Supplementary Material.

The purpose of this part is to derive expressions for the Kirkwood-Buff integrals (KBIs) in binary systems. For binary mixtures, Kirkwood and Buff [7] obtained the following expressions for the partial molar volumes, the isothermal compressibility and the derivatives of the chemical potential with respect to concentrations

$$v_1 = \frac{1+(G_{22}-G_{12})c_2}{c_1+c_2+c_1c_2(G_{11}+G_{22}-2G_{12})} \tag{A-1}$$

$$v_2 = \frac{1+(G_{11}-G_{12})c_1}{c_1+c_2+c_1c_2(G_{11}+G_{22}-2G_{12})} \tag{A-2}$$

$$kTk_T = \frac{1+c_1G_{11}+c_2G_{22}+c_1c_2(G_{11}G_{22}-G_{12}^2)}{c_1+c_2+c_1c_2(G_{11}+G_{22}-2G_{12})} \tag{A-3}$$

$$\left(\frac{\partial \mu_1}{\partial x_1}\right)_{T,P} = \frac{kT}{x_1(1+x_1c_2(G_{11}+G_{22}-2G_{12}))} \tag{A-4}$$

$$\left(\frac{\partial \mu_1}{\partial c_1}\right)_{T,P} = \frac{kT}{c_1(1+c_1(G_{11}-G_{12}))} \tag{A-5}$$

where P is the pressure, T is the absolute temperature, v_α is the partial molar volume per molecule of species α , μ_1 is the chemical potential per molecule of component 1, k is the Boltzmann constant, k_T is the isothermal compressibility, c_α is the bulk molecular concentration of component α and x_α is the mole fraction of component α. Because $c_1 v_1 + c_2 v_2 = 1$, one can solve Eqs. (A-1, A-3 and A-5) to obtain the following expressions for the KBIs

$$G_{11} = kTk_T - \frac{1}{c_1} + \frac{c_2 v_2}{c_1^2 \mu_{11}^{(c)}} \tag{A-6}$$

$$G_{22} = kTk_T - \frac{1}{c_2} + \frac{v_1^2}{c_2 v_2 \mu_{11}^{(c)}} \tag{A-7}$$

and

$$G_{12} = kTk_T - \frac{v_1}{c_1 \mu_{11}^{(c)}} \tag{A-8}$$

where $\mu_{11}^{(c)} = \frac{1}{kT}\left(\frac{\partial \mu_1}{\partial c_1}\right)_{T,P} = \left(\frac{\partial \ln a_1}{\partial c_1}\right)_{T,P}$, a_1 is the activity of component 1 and k is Boltzmann's constant. Eq. (A-6) was derived by Zimm [9] (there is a misprint in Zimm's paper (eq. (10)) where the sign is (-) instead of (+) before the third term in the left hand side of eq. (A-6).

The KBIs can be also expressed in terms of $\mu_{11}^{(x)} = \frac{1}{kT}\left(\frac{\partial \mu_1}{\partial x_1}\right)_{T,P} = \left(\frac{\partial \ln a_1}{\partial x_1}\right)_{T,P}$.

Eqs. (A-1, A-3 and A-4) provides the following expressions for the KBIs

$$G_{11} = kTk_T - \frac{1}{c_1} + \frac{c_2 v_2^2 (c_1 + c_2)^2}{c_1^2 \mu_{11}^{(x)}} \tag{A-9}$$

$$G_{22} = kTk_T - \frac{1}{c_2} + \frac{v_1^2(c_1 + c_2)^2}{c_2\mu_{11}^{(x)}} \tag{A-10}$$

and

$$G_{12} = kTk_T - \frac{v_1 v_2(c_1 + c_2)^2}{c_1\mu_{11}^{(x)}} \tag{A-11}$$

These expressions are the same as those obtained by Matteoli and Lepori [10].