Supplementary information

Table S1: Coating efficiency* of MHCMPs produced by varying the salt concentration and number of cycles

Salt concentration (M) for single-cycle production	Coating efficiency (%)		No. of cycles with 0.4 M salt solution	Coating efficiency (%)
0.2	7.9 ± 1.1		2	5.0 ± 0.2
0.4	7.2 ± 0.3		3	4.2 ± 0.2
0.6	6.6 ± 0.2		4	3.2 ± 0.1
0.8	7.3± 0.5		5	3.6 ± 0.4
1.0	7.6 ± 0.5		6	4.1 ± 0.7

*Coating efficiency is the ratio of actual mass of incorporated in the MHCMPs to the maximum mass that can be incorporated as per stoichiometry.

The maximum mass of metal hydroxides that could be incorporated was calculated according to the following reaction stoichiometry:

$$FeCl_3 \cdot 6H_2O + AlCl_3 \cdot 6H_2O + 6NH_3 + 6H_2O \rightarrow Fe(OH)_3 \cdot Al(OH)_3 + 6NH_4Cl + 12H_2O.$$

For the conditions giving the maximum incorporated mass in this study (0.4 M salt solutions with 6 cycles), the maximum mass of metal hydroxides that can be incorporated theoretically is about 444 mg.

Table S2: Coating efficiency of different metal hydroxides

Metal hydroxides incorporated	Mass incorporated (mg)	Coating efficiency (%)
$Ni(OH)_2$	5.4	5.8 ± 1
$Co(OH)_2$	5.8	6.2 ± 0.6
$Cu(OH)_2$	5.1	5.2 ± 0.4

Coating efficiency was calculated by the same method as used in the previous table. The stoichiometric reactions used for calculating coating efficiency involved the precipitation reaction for the used metal salts with sodium hydroxide.

Text S1: Recycling of the effluent coming out during the MHCMP production

The excess salt solution and NH_4OH during the MHCMP production from 0.4 M iron-aluminium salt solution were collected as duplicates, each consiting of merged effluent from the production of three MHCMPs. These effluents were collected before the washing step and hence did not consist of the water used for washing of the MHCMPs and thereafter centifuged (1670 g, 5 minutes), which resulted in a pellet and the supernatant.

The supernatant consisted of excess/unreacted (NH_4OH) and ammonium chloride ions generated during the reaction. The pellet from the centrifuged effluent consisted of the metal hydroxides that were formed as a result of the reaction between excess salt solution and NH_4OH. After the supernatants had been decanted for reusing the NH_4OH, the pellet was washed throughly with distilled water to remove any ammonium chloride ions that might be present. The effluents was collected as duplicates, and the pellet from one set of effluent was dried overnight in an oven at 100 °C to check the dry weight. The dry weight of the iron-aluminium hydroxides from the pellet were found to be 166 mg (note that each pellet is a merge of effluent generated during production of three MHCMPs). As per the reaction stoichiometry, this indicates that about 75% of the total metal hydroxides formed from the MHCMP process could be recovered from the effluent. Considering that about 7% of the total metal hydroxides are incorporated in MHCMPs (while using 0.4 M salt solution at a single cycle) it was calculated that about 80% of the total metal hydroxides present in the effluent could be collected. And since the metal hydroxides in the effluent represent the excess iron and aluminium that are generated from the MHCMP production process, it can be said that about 80% of the total iron and aluminium in the effluent could be recollected and reused. Some of the losses could be due to metal hydroxides left in the tubings during the process.

For showing that metal hydroxides collected from the effluent could be reused, the pellet from the other batch of effluent, which was not dried was used, to which 3 ml of 4M HCl was added. The metal hydroxides were solubilized as per the following reaction:

$$Fe(OH)_3 \cdot Al(OH)_3 + 6HCl + 6\,H_2O \rightarrow FeCl_3 \cdot 6H_2O + AlCl_3 \cdot 6H_2O$$

The resluting solution was 4.5 ml in total volume and hence considering that about 166 mg of metal hydroxides was persent in that solution, it resulted in 0.2 M iron-aluminium salt solution which was used for producing another set of MHMCPs as discussed under the results section. Thus, the excess chemicals generated as effluent during MHCMP production process could thereby be reused, and in the case of MHCMPs produced using a single cycle from 0.4 M salt solution, about 80% of the total metal salts used as starting material could be succesfully used for further MHCMP productions. For multiple cycles the excess material present in the effluent will be higher, however by adjusting the addition of HCl as per the stoichiometry, it would still be possible to recycle the metal salts.

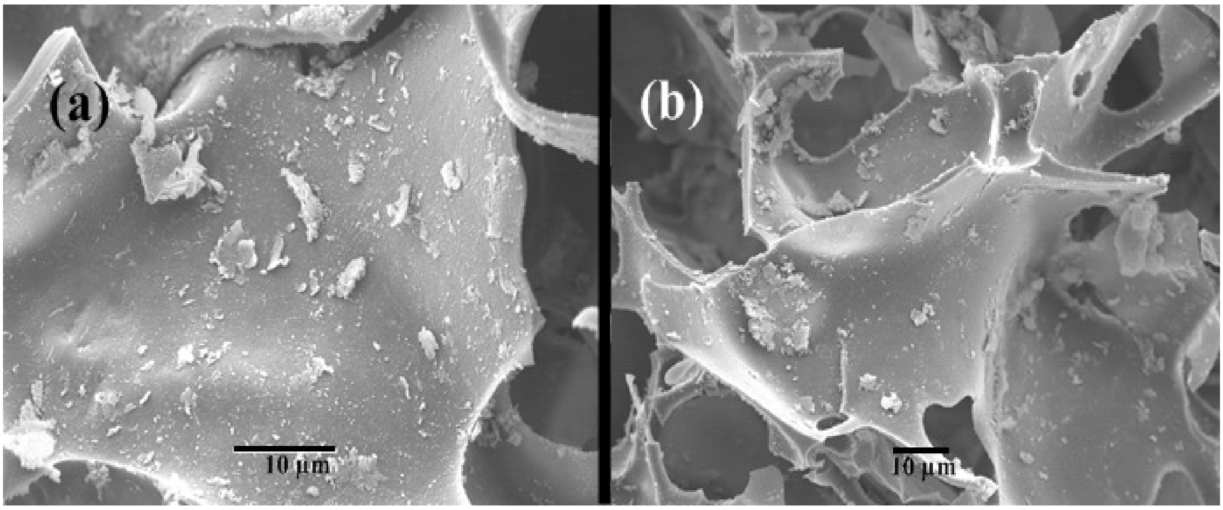

Figure S1: SEM images of two critical-point-dried MHCMPs produced using (a) single cycle with a 1 M salt solution (b) single cycle with a 0.4 M salt solution. It can be seen from the images that the metal hydroxide deposits on the polymer backbone are smooth on some parts and appear as flakes on others.

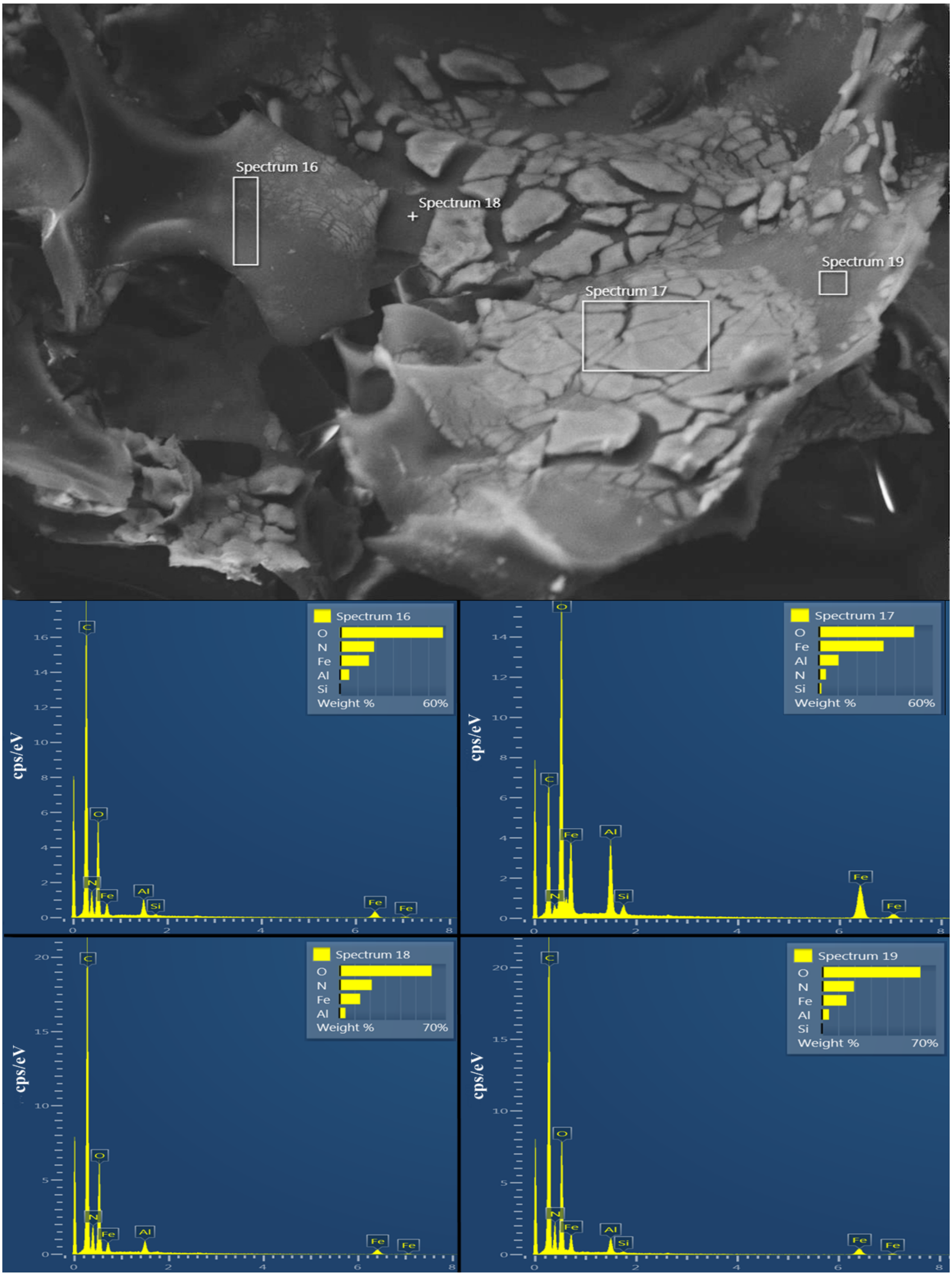

Figure S2: SEM image of a MHCMP prepared in a single cycle using a 1 M salt solution, together with the spectra giving the elemental composition of the corresponding regions in the SEM image. Regions showing patches of coating (spectrum 17 in the SEM image) as well as regions showing smooth coating (spectra 16, 18 and 19) contain Fe and Al.

Figure S3: Photographs of whole MHCMPs containing two different metal hydroxides (upper row), and their cross-sections (lower row): (a) copper (II) hydroxide, (b) iron (III)-aluminium hydroxide. The different MHCMPs were produced using a single cycle with 0.4 M salt solutions.

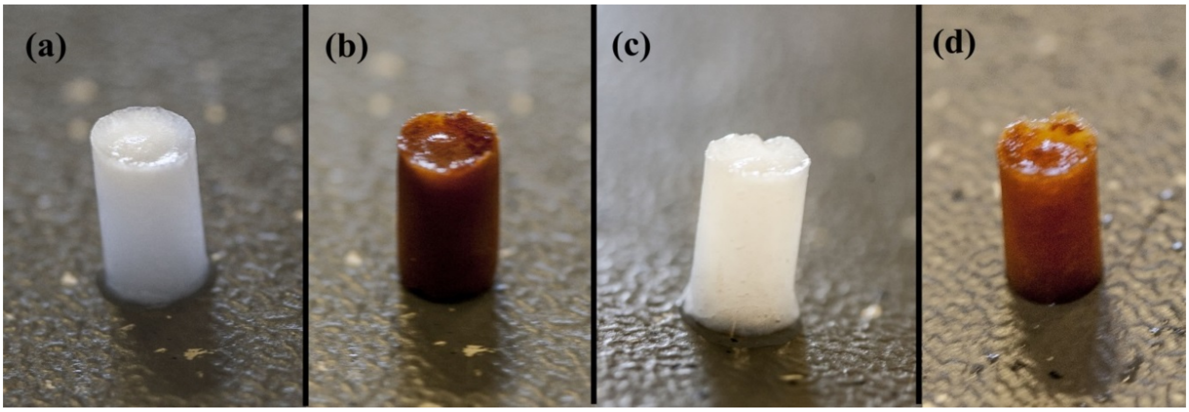

Figure S4: Photographs of (a) unmodified pAAm cryogel (b) MHCMPs produced using a single cycle with a 0.4 M iron and aluminium salt solution (c) pAAm cryogel regenerated using 5 ml of 4 M HCl at 0.5 ml/min (d) MHCMP produced from the regenerated pAAm cryogel shown in c using the same conditions employed for b.

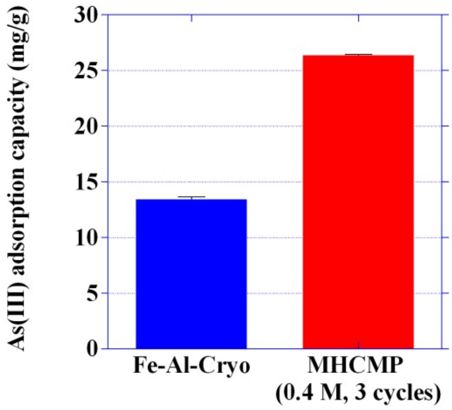

Figure S5: Comparison of adsorption capacities of Fe-Al-Cryo[17] and MHCMP produced from 0.4 M salt solution using 3 cycles, both tested at an initial As(III) concentration of 25 and 23 mg/L respectively.