

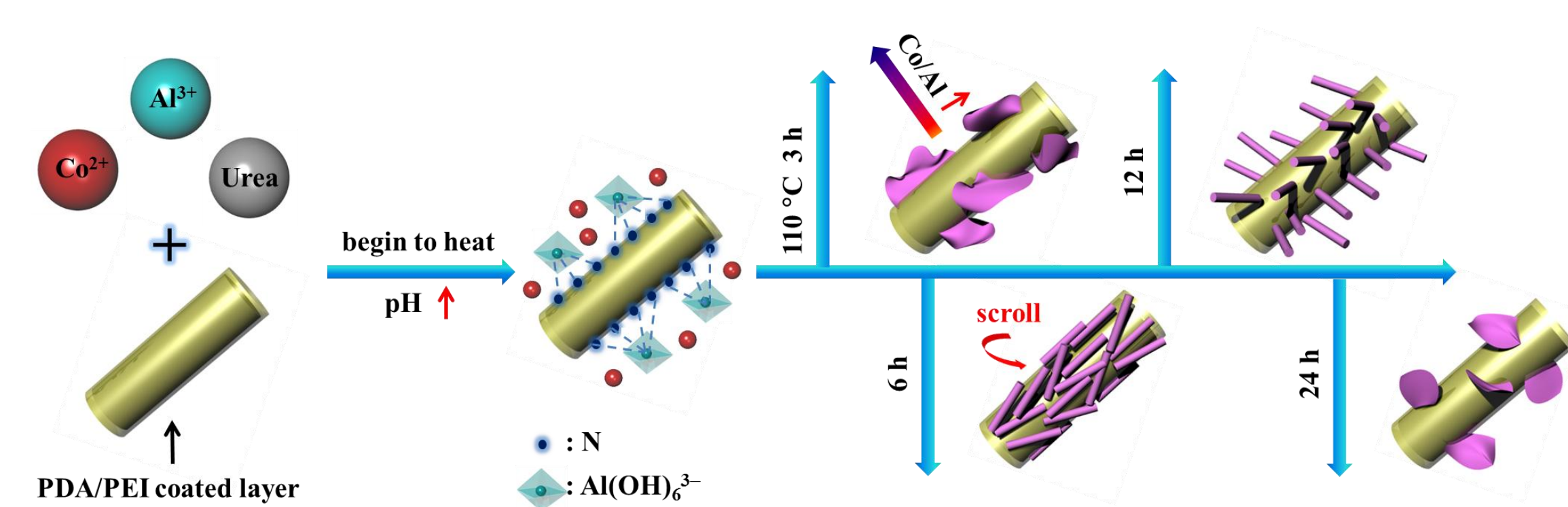


# General Strategy for Synthesis of Morphology-Tunable Layered Double Hydroxide on Arbitrary Substrates with a Mussel-Inspired Intermediate Layer: from Nanosheets to Nanoscrolls

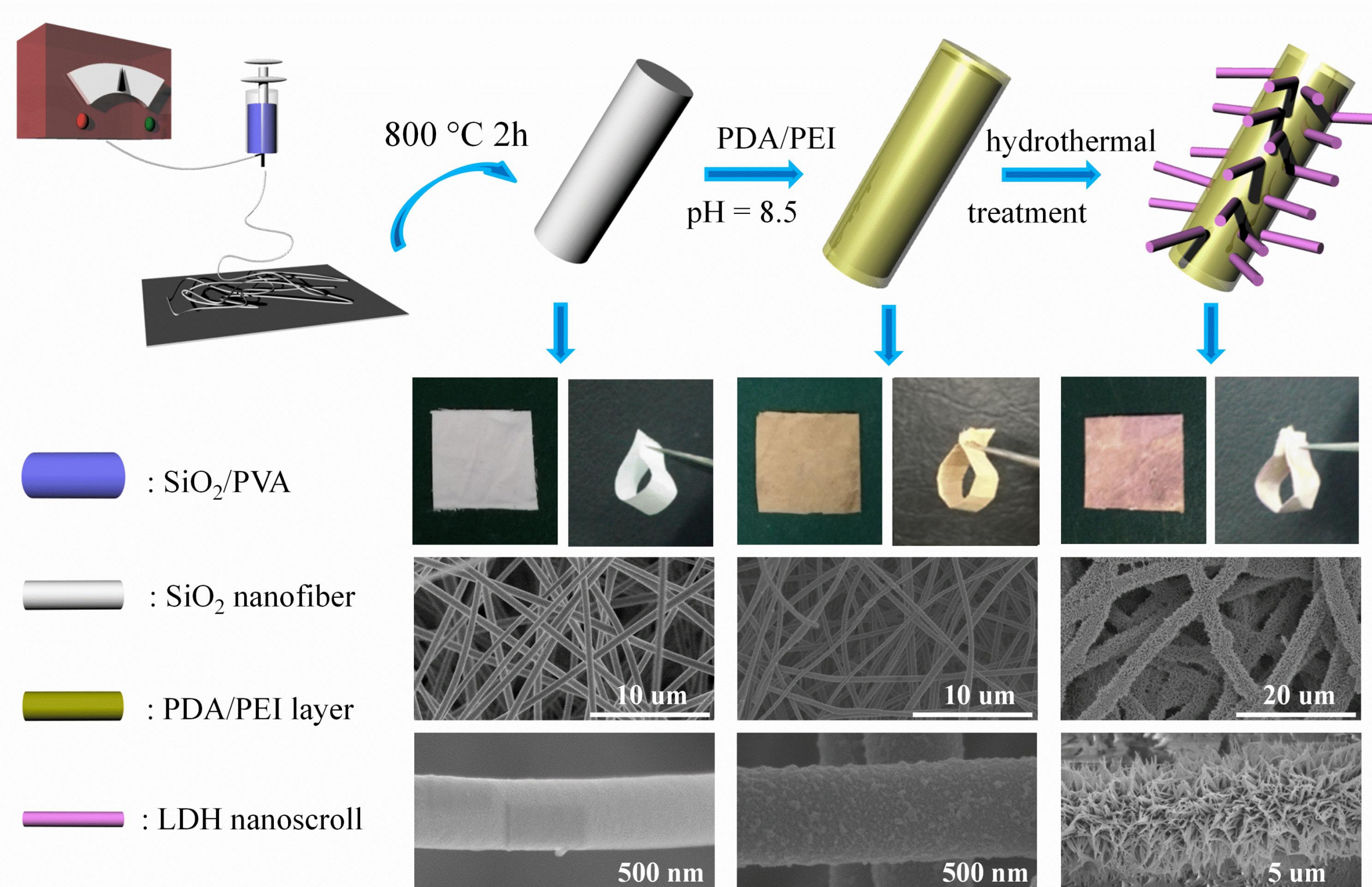
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## Introduction

Synthesis of morphology-controlled nanocrystals on target substrates presents an enticing prospect for fabricating multifunctional devices.<sup>1-3</sup> Herein, layered double hydroxides (LDH) with different morphologies were fabricated via hydrothermal treatment on various polydopamine/polyethylenimine (PDA/PEI) modified substrate surfaces. The morphology of the LDH nanostructures from ultrathin nanosheets, parallel aligned nanoscrolls and vertically aligned nanoscrolls could be tuned facily by reaction times, temperatures and metal-salt concentrations.

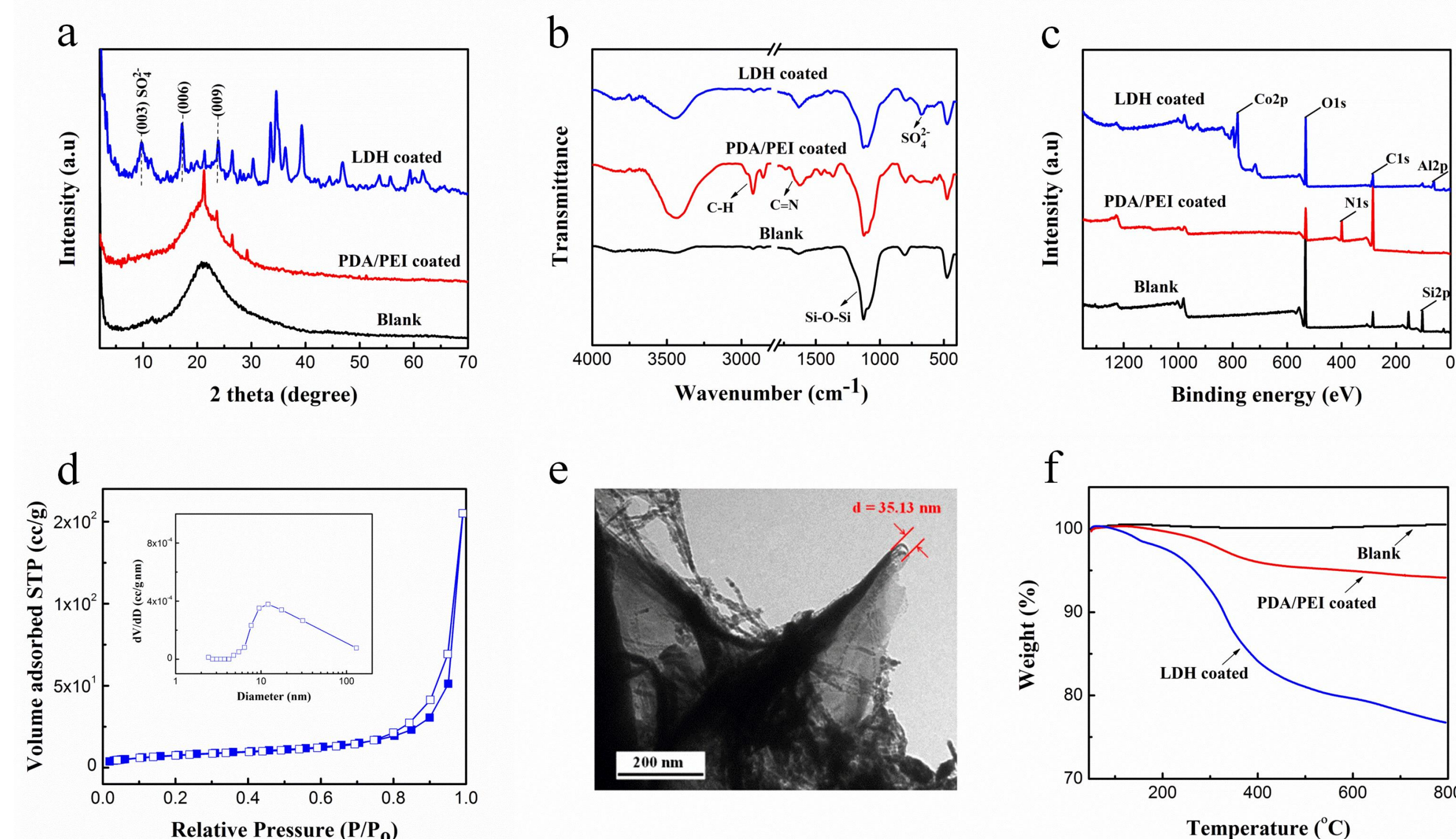


## Preparation steps



**Fig. 1** Schematic illustration of the preparation steps of LDH coated hybrid nanofiber membranes and their corresponding optical and SEM images.

## Structure characterization



**Fig. 2** Characterization of structure properties of the prepared membranes. (a) XRD patterns, (b) FT-IR spectra, (c) XPS spectra, (d)  $N_2$  adsorption-desorption isotherm of the LDH coated nanofiber membranes, (e) TEM image showing the LDH nanoscrolls on  $SiO_2$  nanofiber, and (f) TGA curves. The insert in (d) is the pore size distribution of the LDH coated membranes.

The XRD curves and FT-IR spectra confirmed the existence of LDHs. The atomic concentration ratio of Co/Al was calculated to be 92.7 in the XPS spectra, which was far beyond the stoichiometric ratio of Co/Al in the initial reaction solution (3:1). The specific surface area of the LDH coated membranes ( $28.82 \text{ m}^2/\text{g}$ ) increased by nearly twentyfold compared with that of the uncoated membranes ( $1.49 \text{ m}^2/\text{g}$ ), and the average pore diameter was about  $33.77 \text{ nm}$ , which was consistent with the TEM results.

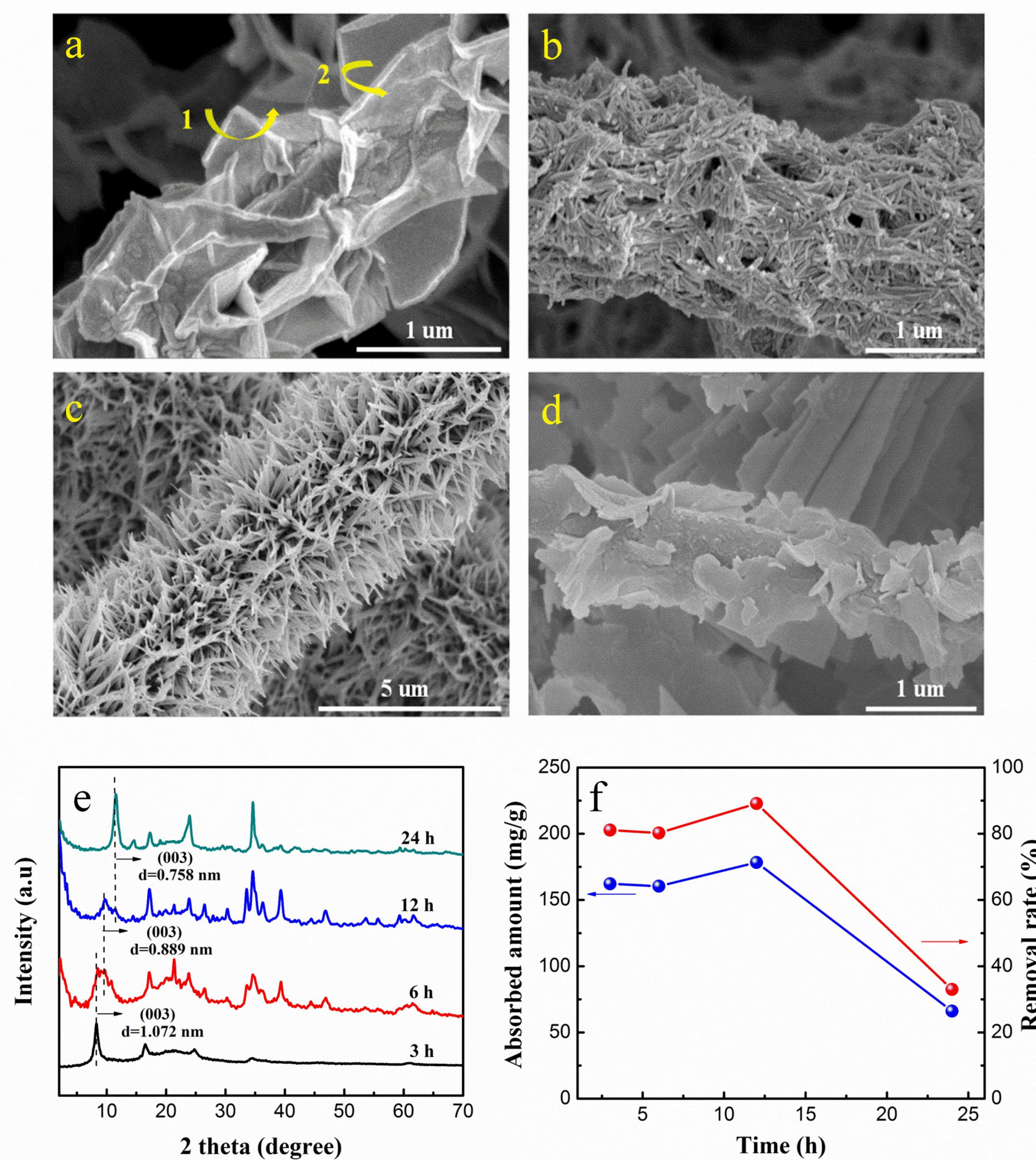
## Conclusions

1. Morphology controllable LDHs with ultrathin nanosheets, parallel aligned nanoscrolls and vertically aligned nanoscrolls on arbitrary substrates were successfully prepared by combining polydopamine /polyethylenimine (PDA/PEI) modified layer and the hydrothermal treatment.
2. The amino group and positively charged surface play critical roles in the formation of ultrathin flexible LDH nanosheets and then LDH nanoscrolls.

## References

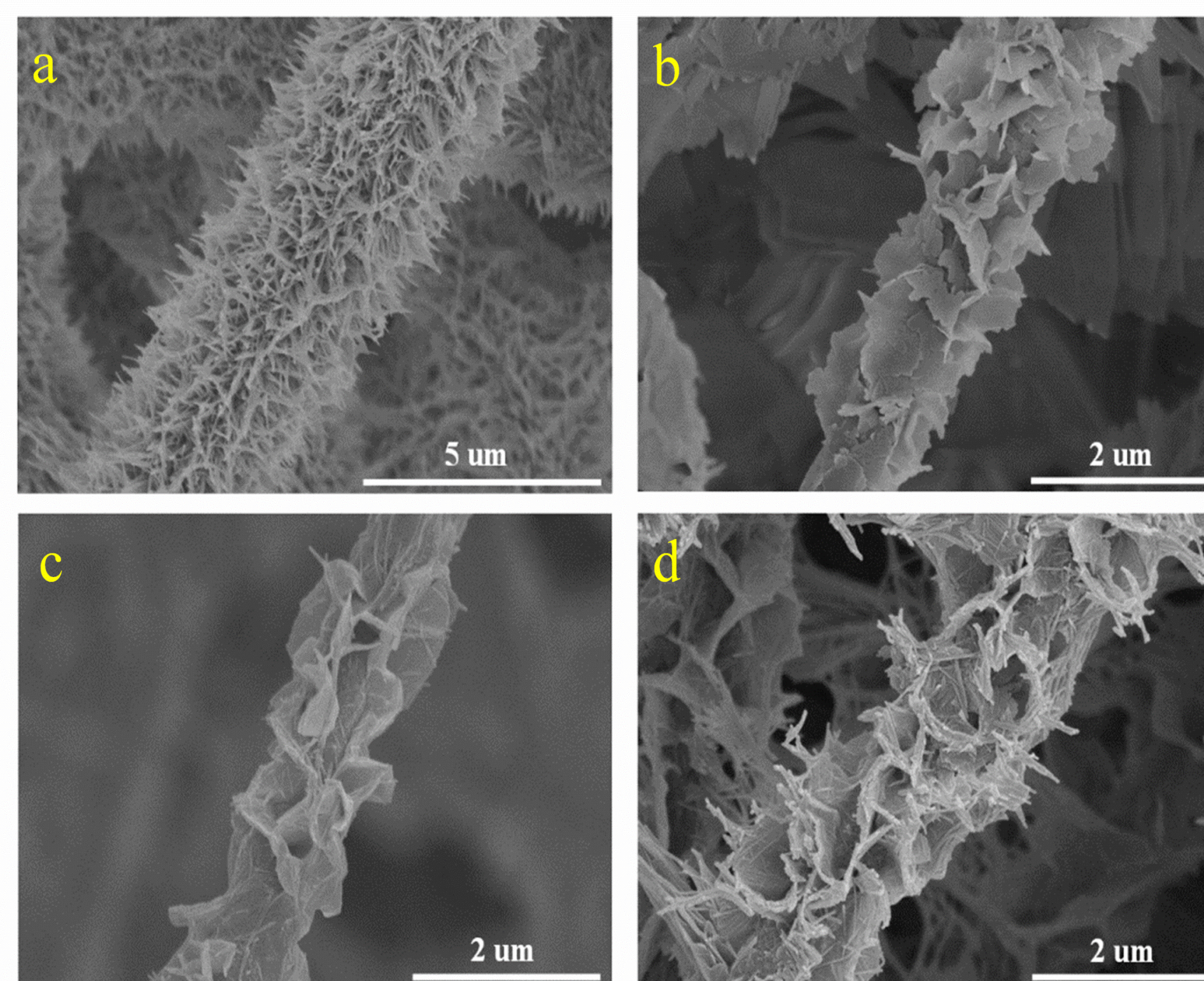
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## Morphological time evolution



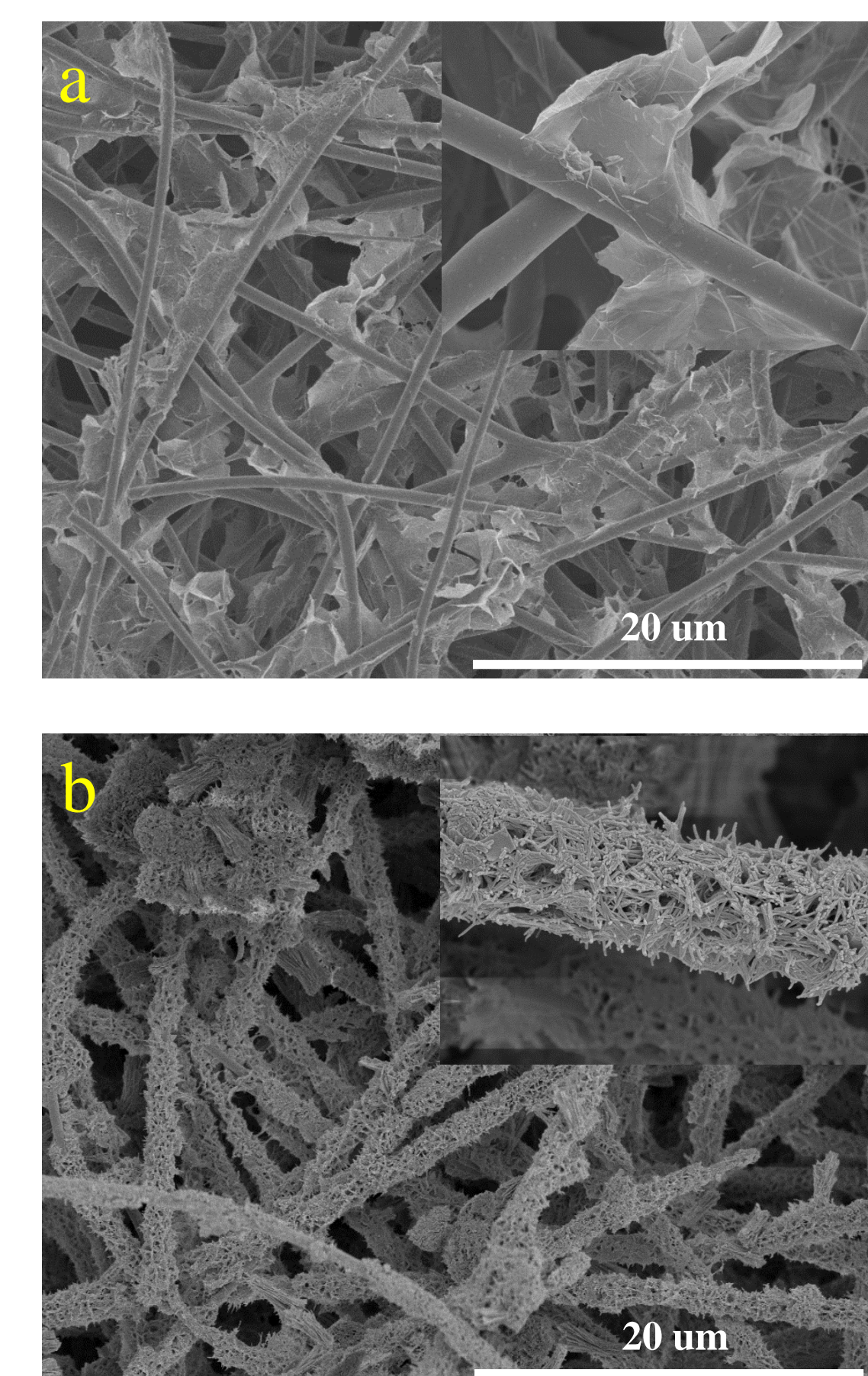
**Fig. 3** SEM micrographs of the LDHs grown on the nanofibers at different times (at  $110^\circ \text{C}$  and the total metal concentration of  $96 \text{ mmol/L}$ ): (a) 3 h, (b) 6 h, (c) 12 h, (d) 24 h and (e) their corresponding XRD patterns. (f) Adsorption properties of the LDH coated samples synthesized at different times for  $100 \text{ mg/L}$  MO aqueous solution.

## Temperature and metal concentration



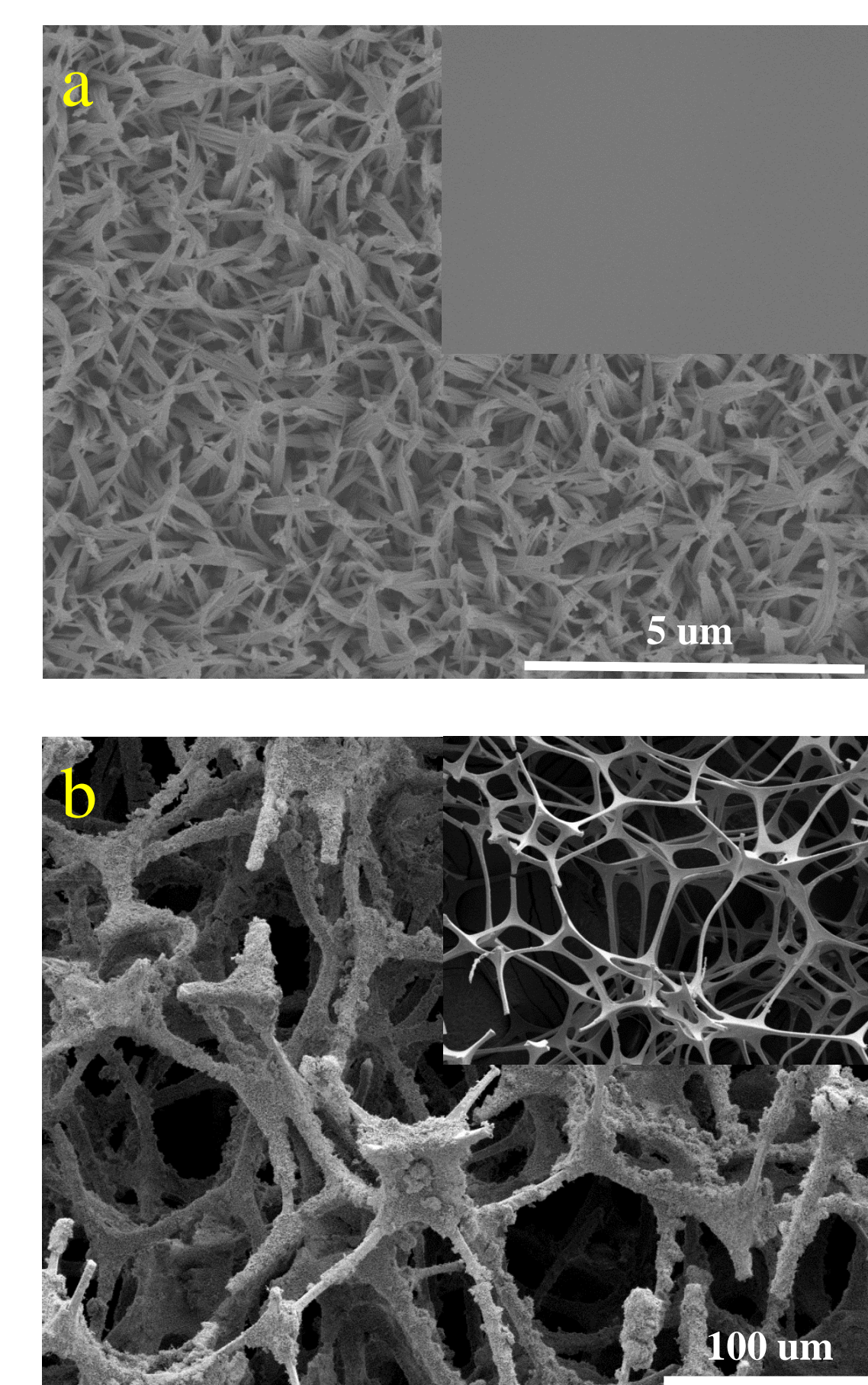
**Fig. 4** SEM micrographs of the LDHs grown on the nanofibers at different temperatures with the total metal concentration of  $96 \text{ mmol/L}$ : (a)  $100^\circ \text{C}$  and (b)  $120^\circ \text{C}$ . SEM micrographs of the LDHs grown on the nanofibers at  $110^\circ \text{C}$  with different metal concentrations: (c)  $12 \text{ mmol/L}$  and (d)  $48 \text{ mmol/L}$ . The reaction time of all experiments was 12 h.

## Targeted experiments



**Fig. 5** SEM images of the LDHs grown on (a) the nascent  $SiO_2$  membranes and (b) the PDA coated nanofibers with different magnifications.

## Various substrates



**Fig. 6** SEM images of the LDHs grown on (a) the aluminum foil and (b) the melamine sponge. The inserts in (a) and (b) are the blank substrates.