

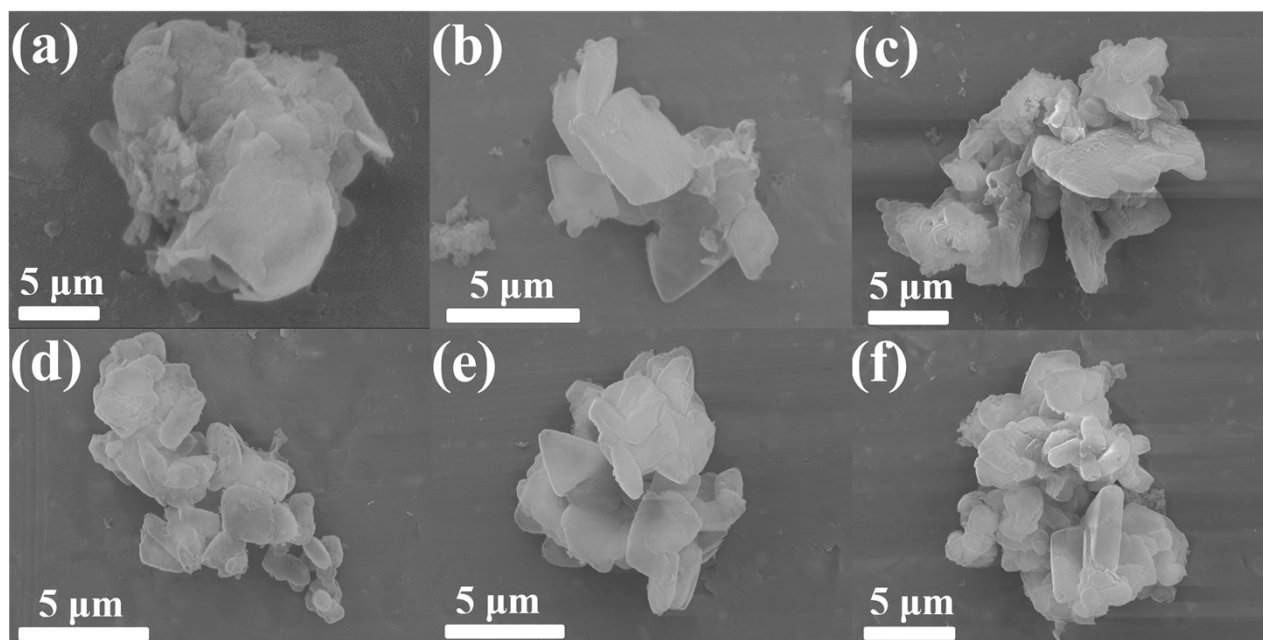
## Supplementary Information

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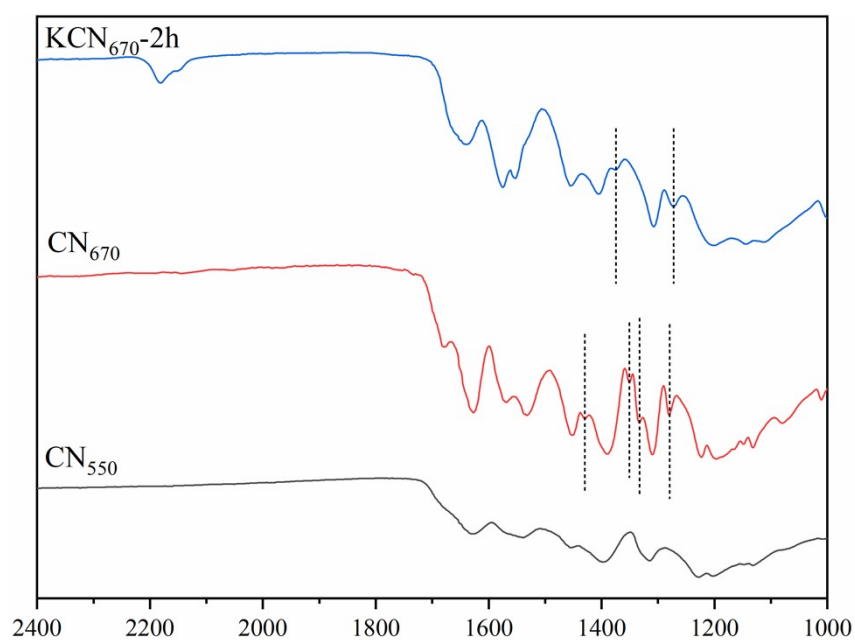
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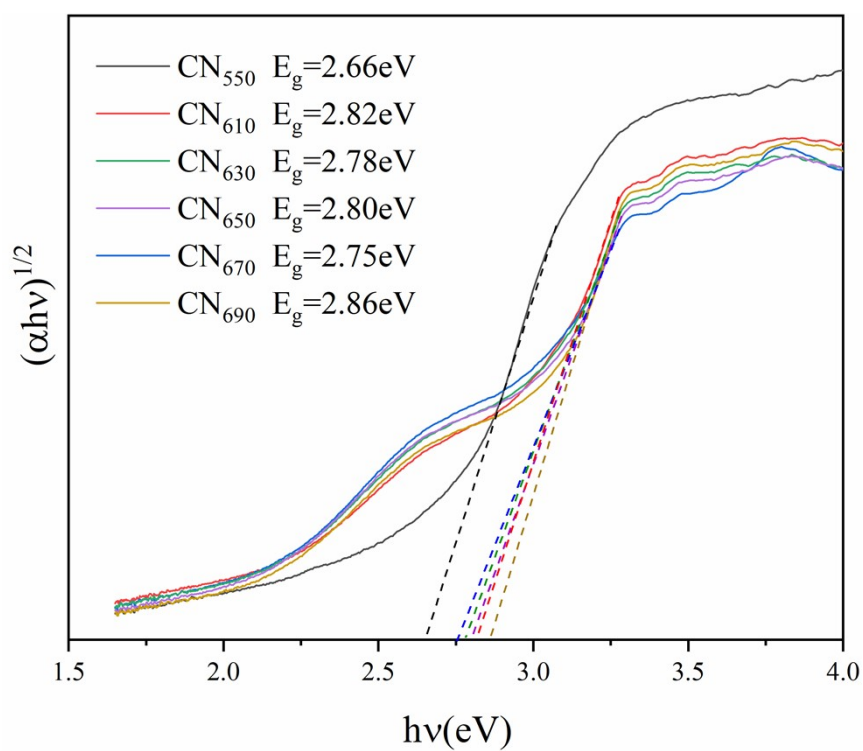
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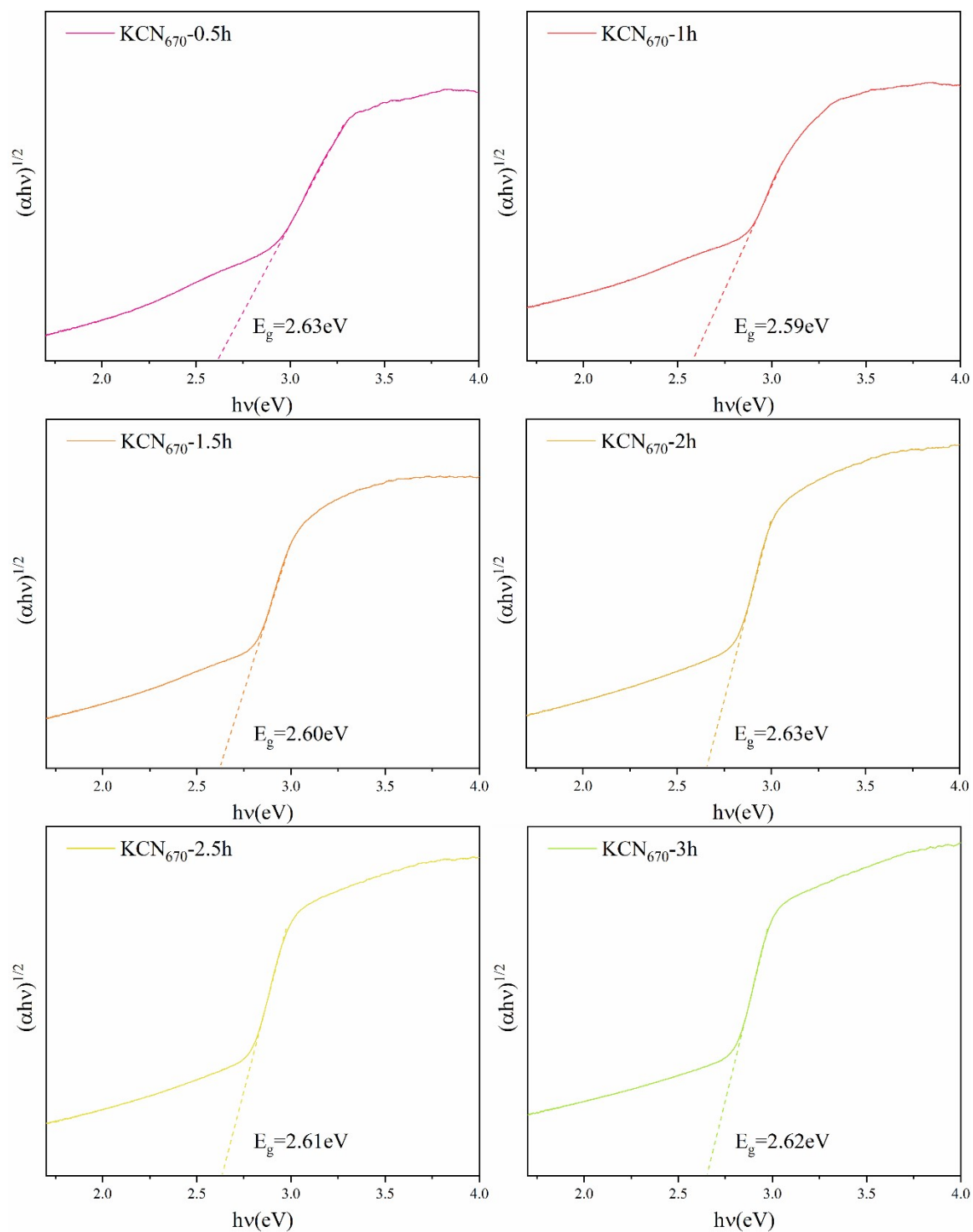
**Fig. S1** FE-SEM images of (a) CN<sub>550</sub>, (b) CN<sub>610</sub>, (c) CN<sub>630</sub>, (d) CN<sub>650</sub>, (e) CN<sub>670</sub>, and (f) CN<sub>690</sub>.



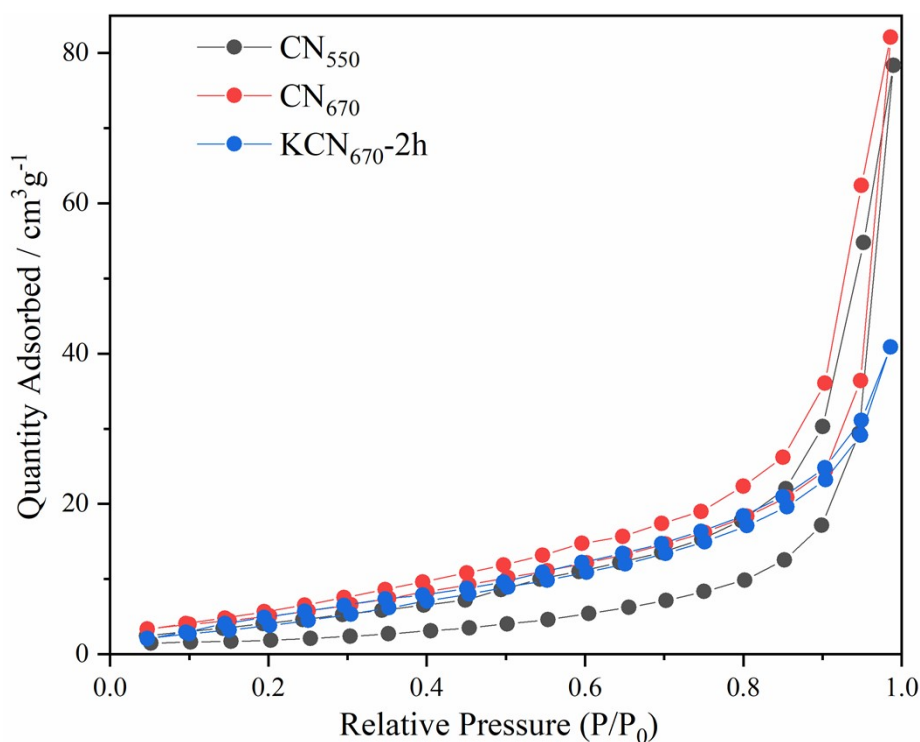
**Fig. S2** FT-IR spectra of CN<sub>550</sub>, CN<sub>670</sub>, and KCN<sub>670</sub>-y.



**Fig. S3** Plots of transformed Kubelka-Munk function for CN<sub>x</sub> samples.



**Fig. S4** Plots of transformed Kubelka-Munk function for  $\text{KCN}_{670-y}$  samples.



**Fig. S5** N<sub>2</sub> adsorption-desorption isotherms of CN<sub>550</sub>, CN<sub>670</sub>, and KCN<sub>670</sub>-2h

**Table S1** The element contents in CN<sub>550</sub>, CN<sub>670</sub>, KCN<sub>670</sub>-1h, KCN<sub>670</sub>-2h and KCN<sub>670</sub>-3h samples estimated by elemental analysis.

| Elemental analysis      | C(wt%) | N(wt%) | H(wt%) | C/N(mol ratio) |
|-------------------------|--------|--------|--------|----------------|
| CN <sub>550</sub>       | 34.5   | 61.1   | 2.21   | 0.658          |
| CN <sub>670</sub>       | 33.6   | 58.6   | 2.11   | 0.669          |
| LiCN <sub>670</sub> -1h | 32.1   | 55.5   | 1.97   | 0.674          |
| LiCN <sub>670</sub> -2h | 29.7   | 50.9   | 1.86   | 0.680          |
| LiCN <sub>670</sub> -3h | 27.5   | 46.4   | 1.72   | 0.691          |
| NaCN <sub>670</sub> -1h | 32.4   | 56.2   | 1.99   | 0.673          |
| NaCN <sub>670</sub> -2h | 30.5   | 52.7   | 1.91   | 0.675          |
| NaCN <sub>670</sub> -3h | 28.9   | 49.4   | 1.85   | 0.682          |
| KCN <sub>670</sub> -1h  | 33.2   | 57.2   | 1.90   | 0.678          |
| KCN <sub>670</sub> -2h  | 32.4   | 55.2   | 1.75   | 0.685          |
| KCN <sub>670</sub> -3h  | 28.8   | 47.8   | 1.65   | 0.703          |

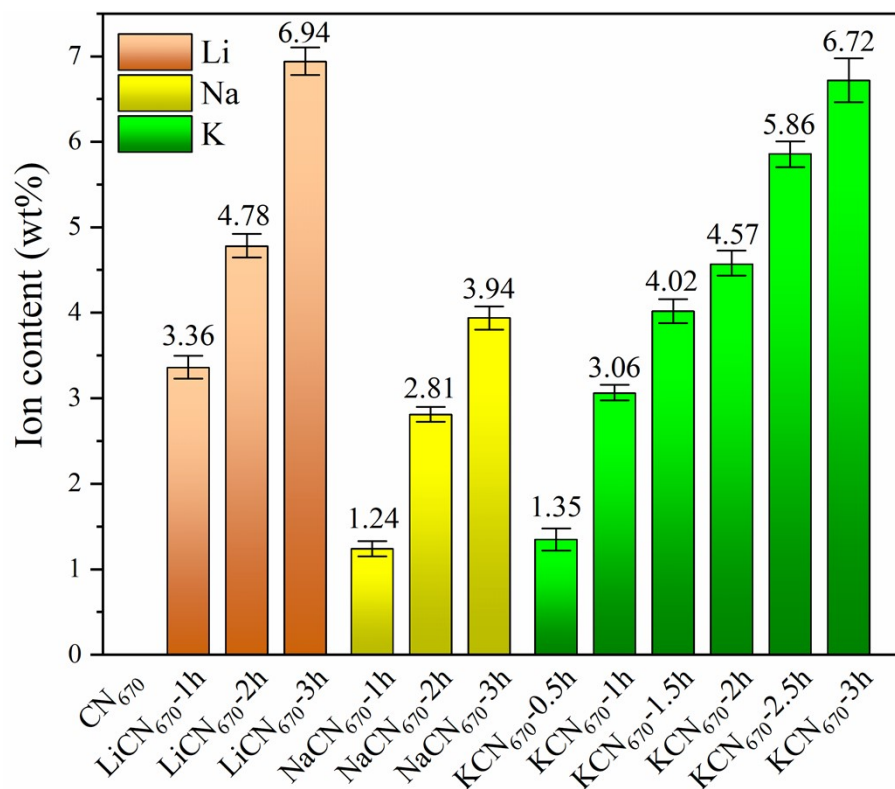
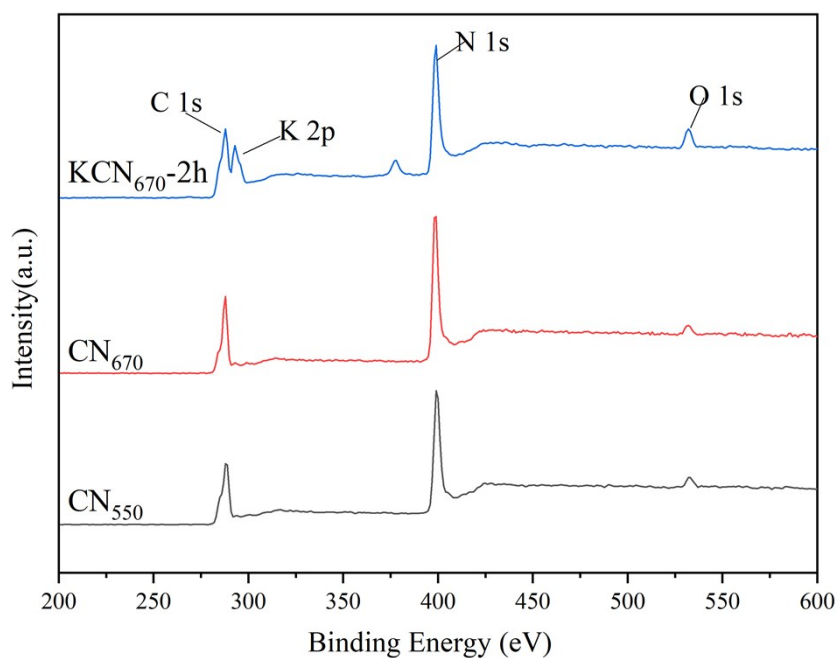


Fig.S6 The Li, Na, K element contents in CN<sub>670</sub>, ACN<sub>670</sub>-1h, ACN<sub>670</sub>-2h, and ACN<sub>670</sub>-3h samples (A=Li, Na, K) estimated by ICP.



**Fig. S7** XPS survey spectra of CN<sub>550</sub>, CN<sub>670</sub>, and KCN<sub>670</sub>-2h.

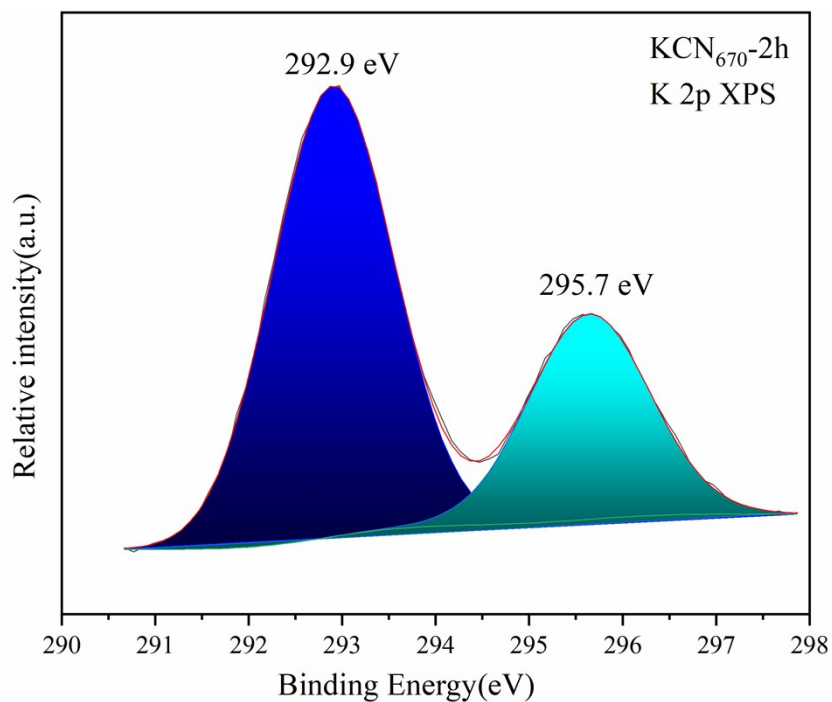


Fig.S8 K 2p XPS spectra of KCN<sub>670</sub>-2h samples.

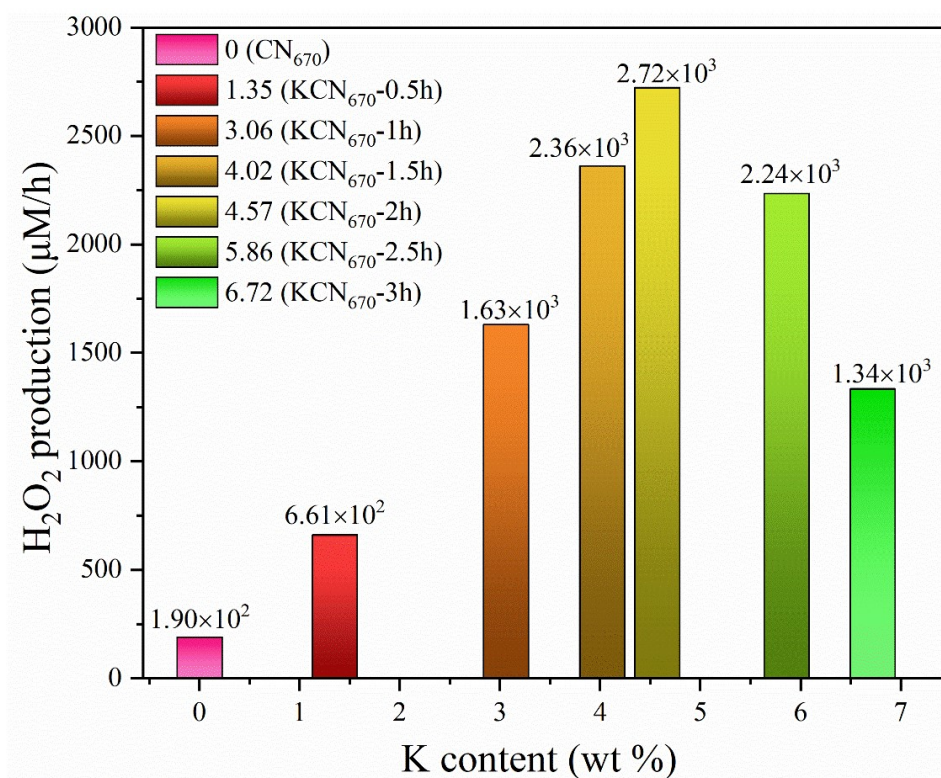
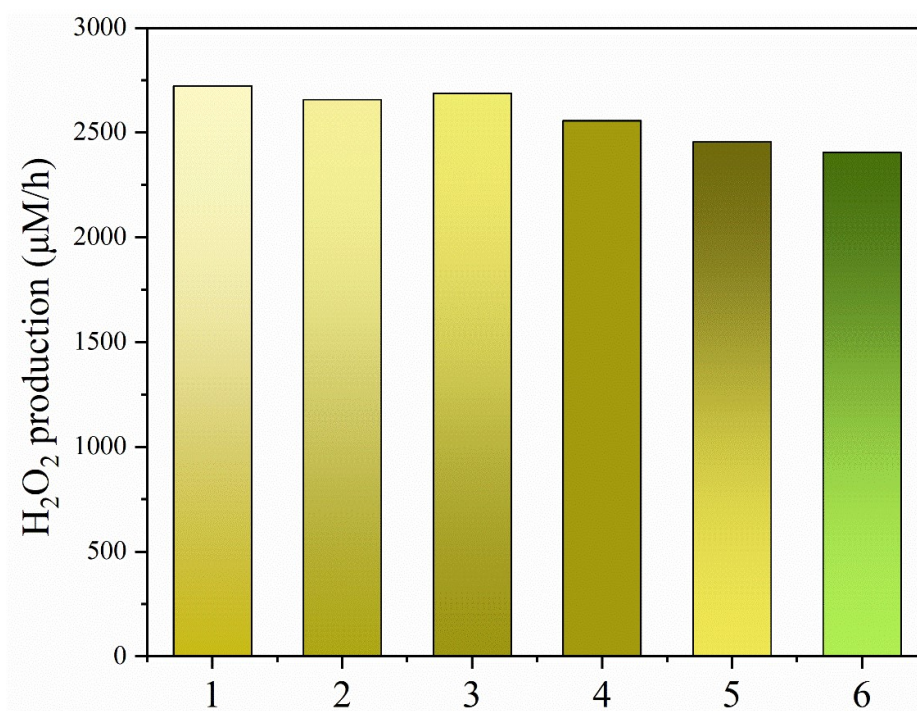


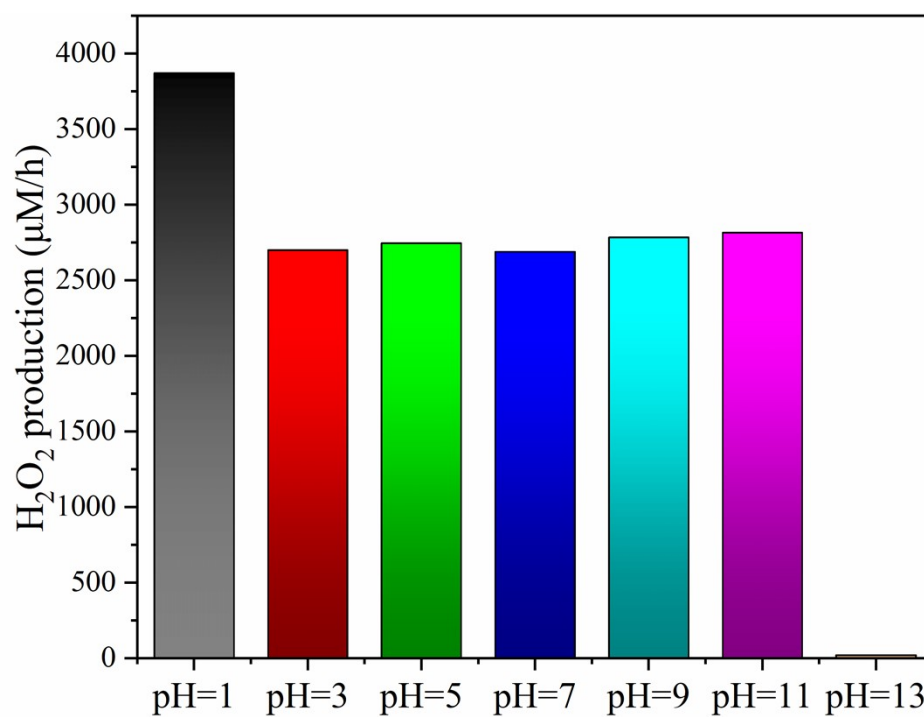
Fig. S9 H<sub>2</sub>O<sub>2</sub> production rate dependence on K content for KCN<sub>670</sub>-y samples.

Table. S2 Summary table of H<sub>2</sub>O<sub>2</sub>-production by different g-C<sub>3</sub>N<sub>4</sub>-based materials.

| Catalysts                                  | Light Source                                         | Reaction Conditions                                    | H <sub>2</sub> O <sub>2</sub> Production (μM h <sup>-1</sup> ) | Reference |
|--------------------------------------------|------------------------------------------------------|--------------------------------------------------------|----------------------------------------------------------------|-----------|
| KCN <sub>670</sub> -2h                     | solar simulator (AM 1.5 G, 100 mW cm <sup>-2</sup> ) | Aqueous (50 mg catalyst, 100 mL, IPA) + Air            | 2.72×10 <sup>3</sup>                                           | This work |
| PDI/CNA                                    | solar simulator (AM 1.5 G, 100 mW cm <sup>-2</sup> ) | Aqueous (50 mg catalyst, 50 mL, IPA) + O <sub>2</sub>  | 1.61×10 <sup>3</sup>                                           | [1]       |
| MPCN                                       | 300 W Xenon lamp (λ>420 nm)                          | Aqueous (50 mg catalyst, 100 mL, IPA) + Air            | 2.21×10 <sup>3</sup>                                           | [2]       |
| KTCN                                       | 300 W Xenon lamp (λ>420 nm)                          | Aqueous (30 mg catalyst, 30 mL, IPA) + O <sub>2</sub>  | 7.2×10 <sup>2</sup>                                            | [3]       |
| KCN-1                                      | 300 W Xenon lamp (λ>400 nm)                          | Aqueous (30 mg catalyst, 100 mL, IPA) + O <sub>2</sub> | 1.35×10 <sup>3</sup>                                           | [4]       |
| K, P, O, S-g-C <sub>3</sub> N <sub>4</sub> | 300 W Xenon lamp (λ>420 nm)                          | Aqueous (150 mg catalyst, 150 mL, IPA) + Air           | 1.18×10 <sup>3</sup>                                           | [5]       |
| K-g-C <sub>3</sub> N <sub>4-x</sub>        | 300 W Xenon lamp (λ>420 nm)                          | Aqueous (10 mg catalyst, 50 mL, EDTA) + O <sub>2</sub> | 7.07×10 <sup>2</sup>                                           | [6]       |
| KOH-doped g-C <sub>3</sub> N <sub>4</sub>  | 500 W Xenon lamp                                     | Aqueous (30 mg catalyst, 30 mL, IPA) + Air             | 21                                                             | [7]       |
| SCN                                        | 300 W Xenon lamp (λ>420 nm)                          | Aqueous (20 mg catalyst, 40 mL, IPA) + Air             | 14                                                             | [8]       |
| Nv-M                                       | 300 W Xenon lamp (λ ≥ 420 nm)                        | Aqueous (50 mg catalyst, 50 mL, IPA) + Air             | 31                                                             | [9]       |
| KDCN-0.2                                   | 300 W Xenon lamp (λ>420 nm)                          | Aqueous (50 mg catalyst, 100 mL, IPA) + Air            | 2.79×10 <sup>2</sup>                                           | [10]      |



**Fig. S10** Cycling experiments of H<sub>2</sub>O<sub>2</sub> production over KCN<sub>670</sub>-2h sample.



**Fig. S11** H<sub>2</sub>O<sub>2</sub> production rates under different pH conditions

**Table S3** Values of elements in equivalent circuit resulted from fitting the EIS data.

| Sample                 | $R_s(\Omega \text{ cm}^{-2})$ | $R_{ct}(\Omega \text{ cm}^{-2})$ | $CPE(F \text{ cm}^{-2})$ |
|------------------------|-------------------------------|----------------------------------|--------------------------|
| CN <sub>550</sub>      | 32.5                          | 9.62E5                           | 3.42E-5                  |
| CN <sub>670</sub>      | 81.1                          | 4.18E5                           | 2.34E-5                  |
| KCN <sub>670</sub> -1h | 84.6                          | 2.43E5                           | 2.35E-5                  |
| KCN <sub>670</sub> -2h | 28.5                          | 1.55E4                           | 3.71E-5                  |
| KCN <sub>670</sub> -3h | 28.1                          | 1.16E5                           | 2.24E-5                  |

## References

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