Graphite felt incorporated with MoS_2/rGO for electrochemical detoxification of high-arsenic fly ash

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1. Introduction – Accumulation of high-arsenic fly ash (HAFA) poses a serious environmental threat due to the toxicity. As(III) oxidation is a necessary step for HAFA detoxification and in situ H_2O_2 electrogeneration via the two-electron-pathway ($2e^-$) oxygen reduction reaction (ORR) is considered a promising technique for As(III) oxidation [1, 2]. MoS_2 has been reported to have attractive $2e^-$ ORR catalytic activity [3]. However, there are still some problems to be solved: (i) its conductivity needs to be improved, (ii) gaseous O_2 is difficult to be effectively utilized, and (iii) the assembled electrode is not flexible and durable enough to treat solid waste.

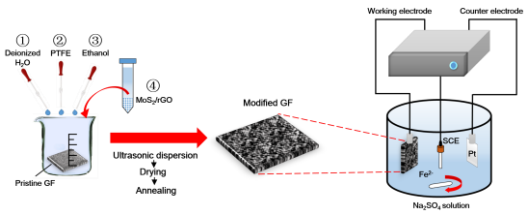

Image 1. Schematic diagram of experimental process.

2. Experimental - the experimental flow is schematically illustrated in Image. 1.

3. Results and Discussion - A novel graphite felt (GF) modified with the MoS_2/reduced graphene oxide (rGO) heterojunction is developed and demonstrated as an efficient electrode for electrochemical detoxification of HAFA, as shown in Image 2(a). The modified GF has abundant microchannels (Image 2(b)), desirable surface properties, and fast electron transfer capability thereby enabling effective utilization of both dissolved O_2 and gaseous O_2 in $2e^-$ ORR. The p-n junction consists of the p-type rGO substrate and uniformly distributed n-type MoS_2 nanoflowers, seeing Images 2(c) and 2(d). Theoretical calculation indicates that gaseous O_2 adsorb stably on sulfur vacancies and is reduced by electrons transferred from rGO. The modified GF exhibits superior ORR catalytic activity such as a high H_2O_2 yield of 47.53 $mg \cdot L^{-1} \cdot h^1 \cdot cm^{-1}$ (Image 2(e)) and onset potential of -0.12 V vs. SCE. The ·OH generated by the autocatalytic mechanism promotes oxidative dissolution of As(III) during detoxification of HAFA and the As removal percentage is 96.10% after electrolysis for 135 min, displayed in Images 2(f) and 2(g). The modified GF with excellent stability and durability has large industrial potential in detoxification of HAFA and other As-bearing hazardous wastes.

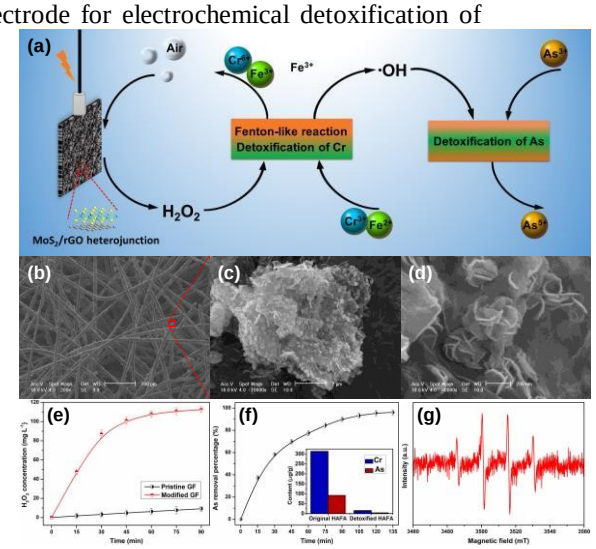

Image 2. (a) Graphical abstract of this study, (b) SEM image of the modified GF, (c) Low-magnification and (d) High-magnification SEM images of MoS_2/rGO heterojunction, (e) H_2O_2 accumulation versus time, (f) As removal percentage of HAFA versus time for the modified electrode (inset showing Cr and As contents in the original HAFA and detoxified HAFA), and (g) ESR spectrum of the hydroxyl radicals detected during the HAFA treatment.

4. Conclusions - This study makes significant contributions. Firstly, the gaseous O_2 reaction path in ORR is confirmed by the combination of experiment and calculation. Secondly, 2e ORR is innovatively applied to detoxification of high-arsenic solid waste.

5. References

[1] M. Sun et al., *Environ. Sci. Technol.* 49 (2015) p. 9289–9297.

[2] H.A. Maitlo et al., Ind. Eng. Chem. 73 (2019) p. 205–213. [3] Y. Xue et al., *Electrochim. Acta* 252 (2017) p. 245–253.