# Supporting Information for Pyrite Oxidation by Hexavalent Chromium: Investigation of the Chemical

# Processes by Monitoring of Aqueous Metal Species

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5 Pages, 2 Figures, 1 Table, 13 references

## **Procedure**

The schematic diagram of experimental apparatus is shown in Figure S1. Experiments were conducted under a controlled atmosphere (argon circulation) to minimise the amount of dissolved oxygen in the system (not controlled) and a Teflon® reactor was used to reduce adsorption of reactants to surfaces. The reactor was  $HNO_3$  cleaned and rinsed with ion-exchanged water between experiments. It was placed inside a glass thermostatically controlled vessel. The temperature was maintained at  $25 \pm 0.1$ °C with a circulating water bath. The glass vessel was closed with a cover containing four main ports. These ports were fitted with a pH electrode (Ross combined electrode), a mechanical stirring motor, a silicon disk septum for solution sampling/injection via a syringe and a pyrite reservoir flask.

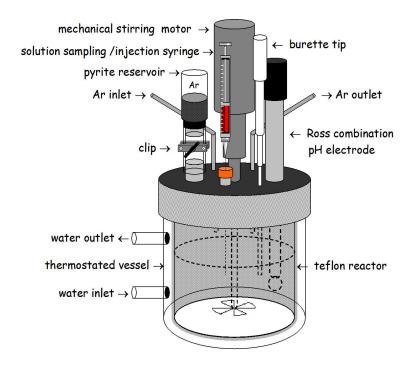


Figure S1: Schematic diagram of reactor used in pyrite oxidation experiments.

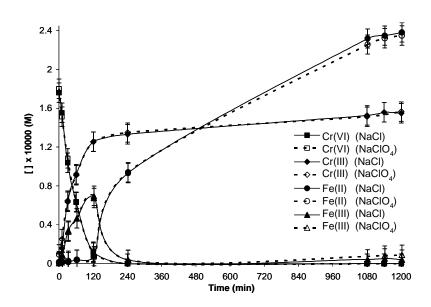
Experiments were started by equilibrating the supporting electrolyte (0.1 M NaClO<sub>4</sub>) under the Ar atmosphere for approximately 2 hours. Then, the appropriate amount of the pre-treated pyrite powder

was introduced in the reactor for equilibration in the inert electrolyte for approximately 1 hour and half.

The pyrite suspension was sufficiently stirred to keep the majority of the particles suspended. The

amount of the mineral used was determined to give a 6 g.L<sup>-1</sup> concentration. After equilibration, an appropriate stock solution of  $K_2CrO_4$  was added to the reactor to give initial Cr(VI) concentrations in the range of 1-7x10<sup>-4</sup> M. A small known amount of  $HClO_4$  was simultaneously added to fix pH values in the range of 2-8. The pH over time was then monitored and adjusted to the constant consign value (pH drifts < 0.1 unit) using a computer-controlled titrator (736 GP Titrino, Metrohm) and the Ross pH combined electrode. The pH readings and added solution volumes were recorded for 20 hours. Samples were withdrawn from the reactor at different time intervals and filtered through 0.22  $\mu$ m pore membrane filters for solution analysis. The analyses were carried out within 5 minutes after sample collection to minimise possible changes caused by continued reactions.

### Influence of physicochemical conditions on Cr(VI) removal: effect of electrolyte anion



**Figure S2:** Effect of the electrolyte anion on the changes in chromium and iron concentrations with time for pyrite oxidation experiments at a pH value of 3.5 ( $[Cr(VI)]_0 = 1.8x10^{-4} \text{ M}$ ,  $[FeS_2] = 6 \text{ g.L}^{-1}$ ,  $[NaClO_4]$  or [NaCl] = 0.1 M).

### Pyrite oxidation by Cr(VI) solutions: investigation of the overall reaction stoichiometry

Sulphur species	R calculated
S(0)	1
$S_2O_3^{2-}$	$\frac{7}{3}$
$S_4 O_6^{2-}$	$\frac{8}{3}$
$SO_3^{2-}$	$\frac{11}{3}$
$SO_4^{2-}$	5

**Table S1:** R ratios ([Cr(III)]/[Fe(III)]) calculated from equations (5) to (9) in the paper.

# **Additional references**

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