

## APPENDIX

The following is a brief summary of the mass balance equations describing the chemical and volumetric changes of metasomatism. The reader is referred to Gresens (1967), Grant (1986), Brimhall and Dietrich (1987), Brimhall et al. (1988), Ague (1994a), Baumgartner and Olsen (1995), Ague and van Haren (1996), Ague (2011), and Durand et al. (2015) for discussion of various aspects of these equations. The mass change of a mobile constituent can be calculated using Equation 6 for a general system with specified geochemical reference frame, Equation 10 for a constant volume system, or Equation 16 for a constant mass system.

### General system

The basic mass balance expression relevant for describing chemical changes in rocks is (e.g., Brimhall et al. 1988; Ague 1994a):

$$V^0 \rho^0 C_i^0 = V' \rho' C_i', \quad (1)$$

in which  $V$  and  $\rho$  are rock volume and density, respectively (including pore space),  $C_i$  is the concentration of a reference (immobile) species  $i$  defining the geochemical reference frame (mass units), and the  $^0$  and  $'$  superscripts refer to the initial and final (altered) states. The volume strain  $\varepsilon$  is given by:

$$\varepsilon = \frac{V' - V^0}{V^0}. \quad (2)$$

Substitution of Equation 2 into Equation 1 and rearranging for volume strain yields:

$$\varepsilon_i = \left( \frac{C_i^0}{C_i'} \right) \left( \frac{\rho^0}{\rho'} \right) - 1, \quad (3)$$

in which  $\varepsilon_i$  denotes that strain is being computed on the basis of reference element  $i$ . The fractional change in mass for some mobile constituent  $j$  ( $\tau^j$ ) is given by:

$$\tau^j = \frac{V' \rho' C_j' - V^0 \rho^0 C_j^0}{V^0 \rho^0 C_j^0} = \frac{V' \rho' C_j'}{V^0 \rho^0 C_j^0} - 1. \quad (4)$$

Noting that  $\varepsilon_i + 1 = V'/V^0$ , substitution yields:

$$\tau_i^j = \frac{\rho'}{\rho^0} \frac{C_j'}{C_j^0} (\varepsilon_i + 1) - 1, \quad (5)$$

providing a quantitative relationship between changes in rock chemical and physical properties. This expression may be simplified further by substituting explicitly for volume change:

$$\tau_i^j = \left( \frac{C_i^0}{C_i'} \right) \left( \frac{C_j'}{C_j^0} \right) - 1. \quad (6)$$

The total change in rock mass  $T^m$  is:

$$T^{rm} = \frac{V'\rho' - V^0\rho^0}{V^0\rho^0} = \frac{V'\rho'}{V^0\rho^0} - 1. \quad (7)$$

From Equation 1, the right side of Equation 7 can be recast to give:

$$T_i^{rm} = \left( \frac{C_i^0}{C_i'} \right) - 1. \quad (8)$$

Equations 3, 6, and 8 give fractional changes; percentage changes are obtained by multiplying by 100 (gains are positive, losses negative).

### Constant Volume or Known Volume Change System

For a system with no volume change, the basic mass balance of Equation 1 simplifies to:

$$\rho^0 C_i^0 = \rho' C_i'. \quad (9)$$

The mass change for a mobile constituent  $j$  is easily obtained from Equation 5 by setting the volume strain to zero:

$$\tau_i^j = \frac{\rho'}{\rho^0} \frac{C_j'}{C_j^0} - 1. \quad (10)$$

As final and initial volumes are equal, the total rock mass change in a constant volume system is controlled by the density ratio:

$$T^{rm} = \frac{V'\rho' - V^0\rho^0}{V^0\rho^0} = \frac{\rho'}{\rho^0} - 1. \quad (11)$$

If the volume change is non-zero, but known, then the change in mass of a mobile constituent  $j$  can be calculated from Equation 5 by substituting in the known value of volume strain. To calculate the total mass change, we begin by rearranging Equation 2:

$$V' = \varepsilon V^0 + V^0. \quad (12)$$

Substituting this expression for  $V'$  into Equation 7 and rearranging gives the expression for the total mass change in a system with known, non-zero volume change:

$$T^{rm} = \frac{(\varepsilon + 1)\rho' - \rho^0}{\rho^0}. \quad (13)$$

If the volume change is zero, this expression reduces to Equation 11.

### Constant Mass or Known Mass Change System

In systems with no overall rock mass change, the concentrations of a reference species in the initial and final states are equal. Thus, Equation 1 simplifies to:

$$V^0\rho^0 = V'\rho'. \quad (14)$$

It follows that the volume strain expression of Equation 3 can be written as follows for a constant mass system:

$$\varepsilon = \frac{\rho^0}{\rho'} - 1. \quad (15)$$

Rearranging this expression to give  $\varepsilon + 1$  and then substituting the equivalent density ratio into Equation 5 yields the mass change for mobile constituent  $j$ :

$$\tau^j = \frac{C'_j}{C_j^0} - 1. \quad (16)$$

As there is no overall rock mass change,  $T^m = 0$ .

In constant mass systems, 1 kg of initial rock has the same mass of 1 kg after alteration. Note, however, that considerable changes in the masses of individual elements may have taken place. The requirement is that the amount of mass lost must equal the amount gained, to keep the overall mass constant.

If the total change in rock mass  $T^m$  is non-zero, but known, then Equation 8 for the total rock mass change can be rearranged and substituted into Equation 3 for volume strain to yield:

$$\varepsilon = \frac{\rho^0}{\rho'} (T^m + 1) - 1. \quad (17)$$

Similarly, substitution of Equation 8 into Equation 6 yields the following expression for the mass change of a mobile constituent in a system with known overall mass change  $T^m$ :

$$\tau^j = \left( \frac{C'_j}{C_j^0} \right) (T^m + 1) - 1. \quad (18)$$