

Remark 1: The phase stability index (Kulik et al., 2013, eqs 21, 22) in Leal's notation:

Using the (extended) activity (in mole fraction scale),  $z$  factor is normalized by dividing by  $RT$ :

$$a_i^z = a_i \exp(-z_i)$$

the (estimated) mole fraction (i.e. extended activity divided by the extended activity coefficient  $\lambda_i$ ) is

$$x_i^z = \frac{a_i^z}{\lambda_i} = \frac{a_i}{\lambda_i} \exp(-z_i)$$

the stability index of a  $k$ -th phase with  $K$  components (in mole fraction scale) is

$$(1) \quad \Omega_k = \sum_i^K x_i^z = \sum_i^K \frac{a_i^z}{\lambda_i} = \sum_i^K \left[ \frac{a_i}{\lambda_i} \exp(-z_i) \right]$$

or in  $\log_{10}$  form:

$$\Lambda_k = \log_{10} \Omega_k$$

Compare this with eq (21) in the manuscript (replacing  $\Lambda_\pi$  with  $\Omega_k$  in the notation):

$$\Lambda_k = \sum_i^K [x_i \exp(-z_i)]$$

Clearly, in place of mole fraction  $x_i$ , an estimate of the mole fraction via the extended activity and activity coefficient  $\frac{a_i^z}{\lambda_i}$  must be used in the general case, also valid if the mole fraction itself is indefinite (amount of phase is zero) or the phase is metastable.

In this context, the extended activity coefficient  $\lambda_i$  includes all terms of the species primal chemical potential  $\mu_i$  but the mole fraction of species in the phase and the standard chemical potential.

For example, in the case of simple ideal mixing in a condensed phase,  $\lambda_i = \gamma_j$  where  $\gamma_j$  is the familiar activity coefficient due to non-zero enthalpy of mixing.

In the case of a non-ideal gaseous fluid,  $\ln \lambda_i = \mu_i - \mu_i^o - \ln x_j$  and it includes  $\ln P$ , the real fugacity  $\ln f_i^o$  correction of the pure gas, and the fugacity coefficient (due to mixing)  $\ln \gamma_i$ .

In case of aqueous species, the extended activity coefficient involves the molality correction to standard chemical potential and the (Debye-Hueckel) activity coefficient, and so on.

Remark 2: As implemented in GEMS,

$$\ln a_i^z = \eta_i - \mu_i^o$$

where  $\eta_i$  is the dual chemical potential (computed from Lagrange multipliers  $y$  and the species formula) and  $\mu_i^o$  is the standard chemical potential (both normalized by dividing by  $RT$ ). To prove this, recall that, from KKT conditions,

$$z_i = \mu_i - \eta_i \quad \text{where} \quad \mu_i = \mu_i^o + \ln a_i$$

is the primal chemical potential of i-th species. By re-arranging, we obtain

$$\ln a_i = z_i + \eta_i - \mu_i^o$$

from where, defining  $\ln a_i^z = \eta_i - \mu_i^o$ , it follows that

$$\ln a_i = \ln a_i^z + z$$

which is identical to the first equation on this page (eq 28 in the manuscript). It also proves that eq (1) on this page should be used as phase stability index equivalently in GEM and xLMA.