (Solarbio, molecular cut-off about 3500 Da) for 3 d. The dialyzed solution was concentrated at 60 °C for 8 h, and the concentration of WK solutions was 5 wt% on average. The obtained WK solutions were stored at 4 °C before further usage.

**S1.3 Preparation of AuNCs@WK**

In a typical experiment, 5 mL aqueous solution of HAuCl₄ (10×10⁻³ M) was added to 5 mL WK solution (2.5 wt%) under vigorous stirring. The pH of reaction mass is adjusted by addition of a 0.5 mL NaOH solution (1 M) 10⁻¹₂, which was then retained at 45 °C for 12 h. The acquired AuNCs@WK were preserved at 4 °C for future application.

**S1.4 Preparation of AuNCs@WK Composite Sponges**

The composite mass is subjected to the freeze-drying method to sponge formation. The obtained suspension of AuNCs@WK (without dialysis) was transferred into a mold and frozen at −20 °C for 24 h. Followed to this the hybrid solidified mass was dried by a freeze-dryer (-106 °C) for 48 h to fabricate 3D porous composite sponges.

**S1.5 Preparation of AuNPs@NPWC**

The solid mass of AuNCs@WK is heated initially to 150 °C for 60 min in a tube furnace with N₂ atmosphere to take out water content, followed by heating to 350 °C (2 °C min⁻¹) for 60 min to develop the incipient conjugated carbon skeleton. It transformed into the polyaromatic N-doped porous carbon matrix with temperature rise from 350 to 700 °C at a rate of 2 °C min⁻¹ and calcined for 2 h. The carbonized material was washed several times with DI water to remove salt residues. The black solid composite material was dried at 60 °C overnight.

**S1.6 Characterization of AuNPs/NPWC**

The crystallographic phase of the as-carbonized composite materials was characterized by X-ray diffraction (XRD, Bruker D8 AVANCE). The surface morphology, porosity and distribution of AuNPs in the composite materials were examined with a field emission gun scanning electron microscope (FEGSEM, Hitachi SU70) with an accelerating voltage of 5 kV. The carbon matrix, size and morphology of nanoparticles were characterized by transmission electron microscopy (TEM) through a JEM-2100F (JEOL, Japan). Nitrogen adsorption/desorption isotherms at 77.3 K were obtained using a Micromeritics TriStar II 3020 static volumetric analyzer. Prior to adsorption measurements, the samples were degassed for 12 h at 100 °C ensuring that the residual pressure fell below 10 mbar. The Brunauer-Emmett-Teller surface area was calculated within the relative pressure range 0.05 to 0.2. Total volume was calculated at P/P₀ = 0.99. The surface element state and the constituent of N-functional groups were detected by X-ray photoelectron spectroscopy (Quantum 2000, PHI, USA), whereas, Raman spectra were taken using (HORIBA LabRAM HR Evolution) with a 532 nm excitation laser. Commercial handy UA meter of Sinocare made was used for UA comparative study, whereas pH comparison study has been performed by commercial pH meter of Thermo scientific made.
S2 Supplementary Tables and Figures

Table S1 Comparison of the elemental compositions of the carbonized materials carbonized at different carbonization temperature (C, H, N elemental analysis)

| Material     | C%    | H%    | N%    |
|--------------|-------|-------|-------|
| AuNPs@NPWC-500 | 47.89 | 1.602 | 9.281 |
| AuNPs@NPWC-600 | 50.77 | 1.344 | 9.418 |
| AuNPs@NPWC-700 | 51.95 | 2.055 | 8.385 |
| AuNPs@NPWC-800 | 57.23 | 3.385 | 4.248 |

Table S2 Effect of carbonization temperature on $S_{BET}$ area of the carbonized materials

| Material     | BET surface area m² g⁻¹ |
|--------------|--------------------------|
| AuNPs@NPWC-400 | 27.0942                  |
| AuNPs@NPWC-500 | 381.2189                 |
| AuNPs@NPWC-600 | 452.6490                 |
| AuNPs@NPWC-700 | 625.0586                 |
| AuNPs@NPWC-800 | 447.6388                 |

Fig. S1 (a) N₂ adsorption–desorption isotherms. (b) Effect of carbonization temperature on the BET surface area. (c) Pore size distributions
Fig. S2 Survey XPS data of (a) AuNPs@NPWC-400, (b) AuNPs@NPWC-500, (c) AuNPs@NPWC-600, (d) AuNPs@NPWC-700 and (e) AuNPs@NPWC-800
**Fig. S3** High resolution N 1s XPS data of (a) AuNPs@NPWC-400, (b) AuNPs@NPWC-500, (c) AuNPs@NPWC-600, (d) AuNPs@NPWC-700 and (e) AuNPs@NPWC-800
Fig. S4 High resolution C 1s XPS data of (a) AuNPs@NPWC-400, (b) AuNPs@NPWC-500, (c) AuNPs@NPWC-600, (d) AuNPs@NPWC-700 and (e) AuNPs@NPWC-800
Fig. S5 High resolution O 1s XPS data of (a) AuNPs@NPWC-400, (b) AuNPs@NPWC-500, (c) AuNPs@NPWC-600, (d) AuNPs@NPWC-700 and (e) AuNPs@NPWC-800
**Fig. S6** XPS core level spectra of Au 4f of (a) AuNPs@NPWC-400, (b) AuNPs@NPWC-500, (c) AuNPs@NPWC-600, (d) AuNPs@NPWC-700 and (e) AuNPs@NPWC-800

**Table S3** Comparison of the elemental compositions of the carbonized material based on XPS analysis

| Material          | Carbon % | Nitrogen % | Oxygen % | Au %  |
|-------------------|----------|------------|----------|-------|
| AuNPs@NPWC-400    | 76.7     | 9.53       | 12.97    | 0.8   |
| AuNPs@NPWC-500    | 80.5     | 7.96       | 10.45    | 1.08  |
| AuNPs@NPWC-600    | 82.33    | 7.63       | 9.14     | 0.9   |
| AuNPs@NPWC-700    | 85.09    | 4.89       | 9.04     | 0.98  |
| AuNPs@NPWC-800    | 86.21    | 4.27       | 9.17     | 0.34  |
Fig. S7 Sensitivity comparison of carbonized composite materials (A) AuNPs@NPWC-800, (B) AuNPs@NPWC-700, (C) AuNPs@NPWC-600, (D) AuNPs@NPWC-500 and (E) AuNPs@NPWC-400

Fig. S8 Sensitivity comparison of AuNPs@NPWC-500 and NPWC-500 using McIlvaine’s buffer pH 3 to 5

Fig. S9 Reproducibility study of AuNPs@NPWC-500 modified electrode for pH sensing
**Fig. S10** Calibration curve showing pH Vs potential (mV) using AuNPs@NPWC-500 modified electrode (for identifications of real sample pH)

**Fig. S11** Stability test of AuNPs@NPWC-500 electrode using McIlvaine’s buffer pH 5

**Table S4** Comparison of pH value detected by commercial pH meter and AuNPs@NPWC-500 electrode

| Sample            | pH sensor (This work) | pH meter |
|-------------------|-----------------------|----------|
| Sweat Sample 1    | 5.53                  | 5.56     |
| Sweat Sample 2    | 5.73                  | 5.77     |
| Sweat Sample 3    | 5.51                  | 5.55     |
| Sweat Sample 4    | 5.81                  | 5.78     |
| Sweat Sample 5    | 5.57                  | 5.60     |
| Urine Sample 1    | 5.65                  | 5.68     |
| Urine Sample 2    | 5.80                  | 5.72     |
| Urine Sample 3    | 5.70                  | 5.69     |
| Urine Sample 4    | 5.74                  | 5.62     |
| Urine Sample 5    | 5.49                  | 5.51     |
Table S5 Comparison of different electrochemical sensor proposed for the determination of UA

| Electrode material         | Linear range (µM) UA | Detection limit (µM) UA | References |
|---------------------------|----------------------|-------------------------|------------|
| AuNPs-β-CD-Gra            | 0.5-60               | 0.21                    | S2         |
| Au-RGO                    | 8.8-53               | 1.8                     | S3         |
| PdAg-NFs/RGO              | 1-150                | 0.081                   | S4         |
| Fe3O4@Au-GA               | 1-300                | 0.05                    | S5         |
| 3DHA-AuNPs*               | 1-60                 | 0.005                   | S6         |
| RGO-ZnO                   | 3-330                | 1.08                    | S7         |
| AuNPs@NPWC-700            | 5-150                | 0.1                     | This work  |

*GA: graphene, RGO: reduced graphene oxide; NFs: nanofibers; 3DHA-AuNPs: 3D-Graphene hydrogel–gold nanoparticles; RGO: reduced graphene oxide

Fig. S12 Stability taste of AuNPs@NPWC-700 modified sensor for the determination of 1 mM UA

Fig. S13 SEM images of AuNPs@NPWC-700 catalyst before (left) and after (right) 100 cycles of CV in 0.1 M PBS (pH = 7.0) containing 0.1 mM UA
**Fig. S14** Selectivity of AuNPs@NPWC-700 towards UA detection

**Table S6** Comparison of UA value detected by commercial UA meter and AuNPs@NPWC-700 electrode

| Sample          | UA sensor (µM L⁻¹) | Commercial UA meter (µM L⁻¹) |
|-----------------|-------------------|------------------------------|
| Urine Sample 1  | 4240              | 4120                         |
| Urine Sample 2  | 3570              | 3530                         |
| Urine Sample 3  | 4678              | 4630                         |
| Urine Sample 4  | 5034              | 5140                         |
| Urine Sample 5  | 4787              | 4480                         |
| Urine Sample 6  | 4787              | 4800                         |
| Urine Sample 7  | 4128              | 4140                         |
| Urine Sample 8  | 4652              | 4440                         |
| Urine Sample 9  | 4082              | 3910                         |
| Urine Sample 10 | 3350              | 3250                         |

**Supplementary References**

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