

Nanocellulose-Paper-Based Analytical Devices with MOFs/Heterojunction Structures for Multiplex SERS Detection

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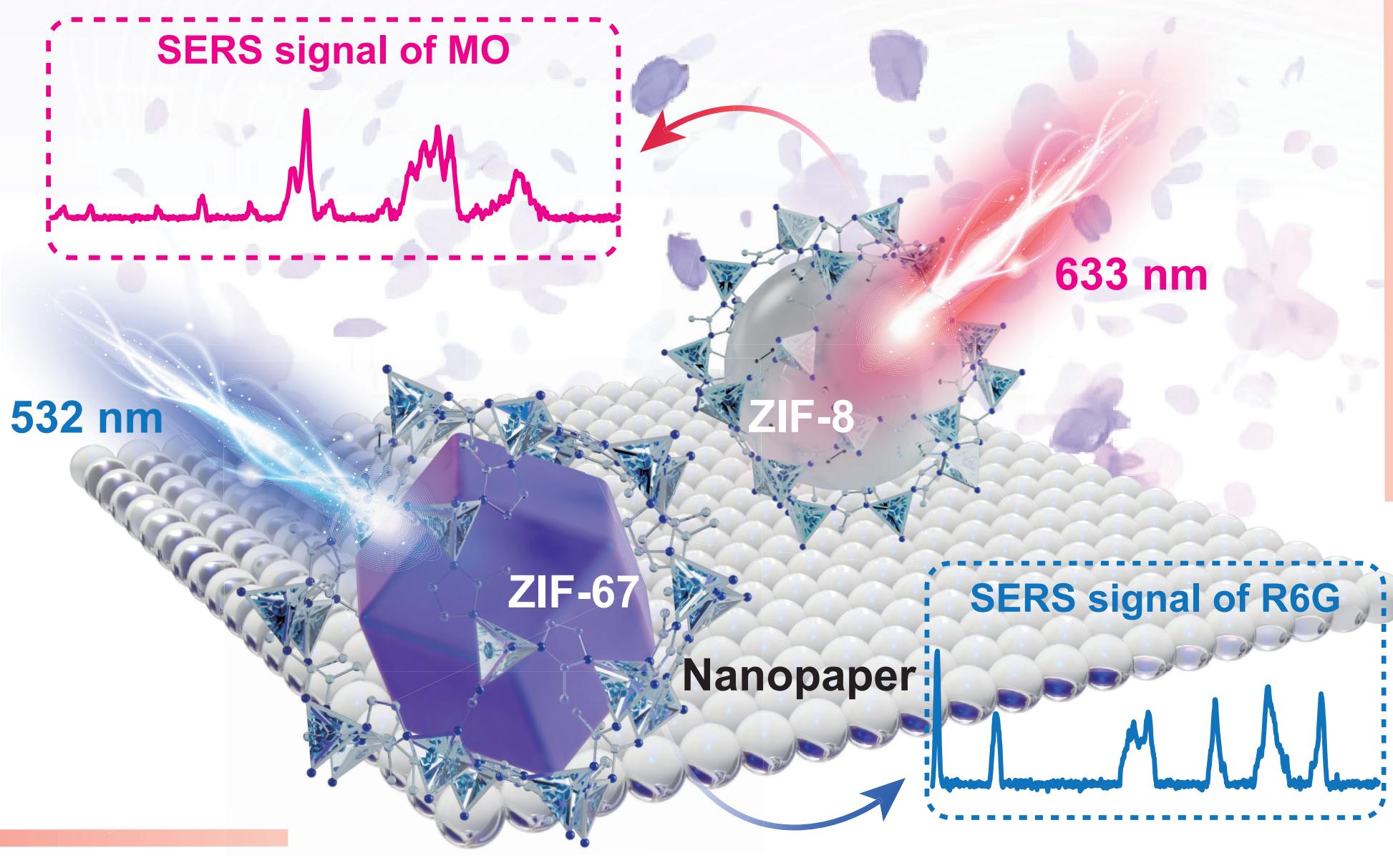
ABSTRACT

Metal-organic frameworks (MOFs) as surface-enhanced Raman scattering (SERS) substrates exhibit selectivity towards analytes with different energy levels. However, due to the energy level mismatch, the multiplex SERS detection on the same substrate has not been developed yet. In this study, we demonstrated nanocellulose-paper-based analytical devices (NanoPADs) with both in-situ ZIF-8/Zn(OH)₂ and ZIF-67/Co(OH)₂ structures. As a proof-of-concept, we independently detected two common environmental pollutants, methyl orange (MO) and Rhodamine 6G (R6G), on NanoPADs under different incident laser wavelengths, achieving limits of detection as 10⁻⁷ M, respectively.

INTRODUCTION

- MOFs have received considerable attention as SERS substrates due to their molecular selectivity [1].
- The existing MOFs-based SERS substrates are restricted by their charge transfer (CT) mechanism and can only detect analytes with specific energy levels [2].

Need to develop MOFs-based SERS substrates for multiplex detection.



RESULTS

- Multiplex SERS detection can be achieved within the ratio range of Co to Zn ions from 3:1 to 1:3 (Fig. 4).
- Compared with the LOD of pure ZIF-8 and ZIF-67 achieved before (10⁻⁷ M), the LOD of our NanoPADs was similar (10⁻⁷ M).

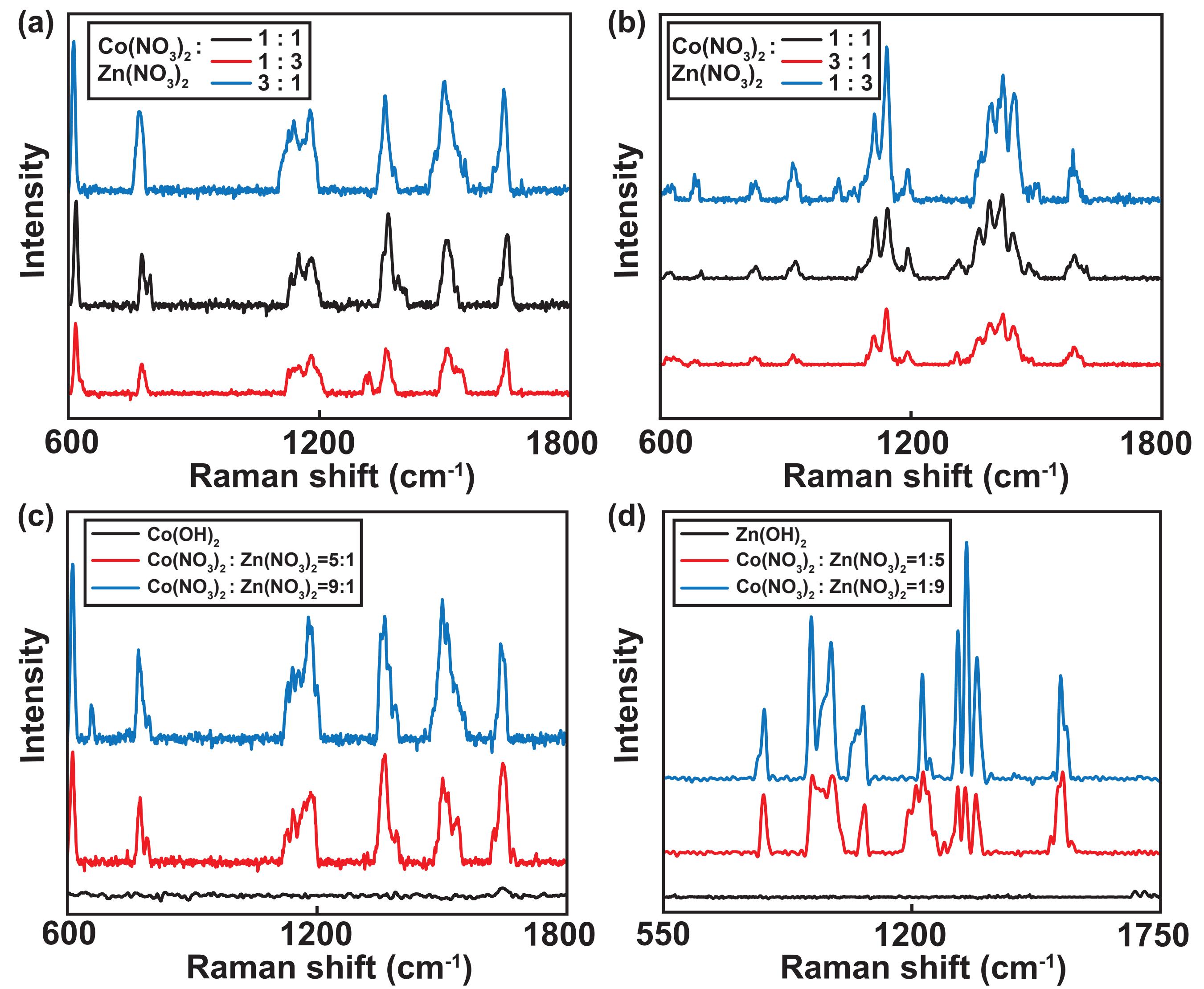


Fig. 4. SERS detection results on different NanoPADs. (a, c) Raman spectra of 100 nM R6G. (b, d) Raman spectra of 100 nM MO. The ratio was defined as the amount of Co(NO₃)₂ to Zn(NO₃)₂ (mol) used in fabricating NanoPADs.

CONCLUSION

The NanoPADs retain the molecular selectivity of MOFs and relatively high sensitivity in multiplex SERS detection, advancing SERS-based device development.

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Developed NanoPADs by generating both in-situ ZIF-8/Zn(OH)₂ and ZIF-67/Co(OH)₂ heterojunction structures on nanopaper (Figs. 1 and 2).

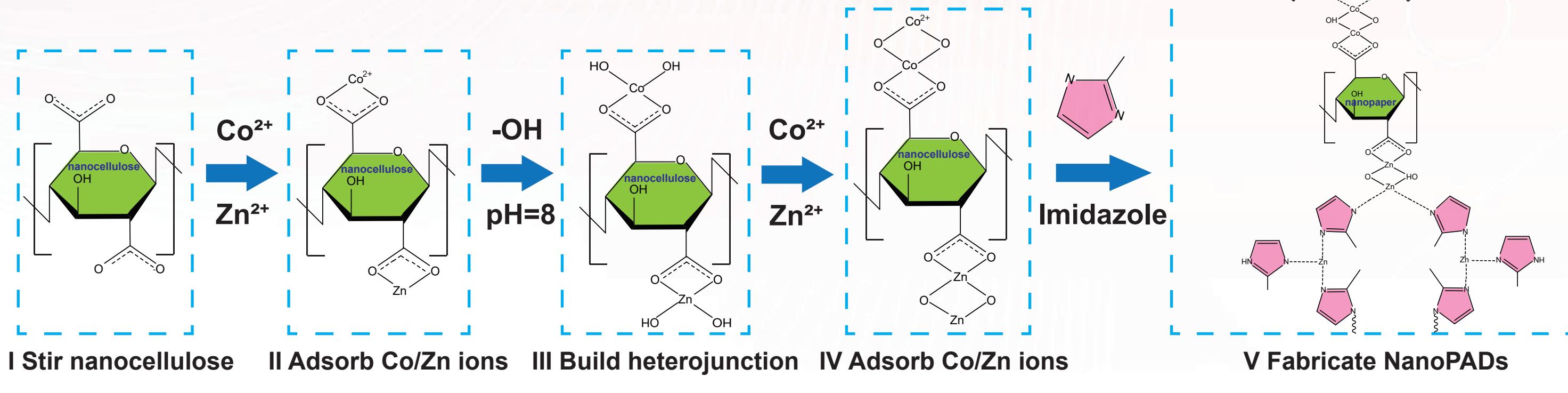


Fig. 1. Fabrication process of NanoPADs.

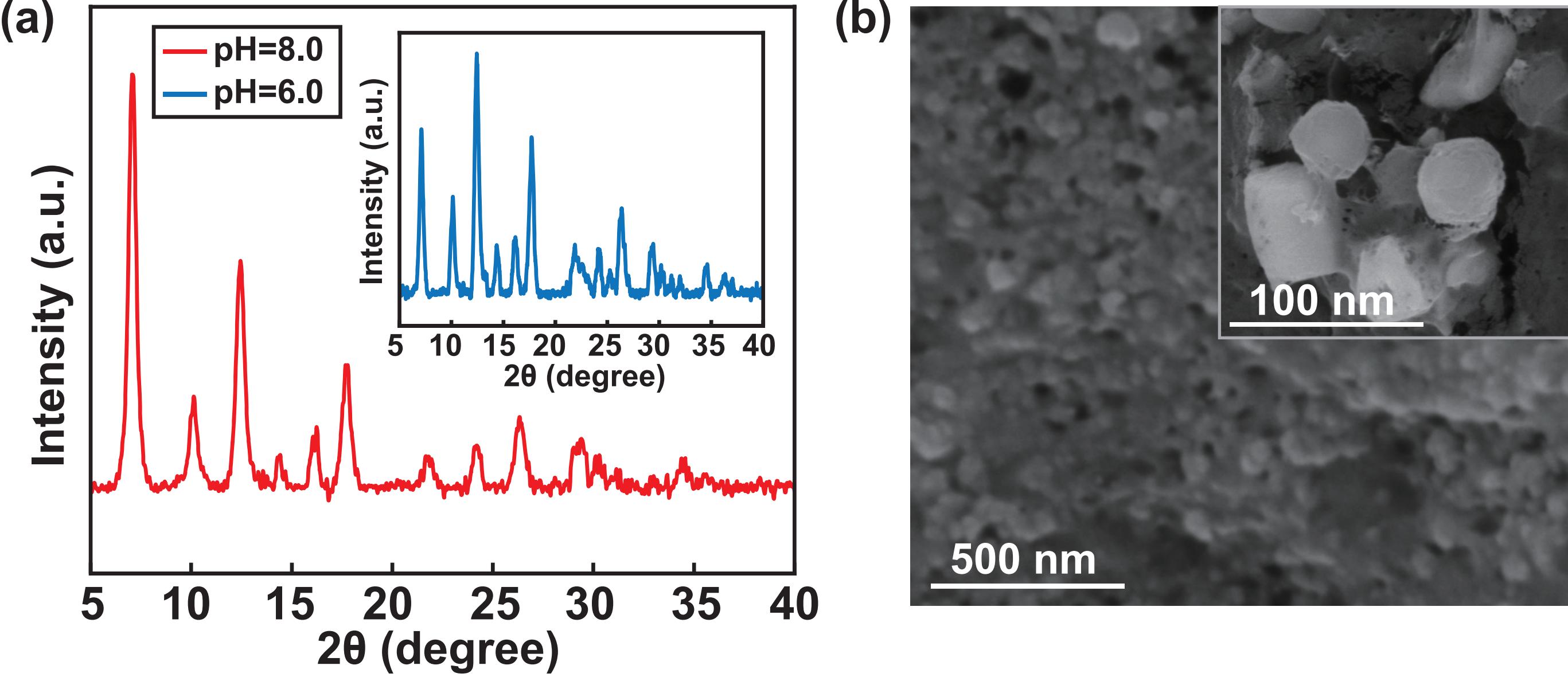


Fig. 2. Characterization of NanoPADs. (a) XRD spectra of both ZIF-67 and ZIF-8 lattice. (b) SEM image of these nanoparticles on NanoPADs.

- In our MOFs/heterojunction structures [3], hydroxides provide additional CT to MOFs to enhance SERS signals (Fig. 3).
- Shift incident laser length from 633 nm to 532 nm to initiate photo-induced CT between ZIF-8 to MO and ZIF-67 to R6G.

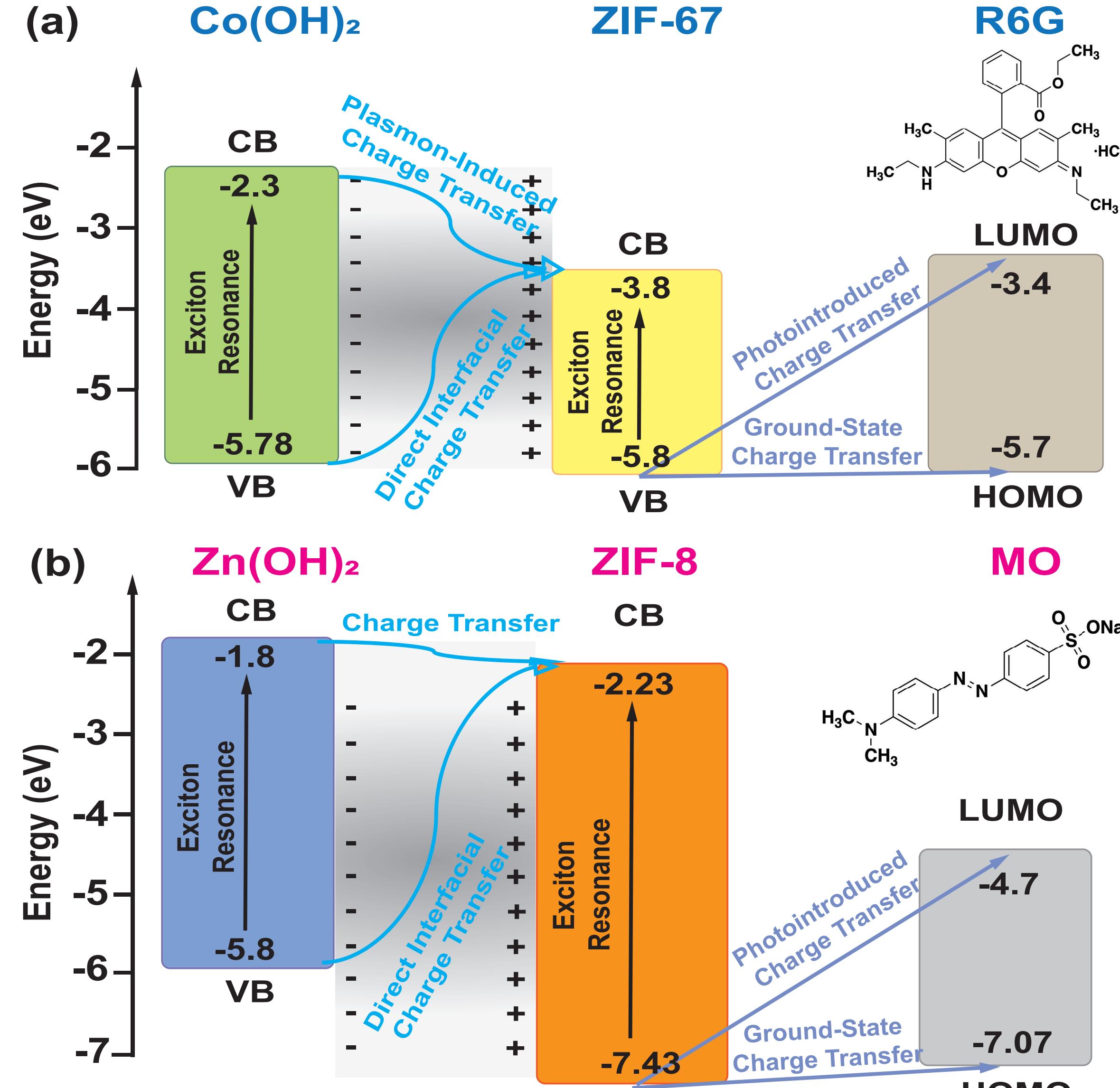


Fig. 3. (a) Energy level diagram of R6G on NanoPADs under 532 nm illumination. (b) Energy level diagram of MO on NanoPADs under 633 nm illumination.

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