



Methods for estimation of long-term non-carbonate neutralisation of acid rock drainage

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ABSTRACT

In the long-term phase of an acid rock drainage (ARD) evolution profile, after any short-term neutralisation capacity provided by carbonate minerals is exhausted, the net acid release is a product of a declining acid generation rate (AGR) and a slower, long-term acid neutralisation rate mainly provided by gangue silicate minerals. At some point, the AGR and the non-carbonate acid neutralisation rate (ANRnc) will be similar. Matching of the AGR and ANRnc near 10 mg H₂SO₄/kg/week is demonstrated in data from 10-year columns. This long-term neutralisation is not measured at present in any accepted assessment tests. Methods to estimate ANRnc, based on silicate mineralogy and solution assays from long-term column leach tests, are compared. Good agreement is demonstrated between rates measured from the solution assay data and those calculated from mineralogy using kinetic databases. More rigorous analysis of the leachate chemistry of selected long-term leach tests also suggests possible cover design criteria based on the maximum AGR that will maintain a pH > 4 in leachate from ARD materials. The data show a distinct break at an AGR of 3 mg H₂SO₄/kg/week, below which no leachate pH is less than 4. The results indicate that an AGR of 10 t H₂SO₄/ha/year is conservative and a suitable cover design target for ARD control that would be matched by ANRnc.

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1. Introduction

In the long-term phase of an acid rock drainage (ARD) evolution profile, the acid generation rate (AGR) declines and the net acid release is a product of continuing but reduced sulfide oxidation, dissolution of previously stored oxidation products (e.g. jarosite) and a slower, long-term acid neutralisation rate mainly provided by gangue silicate minerals. For ARD wastes that are classified as net acid producing in standard acid–base accounting assessment, any short-term neutralisation capacity provided by carbonate minerals (measured in Sobek tests) will be exhausted by the last stages of the ARD profile. At some point, the AGR and the non-carbonate acid neutralising rate (ANRnc) will be similar. A measure of the inherent ANRnc of the main gangue silicate minerals in an ARD waste would therefore provide a major tool in planning cover design and closure.

This long-term neutralisation is not measured at present in any accepted assessment tests. The earlier silicate neutralisation potential

(NP) estimates (static Sobek tests) from Jambor et al. (2000), suggesting that most silicate minerals do not contribute significantly to early NP (largely carbonate-derived), have been supplemented by a new set of estimates for intrusive rocks (Jambor et al., 2006). These results show that some silicate minerals do contribute to early neutralisation but that Sobek estimates of long-term NP are likely to be of questionable value in the management of the ARD profile. Kinetic estimates, while still carrying significant uncertainties, are likely to simulate the long-term acid production/neutralisation balance more closely.

Justification and validation of methods to determine ANRnc, based on silicate mineralogy compared with solution assays from long-term column leach tests, are first presented in this paper. Some of the data have been considered in previous conference papers (Ciccarelli et al., 2008, 2009), and these will be acknowledged where they are used. There are limitations on the estimate from each method but comparison of the values can provide insight to the value of the estimates in ARD management. In this paper we will show that long-term ANRnc can match the remaining AGR in real wastes providing a focus on the estimated matching of ANRnc to AGR for different wastes and potential implications for cover design. Cover design criteria based on oxygen rate (OR) control so that the maximum AGR will maintain a pH > 4 in leachate from ARD materials are discussed.

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2. Methods

2.1. Silicate mineralogy

Potentially the simplest method of estimating ANRnc is to calculate this parameter from the known mineralogy of the waste determined at the time that the AGR is measured. Estimates of the mass fractions of the minerals were obtained using X-ray diffraction (XRD) combined with bulk assay of the whole sample. The XRD was performed with Diffraction Technology Mini Materials Analyser (MMA). Copper K α radiation was used at the conditions of 30 kV and 20 mA. The step size and scanning rate were 0.02° and 0.2°/min, respectively. The phase identification was carried out with Diffraction Technology Traces V6. Quantitative mineralogy was calculated from bulk assay data and X-ray diffraction phase identification. The percentages of each phase were calculated from reconciliation of the two analyses, achieved by linear mixing calculations of mineral chemistry (from XRD phases) and bulk compositions, using a standard matrix software package. A similar process has been described by Paktunc (2001) using the computer program MODAN to solve simultaneous mass balance equations based on whole rock analysis and mineral compositions. This method still directly identifies the minerals from XRD but also infers percentages of known minerals in the sample not specifically identified in the XRD. The presence of these inferred phases (e.g. pyrite at low concentrations) was then verified in SEM/EDS examination of the sample. The uncertainties with this method are that it may not include contributions from unrecognized amorphous or ultrafine secondary mineral products and that XRD will identify the main mineral phases but not their compositional variation. Estimates using mineralogy alone therefore should be validated against other measured parameters if possible. Nevertheless, it is an obvious starting point for estimates of the neutralisation capacity and rates available from non-carbonate minerals.

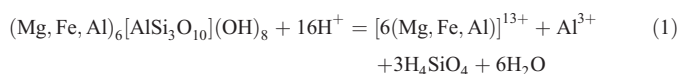
The estimate of ANRnc then requires a database of the kinetics of mineral dissolution as a function of pH. The major 64-page U.S. Geological Survey report from Palandri and Kharaka (2004) with 330 references has compiled dissolution rate parameters, conforming to a general Arrhenius-type rate equation, for over 70 minerals, including phases from all the major classes of silicates, most carbonates, and many other non-silicates. Their rate expression for acidic conditions has the form:

$$\log(\text{rate}) = \log(A) - E / (2.3025RT) - n\text{pH} (\text{mol m}^{-2} \text{s}^{-1})$$

where the parameters A (pre-exponential factor, $\text{mol m}^{-2} \text{s}^{-1}$), E (activation energy, kJ mol^{-1}) and n (reaction order with respect to H^+) are provided in the database.

The compilation covers all of the common minerals likely to be found in ARD wastes and in adjacent non-value parts of most ore deposits. The database provides a set of rates with their pH dependence from which it is possible to estimate the ANRnc from silicates (after steady state dissolution is reached) in four steps.

1. The acid dissolution rate of the mineral at specified pH values is obtained in $\text{mol m}^{-2} \text{s}^{-1}$ from measurement (e.g. Weber et al., 2005) or the compiled data of Palandri and Kharaka (2004).
2. The acid neutralisation factor for each mineral is derived from the stoichiometry of the mineral reaction with acid (i.e. ratio of H^+ to mineral illustrated for chlorite in Eq. (1), i.e. 16) and the adjusted acid neutralisation rate in $\text{mol H}_2\text{SO}_4 \text{ m}^{-2} \text{s}^{-1}$ is calculated from the acid neutralisation factor applied to the acid dissolution rate for each mineral at the specified pH.



3. The long-term rate ANRnc for each mineral in $\text{mg H}_2\text{SO}_4/\text{kg}/\text{week}$ is then calculated from the conversion of $\text{mol H}_2\text{SO}_4$ to $\text{mg H}_2\text{SO}_4$, the area from m^2 to 1 kg of $1.0 \text{ m}^2 \text{ g}^{-1}$ (i.e. 1000) and the time from s to weeks. The estimate assumes a 38–75 μm fraction with specific surface area approximately $1.0 \text{ m}^2 \text{ g}^{-1}$ for each mineral (discussed below).
4. The combined ANRnc in $\text{mg H}_2\text{SO}_4/\text{kg}/\text{week}$ for the waste is then estimated from the mass fractions of the minerals in the mineralogical determination. This represents the long-term neutralisation rate ANRnc from non-carbonate (silicate) minerals in the waste at the pH specified in the calculation.

The assumption of the 38–75 μm fraction and several other qualifications on these estimates have been thoroughly discussed by both Palandri and Kharaka (2004) and Eary and Williamson (2006). For instance, Strömberg and Banwart (1999) have shown that 80% of the reactivity in waste rock occurs in particles less than 250 μm . The leach columns generally use a –3.2 mm size fraction of the waste material including high surface area fines. This may give different dissolution rates, depending on how much fine material is present, but the choice of the 38–75 μm fraction with average surface area of $1 \text{ m}^2/\text{g}$ is reasonable and convenient for these estimates and unlikely to introduce errors of factors greater than two. Eary and Williamson noted that, at the current time, databases of reaction kinetics are sufficiently detailed to allow calculations of the rates of silicate dissolution reactions relative to the rate of pyrite oxidation. They used the data from the Palandri and Kharaka compilation to estimate the rates of reaction for common silicate rocks (i.e. granite, monzonite, granodiorite, diorite and gabbro) found in ARD environments. Their rate calculations point to the importance of the mafic mineral content for neutralising acidity in that minerals, such as anorthite (calcic plagioclase), forsterite, augite, and hornblende, may react rapidly enough to keep up with the rate of acid generation by pyrite oxidation (0–3 wt.%), whereas the felsic minerals, such as K-feldspar and albite, react too slowly to mitigate early acid generation. Comparison of the calculated results with experimental results obtained from the literature for the different rock types has shown that, in general, the calculated results are comparable to the experimental results for the silicic rocks for which neutralisation rates are low but generally higher than the experimental values for the more mafic rock types (Jambor et al., 2006; Eary and Williamson, 2006). They conclude that, overall, kinetic simulation procedures provide a relatively simple and inexpensive means, based on petrological and mineralogical data in the absence of detailed experimental data, to conduct scoping studies of likely pH conditions for non-carbonate-rocks that can be used to optimize sample selection for more detailed experimental testing and estimation of potential impacts from acid mine drainage. We have also taken this as a reasonable starting point for scoping studies of ANRnc compared with AGR from real wastes.

2.2. Estimates from solution assays

An alternative method of estimating ANRnc can be formulated using solution assay data from long-term column leach tests and the acid neutralisation capacity non-carbonate ANCnc ($\text{kg H}_2\text{SO}_4/\text{t}$) formulation first developed by Paktunc (1999) and extended by Weber et al. (2005). The column leach tests were conducted in the EGi laboratories using the standard methodology fully described in the AMIRA/EGi ARD Test Handbook (2002) Appendix F. This test utilises a plastic Buchner funnel 175 mm in diameter and 100 mm high, giving a capacity of about 2.5 L typically holding about 2 to 2.5 kg of –4 mm crushed rock, tailings or sediment. The free draining leach column operation is designed to achieve a weekly wet–dry cycle and a 4-weekly leaching cycle. Heat lamps, maintaining surface temperatures of 30–35 °C, are used to ensure drying of the sample between weekly solution applications. The leach solution is applied weekly (100 mL/kg) and the 4-weekly flush

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