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A corrected mixture law for B/A

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A derivation is presented that corrects an expression for the effective acoustic nonlinearity parameter of a mixture of immiscible liquids. The derivation is based upon a mass fraction, rather than volume fraction, formulation.

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Recent interest in applying measurements of the acoustic nonlinearity parameter B/A of biological materials to infer tissue composition^{1,2} has led to a re-examination³ of the underlying mixture laws for B/A . In his 1983 paper,⁴ Apfel derived a relation that gives the effective nonlinear parameter of a system of immiscible liquids, given the densities, compressibilities, B/A values, and volume fractions of the components. For a system of n components, he showed that

$$\beta = \sum_{i=1}^n \beta_i X_i \quad (1)$$

and

$$\beta^2 \frac{B}{A} = \sum_{i=1}^n X_i \beta_i^2 \left(\frac{B}{A} \right)_i, \quad (2)$$

where β_i and X_i are the compressibility and volume fraction of component i , respectively, and the unsubscripted values refer to effective properties of the mixture. Equation (1) is just the familiar mixture law for compressibility that Chamberlé⁵ has shown can be derived directly from considerations of conservation of mass. Equation (2) was derived by considering the effect of applying a pressure increment to the mixture, but its derivation fails to consider that the relative volume fractions themselves may change due to the application of the pressure increment. Although the contribution of this change in volume fraction with pressure may be taken into account explicitly,⁶ we present an alternative derivation that avoids this difficulty by formulating the problem in terms of mass fractions.

Let us define Y_i as the mass fraction of the i th component of an n -component mixture:

$$Y_i = M_i / M_{\text{tot}}, \quad (3)$$

where M_i is the mass of component i and M_{tot} is the total mass of the mixture. Note that Y_i and X_i are related to one another by $X_i = (\rho / \rho_i) Y_i$, where ρ is the density of the mixture and ρ_i is the density of the i th component. Let us now consider the intensive quantity $\nu = 1/\rho$, the specific volume (volume per unit mass) of an n -component mixture.

If we assume that the total volume of the mixture equals the sum of the component volumes (i.e., we assume the components are not interactive and are not mutually soluble), it follows that

$$\nu = \sum_{i=1}^n \nu_i Y_i. \quad (4)$$

If Eq. (4) holds, so do its partial derivatives with respect to pressure p (the entropy s is held constant in all differentiations). Note that while the component volume fractions of a mixture may change with pressure, the mass fractions do not: $\partial Y_i / \partial p = 0$. The result of differentiating equation (4) is, therefore,

$$\left(\frac{\partial \nu}{\partial p} \right)_s = \sum_{i=1}^n \left(\frac{\partial \nu_i}{\partial p} \right)_s Y_i, \quad (5)$$

and likewise to higher orders,

$$\left(\frac{\partial^2 \nu}{\partial p^2} \right)_s = \sum_{i=1}^n \left(\frac{\partial^2 \nu_i}{\partial p^2} \right)_s Y_i. \quad (6)$$

Substituting $\rho = 1/\nu$ into the left-hand side of Eq. (5) and noting that the sound speed $c^2 = (\partial p / \partial \rho)_s$ yields

$$\left(\frac{\partial \nu}{\partial p} \right)_s = \left(\frac{\partial (1/\rho)}{\partial p} \right)_s = -\frac{1}{\rho^2} \left(\frac{\partial \rho}{\partial p} \right)_s = -\left(\frac{1}{\rho c} \right)^2. \quad (7)$$

Since Eq. (7) must be true for the mixture *and also for each component individually*, Eq. (5) becomes

$$\left(\frac{1}{\rho^2 c^2} \right) = \sum_{i=1}^n \frac{Y_i}{\rho_i^2 c_i^2}.$$

Making use of the expressions $\beta = (\rho c^2)^{-1}$ and $X_i = (\rho / \rho_i) Y_i$ results in an expression relating the compressibility of the mixture to that of the components, viz.,

$$\beta = \sum_{i=1}^n \frac{\rho Y_i \beta_i}{\rho_i} = \sum_{i=1}^n X_i \beta_i, \quad (8)$$

which is just Eq. (1).

Similarly, we can use Eq. (6) to arrive at a mixture law for B/A . Using the relation $B/A = 2\rho c (\partial c / \partial p)_s$, the left-hand side of Eq. (6) can be written as:

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$$\begin{aligned} \left(\frac{\partial^2 v}{\partial p^2}\right)_s &= \left(\frac{\partial(-1/\rho^2 c^2)}{\partial p}\right)_s = \frac{2}{\rho^3 c^2} \left(\frac{\partial \rho}{\partial p}\right)_s + \frac{2}{\rho^2 c^3} \left(\frac{\partial c}{\partial p}\right)_s \\ &= \frac{2}{\rho^3 c^4} \left(1 + \rho c \frac{\partial c}{\partial p}\right) \\ &= \frac{2}{\rho^3 c^4} \left(1 + \frac{B}{2A}\right). \end{aligned} \quad (9)$$

Since Eq. (9) must be true for both the mixture and its components, Eq. (6) becomes

$$\frac{2}{\rho^3 c^4} \left(1 + \frac{B}{2A}\right) = \sum_{i=1}^n \frac{2Y_i}{\rho_i^3 c_i^4} \left[1 + \left(\frac{B}{2A}\right)_i\right],$$

where all quantities on the left-hand side are properties of the mixture as a whole. Using $\beta = (\rho c^2)^{-1}$ and the definition $K \equiv 1 + B/2A$ results in the desired relation

$$\beta^2 K = \sum_{i=1}^n \frac{\rho Y_i \beta_i^2 K_i}{\rho_i} = \sum_{i=1}^n X_i \beta_i^2 K_i. \quad (10)$$

This expression is similar to Eq. (2), but relates the B/A value of the mixture to those of the components through K , the coefficient of nonlinearity. Actually, Apfel first reported⁷ the result given in Eq. (10), but then later incorrectly modified it, substituting B/A for K . The coefficient of non-

linearity K contains information about nonlinearities arising from convection,⁸ as well as those from the equation of state of the material via B/A . Future methodologies which use mixture laws for the nonlinearity parameter should take the corrected relation, Eq. (10), into account.

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⁶One of the authors, Zhe-ming Zhu, has independently derived the result given in Eq. (10) by taking the pressure derivative of the effective compressibility [Eq. (8)] and noting that the effective B/A is given by $d(1/\beta)/dp - 1$ (private communication).

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