Table I
Effect of Temperature on the Reduced Volumes for Several Values of Energy Parameters

RT, cal	parameters $v_{21} = v_{12}$, cal/cm^3	$\tilde{V}$ calcd from eq 13	$\tilde{V}$ calcd from eq 8 in ref 1	deviation,[a] %
600	10	1.2131	1.2079	0.4
	20	1.2173	1.2098	0.6
	−10	1.2404	1.2338	0.5
	−20	1.2840	1.2702	1.1
	−30	1.3358	1.3209	1.1
800	10	1.3281	1.3199	0.6
	20	1.3399	1.3206	1.5
	−10	1.3797	1.3688	0.8
	−20	1.4767	1.4371	2.8
	−30	1.6452	1.5516	6.0

[a] Defined as $100(\tilde{V}_{13} - \tilde{V}_8)/\tilde{V}_8$.

Table II
Specific Volumes of Two Binary Mixtures Containing Polyisobutylene at 25 °C

solvent	segment fraction of polymer	exptl specific vol, cm^3/g	calcd specific vol,[a] cm^3/g	abs error, %[c]	calcd specific vol,[b] cm^3/g	abs error, %[c]
benzene	0.1989	1.1297	1.1312	0.13	1.1310	0.12
	0.4355	1.1202	1.1223	0.19	1.1219	0.15
	0.5923	1.1135	1.1156	0.19	1.1151	0.14
cyclohexane	0.1971	1.2267	1.2268	0.008	1.2253	0.11
	0.3731	1.1979	1.1981	0.017	1.1959	0.17
	0.5445	1.1700	1.1702	0.017	1.1679	0.18

[a] Values calculated from eq 13. [b] Values calculated from Renuncio's equation. [c] Defined as $100|V_e - V_c|/V_e$.

Moreover, the specific volumes of benzene–polyisobutylene and cyclohexane–polyisobutylene mixtures have been calculated with the ν_{21} and ν_{21} values quoted in ref 1. The results are shown in Table II together with the experimental data reported in the literature.[6] For the benzene–polyisobutylene system, eq 13 and Renuncio's equation give the same absolute errors, but for the cyclohexane–polyisobutylene system, eq 13 is far superior, because the deviation from experiment is about ten times smaller than that associated with Renuncio's equation.

References and Notes

(1) J. A. Renuncio and J. M. Prausnitz, *Macromolecules*, 9, 898 (1976).
(2) D. S. Abrams and J. M. Prausnitz, *AIChE J.*, 21, 116 (1975).
(3) E. A. Guggenheim, "Mixtures", Clarendon Press, Oxford, 1952, p 217.
(4) J. H. Hildebrand and R. L. Scott, "The Solubility of Nonelectrolytes", Dover Publications, New York, N.Y., 1964, p 125.
(5) B. E. Eichinger and P. J. Flort, *Trans. Faraday Soc.*, 64, 2035 (1968).
(6) J. A. Renuncio and J. M. Prausnitz, *Macromolecules*, 9, 324 (19976).

Reply to the Note by V. Brandani

J. M. PRAUSNITZ* and J. A. R. RENUNCIO

Department of Chemical Engineering, University of California, Berkeley, California 94720.
Received August 21, 1978

We are most grateful to Vincenzo Brandani for having accepted the challenge with which we ended our admittedly speculative paper on nonrandomness: "These remarks. . .are intended. . .to stimulate reexamination of fundamental concepts in solution thermodynamics".

Regrettably, we erred in failing properly to take into account the consequences of the density dependence in our definitions of local site fractions [eq 7 of our paper]. As a result of this density dependence, "parameter" P^* [eq 14] is also density dependent. As a result, the equation of state for a mixture is not the same as that of Flory but contains an additional term, as shown correctly in Brandani's equation 14. Similarly, the residual entropy in the nonrandom treatment is different from that in Flory's random treatment.

Fortunately, our overall conclusions are not destroyed by Brandani's corrections. On the contrary, Brandani's calculations suggest that the effect of nonrandomness is perhaps more important than we claimed; with his corrected results, he obtains significantly better agreement with experiment than we did.

While we are thankful to Brandani for having found our mathematical error and having worked out the correct results, it is clear that the empirical flavor of our nonrandom "theory" remains. As before, it is our hope that the (now happily corrected) approximation which we have presented has called attention to the importance of partial order in liquid mixtures, and that it may stimulate research toward a more rigorous answer to the problem of nonrandomness in mixtures of dense fluids.

0024-9297/78/2211-1294$01.00/0