**JoVE55062R1: Response to Reviewer Comments**

Editorial Comments

***1.6 - how is the leachate solution stored (under what conditions)?***

Response: The leachate is relatively stable since it is acidic, so it can be stored at ambient temperature. It is best not to store in the fridge since some components may precipitate at lower temperature (this is done later under controlled conditions to purify the leachate). To address the particular comment, the last part of step 1.6 was replaced by: “The leachate can be stored at ambient temperature, but it should preferably be further processed (purified and carbonated) shortly afterwards to avoid uncontrolled precipitation of dissolved compounds. The solids can be further processed immediately (wet cake), or be left to dry at ambient temperature if to be processed later.”

***1.11 - Please describe centrifuge speeds as "x g" instead of the machine-dependent "rpm".***Response: that value is actually in g, rather than rpm, that was a mistake. So the centrifuge speed was 2500 g. It has been corrected.

***5.2.7 - please provide a reference for ICP-OES or cite manufacturer's instructions, if applicable.***

Response: The following has been added to 5.2.7: “Solutions are diluted using 2wt% HNO3 diluent, such that the concentration falls in the linear range of the instrument calibration (0-2 mg/L). Yttrium, at 2 mg/L, is added to each diluted sample as an internal standard. The ICP-OES instrument is operated with basis on the manufacturer’s recommendations for analysis of metals in wastewaters.38”

***-The highlighted section of the manuscript exceeds our 2.75 page limit for filmable content. Please reduce the highlighting to no more than 2.75 pages.***

Response: highlighting is now made only to the specific portions of the procedure that will be filmed. The highlighted text alone (deleting the text not highlighted, which is mainly clarification comments) is now only 2 pages in length. The tasks are simple enough to not require extensive filming.

***-Please make sure that your references comply with JoVE’s instructions for authors. For more than 6 authors, list only the first author then et al.):***

Response: The list of reference has been corrected according to JoVE’s instructions. In the two cases that studies were conducted by more than 6 authors, only the first author was listed and then “et al.” was added.

***•Formatting:***

***-Please remove references to the video.***

Response: the reference to the video has been removed.

***-Please use “x g” rather than “rpm” for centrifuge speeds. Also, use “x g” rather than “g” alone to avoid confusion with units of mass.***

Response: this has been done as suggested.

***-References – Please include DOI where available.***

Response: DOI were included where available.

***•Length exceeds 2.75 pg of highlighted material and should be reduced accordingly.***

Response: highlighting is now made only to the specific portions of the procedure that will be filmed. The highlighted text alone (deleting the text not highlighted, which is mainly clarification comments) is now only 2 pages in length. The tasks are simple enough to not require extensive filming.

***•Please copyedit the manuscript for numerous grammatical errors, in particular correct use of prepositions. Some examples are listed below:***

Response: The manuscript has been proof-read to correct grammatical errors, including the corrections described below.

***-1.1, 4.8 – “particles size”***

Response: The particular phrase was replaced by “particle size”

***-1.4, 3.2, 4.2 – Should be “in the proper position”***

Response: “at the proper position” was replaced by “in the proper position” throughout the Protocol section.

***-5.2.2, 5.2.4 – Should be “to 4-5”***

Response: “at 4-5” was replaced by “to 4-5” wherever required.

***-5.2.4 – Please clarify “to avoid accounting the precipitation of metals as adsorption capacity”***

Response: Step 5.2.4 has been re-written to provide more clarity. Essentially, without pH adjustment, nickel is removed from solution both via precipitation and adsorption mechanisms. By adjusting the pH to below the precipitation point of nickel, only adsorbed nickel remains in the solid phase, and mass balance of the liquid phase can be done to determine that adsorbed quantity.

***-Line 450 – “in more readily formation of PCC”***

Response: To correct that, the phrase “However, it presents lower acid dissociation constant than acetic acid, suggesting that the employment of the latter should result in more readily formation of PCC.” Was replaced by the phrase “However, it presents lower acid dissociation constant than acetic acid, suggesting that the precipitation of calcium carbonate should be more readily achievable after employment of acetate solutions as the extraction agent.”

***-Line 455 – “should subjected to”***

Response: The particular phrase was replaced by “should be subjected to”.

***•Additional detail is required:***

***-1.2 – What is attached to the cap?***

Response: this step has been re-written to provide more clarity on the reactor components.

***-1.11 – How much should be placed in each test tube?***

Response: it depends on the size of centrifuge tube/bottle used (it varies by laboratory, and often laboratories will have several), so it is best not to specify as the amount and size of tube/bottle. It is advisable to use larger tubes/bottles, to minimize the number needed and simplify the work. What is more crucial is the centrifuge speed (g’s), time, and what to do afterwards. The section has been revised to improve clarity.

***-3.5, 4.5 – Wash with what?***

Response: In both instances, DI water. The steps have been revised.

***-5.2.1 – Please clarify “activated material”. Is this the material from step 4.8?***

Response: Yes (number have now changed). The phrase “Disperse 1 g of the activated material in 100 ml of the contaminated solution” was replaced by the phrase “Disperse 1 g of the hydrothermally converted material resulting from step 4.6, in 100 ml of the contaminated solution.”

***-5.2.2, 5.2.4 – How is pH monitored?***

The phrase “The solution is continuously stirred, at low speed, using magnetic stirring bar on stirring plate, and the pH is continuously monitored by maintaining a pH electrode soaked in the solution…” was added to these steps.

***-5.2.6 – How much acid is added? To what pH?***

Response: the amount of acid is based on achieving 2wt% concentration, which should lower the pH to less than 2. The following has been added to clarify: “Acidify the solution with HNO3 (to 2 wt% nitric acid concentration), in order to reduce its pH to <2. This step is performed to ensure the ions remain in solution during storage (at ambient temperature) prior to further analysis.”

***-5.2.7 – Please provide a citation for ICP-OES.***

Response: A reference for the ICP-OES method has been provided, as well as a succinct description of the methodology.

1st Reviewer Comments

***-Line 60 – Define PCC.***

Response: The term PCC in the abstract section has been defined.

***-Line 73 – Needs references.***

Response: References have been added for the first phrase of the introduction part.

***-Line 142 – Why using 731 ml of CH3COOH (2M)? Was it calculated for some reason?***

Response: this has been clarified in the discussion section as follows: “The acetic acid-to-calcium molar ratio used in each extraction step was 2:1 (based on the mass of slag, calcium content of the slag and volume of acetic acid solution), meaning the total ratio over two extraction steps was 4:1. Since calcium acetate has an acetate-to-calcium ration of 2:1, double the stoichiometric amount is used, as was found necessary by Chiang et al.5”

***-Line 169 – Is it minimum 10 minutes?***

Response: The phrase “at 2500 rpm for 10 min.” has been replaced by “at 2500 rpm for minimum 10 min.”

***-Line 178 – Not clear.***

Response: The Note part of Protocol’s section 2 has been changed in order to be clearer to the reader.

***-Line 181 – A statement is necessary before starting the procedure for example, the procedure can be summarized as:***

Response: We understand your suggestion, but we believe it is not appropriate to the journal requirements. The note is not meant as an introduction to each step in the procedure, but rather as a note to highlight some aspect of the procedure. Hence, it does not seem appropriate to include the statement suggested. Looking at other JoVE articles, this does not seem to be a common practice.

***-Line 228 – A separate calculation is necessary about the percentage recovery of residue from the slag.***

Response: A statement regarding the amount of residual solids resulting after each Ca-extraction stage was made in the Note part of Protocol’s section 4.

***-4.7 – Not clear! What procedure was adopted in this investigation? Milling or grinding?***

Response: The zeolitic material has small primary particles (as observed microscopically), but they become agglomerated upon drying, hence they are disaggregated rather than milled. The description of this step was changed to “Disaggregate the granular material using a mortar and pestle.”

***-5.2.2 – two 'in order to'; consider for revision.***

Response: the step has been revised. A statement has also been added to the discussion section to justify the pH adjustment: “The pH of the synthetically prepared contaminated solutions of Ni2+ was controlled to 4-5 during the test to, firstly, prevent dissolution of the material in the initial acidic environment of the synthetic solution, and, secondly, to adjust the pH to that typically found in heavy metal remediation conditions.39”

***-5.2.4 – Consider for revision.***

Response: revised for clarity and justification has been moved to discussion section.

***-5.2.7 – Some other procedure can also be used. So, statement should be rewritten considering other options of analysis.***

Response: A statement has been added to suggest alternative forms of measurement: “Alternate techniques for determination of nickel concentration in solution, such as inductively coupled plasma mass spectrometry (ICP-MS) and atomic absorption spectroscopy (AAS), are also suitable for this step.”

***-Line 301 – Replace “collected” with “presented”?***

Response: The word “collected” was replaced by “presented”.

***-Line 303 – Why ICP-MS technique was not used to measure Ni2+ concentration?***

Response: The two phases of the process (carbonation and hydrothermal conversion) were held in different laboratories. That is why two different methodologies were used to measure the elemental composition of the leachate solution (ICP-MS) and the concentration of Ni2+ of the contaminated solutions (ICP-OES). In fact, given the concentration range of the solutions, either technique is suitable in both cases. ICP-MS is more sensitive to very low concentrations, but the concentrations here are of much higher value than the linear calibration range of ICP-MS and ICP-OES, so in both cases the samples undergo dilution prior to analysis.

***-Line 305 – Why only acetic was considered as the leaching agent?***

Response: This is discussed in the second-to-last paragraph of the introduction. In one of our previous works (Hydrometallurgy 147–148 (2014) 90–94), we tested both acetic acid and succinic acid, and found that only the former allowed precipitation of calcium carbonate upon carbonation. In the case of succinic acid, calcium succinate precipitated. There are also other works in literature (Int. J. Greenhouse Gas Control 9, 334–346) that show that oxalic acid is not suitable, due to precipitation of oxalate. An additional reference has been added to this discussion as follows: “Eloneva et al.36 compared sixteen extracts for calcium removal from steelmaking slags, and found acetic acid to be the most efficient (best performance between 0.5M and 2M extractant concentrations) and most successful (highest calcium recovery at ~100%).”

***-Line 319 – Replace “XRD pattern” with “XRD technique”?***

Response: “XRD pattern” was replaced by “XRD technique”

***-Line 320 – What is PSD analysis?***

Response: “PSD analysis” was replaced by “particle size distributions (PSD)”.

***-Line 323 – Chemical formula of Nesquehonite?***

Response: The chemical formula of Nesquehonite is Mg(HCO3)(OH)·2(H2O). The chemical formulas of calcite, nesquehonite and magnesian calcite have been added in brackets.

***-Line 325 – The phrase is not clear.***

Response: The phrase “From the particle size distribution analysis of the material (Figure 2c), it becomes clear that the average particles size is small and the size distribution is within a narrow range.” was replaced by “From the PSD analysis of the material (Figure 2c), it becomes clear that its mean particle size is small and its particle size distribution is narrow”.

***-Line 389 – The material also contains tobermorite gel, which also has ability to adsorb Ni2+.***

Response: “zeolitic minerals” has been replaced by “sorbent material”. Also, the following has been revised/added in the discussion: “Tobermorite is one of the undesirable mineral phases that can form; its layered crystal structure lends to reduce specific surface area,39 a trait important for sorbents, though it has been reported that tobermorite can act as a sorbent through ion exchange mechanism.40”

***-Line 441-452 – Some part of this section should be discussed in previous sections.***

Response: The original paragraph has been moved to the introduction part. Its place in the discussion part is now taken by the following new paragraph: “To this end, among the several extractants investigated in literature,7,8,10,11,34-36 acetic acid was selected as the most suitable for the aim of this study. The particular extraction agent causes the release of high amounts of calcium from the slag to the solution, while retaining the largest part of Si and Al in the resulting residues, promoting that way the parallel formation of PCC and zeolites. The acetic acid-to-calcium molar ratio used in each extraction step was 2:1 (based on the mass of slag, calcium content of the slag and volume of acetic acid solution), meaning the total ratio over two extraction steps was 4:1. Since calcium acetate has an acetate-to-calcium ration of 2:1, double the stoichiometric amount is used, as was found necessary by Chiang et al.5”

***-Line 488-498 – Author should write a few lines considering the cost - effectiveness of the developed process: why this process is suitable than other processes?***

Response: Economics are definitely important to decide whether a new technology is ready for industrial implementation, but this is not something that JoVE is looking for in their articles. The main objective here is to clarify how experiments are performed so that others can implement a technique, verify the results, or continue developing the technology. Another thing to remember, when it comes to waste valorization, is that the economics depend not only on production costs, but also external factors, e.g.: carbon credits for CO2 utilization and avoidance of waste disposal fees. The existence of these can swing a process from being tens or hundreds of dollars/euros per tonne (CO2 or slag) in the red to tens or hundreds in the black. I hope you can understand that this type of analysis better suited elsewhere. The following has been added to the concluding paragraph to touch on the economics: “Economics (the need to pay for virgin materials versus the avoidance of disposal fees for waste materials, or the financial return on utilizing the products for higher or lower value applications) should play a role in the identification of a suitable mineral feedstock. Substitution of other process inputs (acetic acid, sodium hydroxide and concentrated CO2) by less costly or more easily recoverable alternatives should also be considered to improve processing costs.”

2nd Reviewer Comments

***–Is there a better way to store the leachate to ensure the solubilized elements stability? Is storage in a fridge would be recommended?***

Response: please note that the solubility of silica reduces at lower temperatures (this is used as a purification strategy, under controlled conditions), so it is best to keep the leachate stored at room temperature. The acidity is enough, and the amount of excess acetic acid sufficient, to keep elements in solution for long periods of time. Storage in a fridge is more crucial for samples from environmental leaching tests, in water or weak ionic solution, since the elements in that case will have propensity to precipitate in the absence of solids.

**–In the point 1-10, please precise if the term leachate refers to the leachate solution. If yes, please use similar term to keep a uniformity within the text.**

Response: The word “leachates” was replaced by “leachate solution”.

***–Is there any requirements regarding the filters porosity?***

Response: Filters porosities have been added where required.

***–Point 3-5, how much water is needed for washing the carbonates?***

Response: the amount of wash water should be enough to remove soluble compounds, and more washing is better than too little, so it is difficult to provide a specific value. Instead, we have added the following recommendation: “Marked reduction in conductivity of the wash filtrate can be used to confirm wash end-point.”

***–How the authors evaluate the overall carbonation and pcc precipitation or zeolites formation efficiencies? These aspects are missing. For example, mass balance for carbonation/pcc and pore volume measurement for zeolites.***

Response: brief descriptions (since they will not be filmed) of the characterization techniques have been added, with appropriate references for more details and examples. Data on specific surface area, pore volume and pore width have also been included in the representative results section, determined by nitrogen adsorption.

Remarks from authors

* Some revisions were made to the protocol section, especially to clarify the recommended liquid-solid separation techniques for each stage. Some terminology was also modified to
* There was a small error in the calculation of one data point on Figures 4 and 5, which have now been revised. This correction does not change any of the discussion.
* The isotherm equation coefficients in Table 1 had been determined numerically instead of analytically (using the linear equations). The values have now been updated to the analytical solution, which is more accurate. This correction does not change any of the discussion.