

Volumetric, Viscometric and Refractive Indices Properties of Binary Mixtures of Acetyl Acetone with 1-Butanol at Different Temperatures

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Abstract

Measurements of thermodynamic and transport properties have been effectively involved in understanding the nature of molecular interactions and physico-chemical behaviour in mixtures. As well as, they are necessary for the designing and optimizing of the industrial equipment. In this paper, experimental densities, ρ , viscosity, η , and refractive indices, n^D , of binary mixtures of acetyl acetone (acac) with 1butanol (Bu) were determined at temperatures of 298.15, 303.15 and 318.15 K and atmospheric pressure over the entire composition range of mixtures. The experimental values of mixtures and pure liquids have been used to calculate the excess volume, V^E , deviation in viscosity, $\Delta\eta$, excess Gibb's free energy of activation for viscous flow, ΔG^{*E} , and deviation in molar refraction, ΔR . The computed results were fitted to the Redlich–Kister polynomial equation to evaluate the adjustable parameters (A_i) and standard deviation (σ). The V^E , $\Delta\eta$ and ΔG^{*E} values of the mixtures have been found to be negative over the whole composition. The negative magnitude suggests the presence of strong intermolecular interaction between unlike molecules in the binary liquid mixtures. A comparative study of these properties together with those available in the literature has been also discussed.

Keywords: Binary mixture; molecular interactions; excess properties; Redlich-Kister equation.

1. Introduction

The studies of intermolecular interactions and the internal structures of mixed binary liquid mixtures remain as significant area of interest in physical chemistry [1, 2]. Physicochemical properties of liquid–liquid mixtures v_{iz} ; density, viscosity, refractive index, surface tension and the thermodynamic behavior of binary mixtures have been studied extensively. These studies were done in order of understanding of the nature of the interactions, and interpreting the interactions between molecules of a mixture in terms of the excess mixing parameters such as excess molar volume, excess viscosity and deviations in refractivity and surface tension [3, 6]. These parameters can be calculated from the direct measurements of density, viscosity, refractive index and surface tension of the mixtures, as well as, pure components at different temperatures. Such data find extensive application in solution theory and molecular dynamics for

both fundamental research and engineering applications in many chemical industries, engineering disciplines and other related areas [6, 7]. Furthermore, in recent years, interest in surface tension and viscosity have rapidly increased because this property plays an important role in the design of contacting equipment with which some chemical processes are carried out, like gas absorption distillation, extraction [8]. The intention of this study is to provide a set of volumetric and transport data in order to assess the influence of temperature and molecular structure on the behavior of mixtures of alcohol (1-butanol) and acetyl acetone. In the present work we report experimental density, viscosity and refractive index data for the binary mixtures of acetyl acetone with 1butanol at (T = 298.15, 303.15, 308.15 K) and at atmospheric pressure in order to provide information about molecular interactions between an acetyl acetone and 1-butanol. From the experimental values, the corresponding derived properties $(V^E, \Delta \eta,$



 ΔG^{*E} and ΔR), were computed and correlated by means of the Redlich-Kister [9] equation. Up to our knowledge, no full data of density viscosity are available in literature for this system.

2. Material and Methods

The analytical grade 1-butanol and acetyl acetone obtained in its highest purity of 99% as claimed by the manufacturer (from alfa Aesar or by Merck) and, thus, no further purification was done. The purity of chemicals was checked by comparing with their densities, and refractive index values with the literature data (Table 2.1). Precautions were taken, such as cooling the chemicals before preparation of samples and reducing to a minimum the vapor space in the vessels, to avoid losses by evaporation during manipulation and after preparation. During the measurements, the mixtures were transferred to the pycnometer, viscometer, or stalagmometer by taking extreme care to avoid external contamination. Mixtures were prepared by mass in glass stoppered bottles. A set of twenty-one compositions were prepared for each system and their physical properties were measured on the same day. All molar quantities were based on the IUPAC relative atomic mass table. Densities of liquids and their mixtures were obtained by using a pycnometer having a bulb volume of 25 cm^3 and diameter of 1mm. the triplicate measurements were performed, and the average of these values was considered in all calculations. The mass measurements were done on a single-pan digital balance to an accuracy of 0.0001 g. Refractive indices were measured using the sodium D line refractometer. The average of three readings was used to calculate the refractive index. In all physical property measurements, a thermostat was used.

2.1. Theory

The excess molar volume of mixing, V^E , representing the non-ideal behavior were derived from equation 2.1

$$V^{E} = V_{m} - \sum_{1}^{2} V_{i} X_{i}$$
 (2.1)

Where V_m refers to molar volume of the mixture which can be calculated from the mixture density, Standard deviation for each case is calculated by ρ_m and the mixture molecular weights as $(V_m = M_m / \rho_m)$ for each temperature; in $(cm^3.mol^{-1})$.

Molar mass of mixture can be calculated from:

$$M = X_{acac} M_{acac} + (1 - X_{acac}) M_{BuOH}$$
(2.2)

The viscosity deviations (excess viscosity, $\Delta \eta$) were calculated using

$$\Delta \eta = \eta_{12} - x_1 \eta_1 - x_2 \eta_2 \tag{2.3}$$

Where η_{12} is the viscosities of the mixture and x_1, x_2 and η_1 , η_2 are the mole fraction and the viscosities of pure components 1 and 2, respectively. Furthermore, the Molar Gibbs free energy of activation of viscous flow can be calculated using the following equation

$$G^{*E} = RT \left[ln \left(\frac{\eta V_m}{\eta_2 V_2} \right) - x_1 ln \left(\frac{\eta_1 V_1}{\eta_2 V_2} \right) \right]$$
(2.4)

Where V_m is the molar volume of the mixture, V_1 and V_2 is the molar volume of the pure component 1 and 2, R is the gas constant, T is the Kelvin temperature, and η is the dynamic viscosity of the mixture, respectively. η_1 and η_2 is the dynamic viscosity of the pure component 1 and 2. x_1 is the mole fraction of component 1.

Refractive indices have been used for the calculation of molar refraction (R_m) obtained by using Lorenz-Lorenz [13, 14] equations 2.5 and 2.6

$$R_m = \left[\frac{n_D^2 - 1}{n_D^2 - 2}\right] \sum \frac{x_i M_i}{\rho_m} \tag{2.5}$$

$$R_i = \left[\frac{n_D^2 - 1}{n_D^2 - 2}\right] \sum \frac{M_i}{\rho_i} \tag{2.6}$$

Where n_D refers to refractive index, R_m is molar refraction of the mixture, R_i is molar refraction of the i^{th} component. Deviation in molar refraction (ΔR) is calculated by equation

$$\Delta R = R_m - (x_1 R_1 + x_2 R_2) \tag{2.7}$$

Where R_1 and R_2 are the molar refractivities of pure components 1 and 2, respectively. R_m is the molar refractivity of the mixtures. All the deviations, (V^E) , $\Delta \eta$, ΔG^{*E} and ΔR), have been fitted to Redlich-Kister [9] polynomial regression (equation 2.8) of the type

$$Y_m^E = x_1 (1 - x_1) \sum_{i=0}^n a_i (2x_1 - 1)^i$$
 (2.8)

$$\sigma = \left[\frac{\sum \left(\Delta Y_{expt} - \Delta Y_{cald}\right)^2}{m - n}\right]^{0.5}$$
(2.9)



Table 2.1: Comparison of experimental values of densities and viscosities of pure liquids with literature values at 298.15, 303.15 and 308.15 K

Component	T (K)	$ ho~({ m g}~/~{ m cm}^3)$		η (n	nPa.s)
		Exp.	Lit.	Exp.	Lit.
	298.15	0.975105	0.969420 [7]	0.756032	0.737 [10]
Acetyl acetone	303.15	0.961046	0.96600 [10]	0.686377	0.702 [10]
	308.15	0.948100	0.96000 [10]	0.679603	0.679 [10]
	298.15	0.803771	0.805500 [11]	2.057800	2.578 [6]
1-Butanol	303.15	0.802265	0.8022 [3, 4]	1.793465	1.916
	308.15	0.801227	0.794000 [12]	1.257244	1.78321 [12]

Mole fraction	Density, $ ho~({ m g}~/{ m ml})$			Visc	Viscosity, $\eta~({\rm mPa}~.~{\rm s})$			
X_{acac}	298.15 K	303.15 K	308.15 K	298.15 K	303.15 K	308.15 K	index	
0	0.803771	0.802265	0.801227	2.057800	1.793465	1.257244	1.3970	
0.03988	0.812525	0.808958	0.798257	1.585634	1.342912	1.090761	1.4005	
0.07500	0.817200	0.813500	0.805537	1.426000	1.196300	1.038200	1.4030	
0.132832	0.828321	0.823126	0.818100	1.153980	0.988365	0.92038	1.4060	
0.18300	0.836100	0.832300	0.821007	1.059000	0.915300	0.854400	1.4085	
0.230442	0.845697	0.842113	0.831600	0.949274	0.824467	0.817174	1.4105	
0.27800	0.853700	0.852900	0.836306	0.854800	0.781700	0.763900	1.4140	
0.334141	0.865148	0.859937	0.843800	0.747515	0.730600	0.711643	1.4175	
0.37700	0.87200	0.865800	0.853745	0.742200	0.698300	0.661500	1.4195	
0.427724	0.88390	0.875292	0.863200	0.728573	0.648574	0.613127	1.4235	
0.477655	0.890704	0.883978	0.869663	0.679697	0.612921	0.604260	1.4260	
0.534178	0.898376	0.894800	0.877573	0.637466	0.601785	0.570373	1.4295	
0.58500	0.907800	0.901900	0.888589	0.618800	0.587100	0.552700	1.4315	
0.625566	0.922667	0.913743	0.892700	0.605318	0.581895	0.549813	1.4355	
0.68800	0.926700	0.918100	0.904100	0.575600	0.558000	0.540600	1.4367	
0.730838	0.934100	0.926900	0.913000	0.541394	0.533729	0.525581	1.4420	
0.79300	0.942400	0.939700	0.918300	0.537700	0.526800	0.517800	1.4435	
0.844434	0.944858	0.939012	0.924200	0.550519	0.536725	0.512006	1.4475	
0.89500	0.958700	0.951000	0.932800	0.576300	0.557400	0.534800	1.4495	
0.955826	0.961786	0.955936	0.944000	0.623772	0.589100	0.564795	1.4520	
1.00000	0.975105	0.961046	0.948100	0.756032	0.686377	0.679603	1.4520	



Where m is the number of data points and n is the number of coefficients.

3. Results and Discussion

The experimental values of densities and viscosities of the acetyl acetone and 1-butanol are compared with the literature values and are presented in Table 2.1. Density, viscosity and refractive index of the binary mixture and pure solvent at (T = 298.15, 303.15 and 308.15 K) are reported in Table 2.2. For all the mixtures, results of densities and refractive index increased with the mole fraction of 1-butanol over the investigated range of temperature. However, the results of viscosities tend to decrease with an increase in concentration of 1-butanol in the mixture.

3.1. Excess Properties of Mixtures

The experimentally obtained Results for molar mass, molar volume, excess molar volumes and viscosities deviations of all binary mixtures as a function of mole fraction and temperature are reported in Tables 3.1 and 3.2. The plots of excess molar volume (V^E) , deviations in viscosity $(\Delta \eta)$ and Excess Gibbs' free energy of activation of flow (ΔG^{*E}) against mole fraction of acetyl acetone are given in Figure 3.1 (a, b and c respectively). The excess molar volumes (V^E) included in Table 3.2 show that the values are negative over the entire mole fraction range for all binary mixtures studied in the present work. This can be explained in terms of contraction in volume that comes from dipole-induced dipole and dipole-dipole interactions and donor-acceptor interactions (hydrogen bonding) between unlike molecules. The excess molar volume curves (Figure 3.1) are not symmetric. The V^E values for the systems are completely negative over the entire composition range, having minima at $x_1 = 0.7$.

The negative values of V^E suggest strong interactions between the unlike molecules of the binary liquid systems. It is known that, alcohols are selfassociated through hydrogen bonding [15]. When two components of acetyl acetone and 1-butanol are mixed, the hydrogen bonding in alcohols are broken, but strong hydrogen bonding (as in Figure 3.2) and dipole-dipole interactions between acetyl acetone with 1-butanol are formed. Because the interaction strength of the hydrogen between unlike

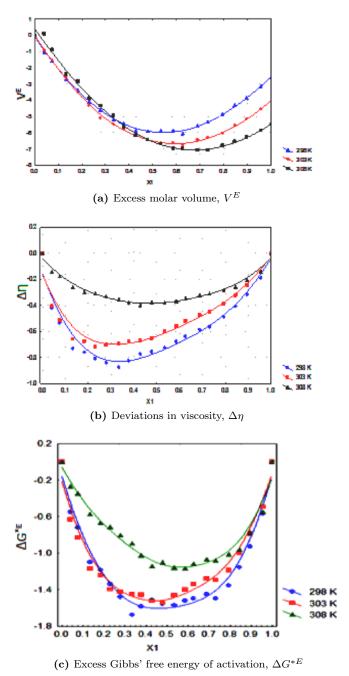


Figure 3.1: Experimental excess properties at 298.15, 303.15 and 308.15 K $\,$



Table 3.1: Molar mass, molar volume and excess Gibbs' free energy of activation at 298.15, 303.15 and 308.15, 323.15 K

Mole fraction	$rac{Molar\ mass}{(g/mol)}$	Molar volume (cm 3 / mol)			Δ	G^{*E} (KJ / mo	ol)
X_i	М	$298.15~\mathrm{K}$	$303.15~\mathrm{K}$	$308.15~\mathrm{K}$	$298.15 { m K}$	$303.15~\mathrm{K}$	$308.15~\mathrm{K}$
0.0000	74.1200	92.21534	92.38841	92.50817	66.62011	67.39625	67.60203
0.03988	75.15687	92.49787	92.90577	94.15127	65.98186	66.68146	67.28335
0.07500	76.07000	93.08615	93.50953	94.43394	65.73466	66.40654	67.16456
0.132832	77.57362	93.65163	94.24267	94.82169	65.22524	65.94519	66.86658
0.18300	78.87800	94.34039	94.77112	96.07467	65.03058	65.76579	66.7097
0.230442	80.11150	94.72841	95.13157	96.33417	64.76973	65.51205	66.60253
0.27800	81.34800	95.28874	95.37812	97.27064	64.52461	65.38438	66.45466
0.334141	82.80766	95.71500	96.29506	98.1366	64.20337	65.23817	66.29589
0.37700	83.92200	96.24083	96.93001	98.29865	64.19927	65.14081	66.11301
0.427724	85.24081	96.43715	97.38556	98.74978	64.1584	64.96652	65.93027
0.477655	86.53904	97.15800	97.89727	99.50868	64.0048	64.83728	65.91256
0.534178	88.00863	97.96413	98.35564	100.2864	63.86634	64.80285	65.7847
0.58500	89.33000	98.40273	99.04646	100.5301	63.80377	64.75825	65.71032
0.625566	90.38471	97.96023	98.91701	101.2487	63.73802	64.73252	65.71515
0.68800	92.00800	99.28564	100.2157	101.7675	63.64659	64.65974	65.68496
0.730838	93.12179	99.69145	100.4658	101.9954	63.5049	64.55399	65.61854
0.79300	94.73000	100.5284	100.8173	103.1667	63.50865	64.52987	65.60958
0.844434	96.07529	101.6823	102.3153	103.9551	63.59531	64.61405	65.60026
0.89500	97.39000	101.5855	102.408	104.4061	63.70634	64.71155	65.72289
0.955826	98.97146	102.9039	103.5335	104.8427	63.93442	64.87844	65.87332
1.00000	100.12000	102.6762	104.1782	105.6007	64.40539	65.2791	66.36564

molecules is stronger than the hydrogen bond between 1-butanol molecules and the dipole interaction, the V^E values are negative for all binary mixtures of acetyl acetone with 1-butanol. The solutions studied show a negative excess of the viscosity $\Delta \eta$ in the whole range of 1-butanol concentrations. A negative excess of the viscosity of a mixture means that its resistance to the flow is smaller than that of the pure components. The intermolecular structures existing in pure liquids are partially destroyed by a mixing process, which makes the flow easier than that of the pure liquids. A review of (Figure 3.1) reveals that, the ΔG^{*E} values are completely negative over the entire range of composition at all the studied temperatures. The negative ΔG^{*E} values become more negative as the temperature increases for acetyl acetone–1-butanol binary mixture. The negative ΔG^{*E} values indicate that dispersion forces are dominant; furthermore, the existence of dispersion forces indicates that the component molecules have different molecular sizes and shapes.

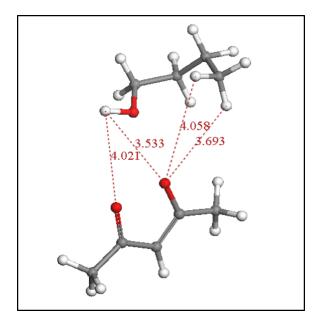


Figure 3.2: Intermolecular forces of hydrogen bonds formed between unlike molecules

In summary, excess values may be affected by:



Table 3.2: Molar mass, molar volume and excess Gibbs' free energy of activation at 298.15, 303.15 and 308.15, 323.15 K

Mole fraction	Excess molar volume (cm ³ / mol)			Ε	Excess viscosity		
X_i	298.15 K	303.15 K	308.15 K	298.15 K	303.15 K	308.15 K	
0	0	0	0	0	0	0	
0.039880	-1.03105	-0.92525	0.086276	-0.42025	-0.4064	-0.14345	
0.075000	-1.56996	-1.51323	-0.89398	-0.53417	-0.51413	-0.17572	
0.132832	-2.68829	-2.59663	-2.40673	-0.7309	-0.65804	-0.26013	
0.183000	-3.36146	-3.49237	-2.8473	-0.76058	-0.67557	-0.29714	
0.230442	-4.08951	-4.32411	-3.88585	-0.80854	-0.71388	-0.30696	
0.278000	-4.58864	-5.11171	-4.34721	-0.84111	-0.70399	-0.33276	
0.334141	-5.19644	-5.50369	-4.96032	-0.87531	-0.69294	-0.35259	
0.377000	-5.44661	-5.7746	-5.63201	-0.82483	-0.67779	-0.37797	
0.427724	-5.86706	-6.16654	-6.1648	-0.77243	-0.67136	-0.39705	
0.477655	-5.91374	-6.41765	-6.4394	-0.75631	-0.65174	-0.37707	
0.534178	-5.88264	-6.63525	-6.69471	-0.72496	-0.6003	-0.37831	
0.585000	-5.87636	-6.62525	-6.99801	-0.67747	-0.55872	-0.36662	
0.625566	-6.0455	-6.77364	-6.99461	-0.63814	-0.51901	-0.34608	
0.688000	-5.57459	-6.44179	-7.07067	-0.58658	-0.47379	-0.31923	
0.730838	-5.33222	-6.3215	-7.06900	-0.56502	-0.45063	-0.3095	
0.793000	-4.83830	-6.02541	-6.80174	-0.4878	-0.38874	-0.28137	
0.844434	-4.30582	-5.52885	-6.55642	-0.40802	-0.32188	-0.25746	
0.895000	-3.85961	-5.15691	-6.28801	-0.31642	-0.24522	-0.20546	
0.955826	-3.12187	-4.54107	-5.86842	-0.18976	-0.14618	-0.14032	
1.000000	0	0	0	0	0	0	

- i. Specific forces between molecules, such as hydrogen bonds, charge transfer complexes, breaking of hydrogen bonds [15, 16].
- ii. The physical intermolecular forces, including electrostatic forces between charged particles.
- iii. The structural characteristics of the component arising from geometrical fitting of one component in to other structure due to the differences in shape and size of the components and free volume [16].

3.2. Optical Properties

The deviations of refractive indices, ΔR , were calculated from the experimental densities and refractive indices equation 2.5, 2.6 and 2.7. The dependences of the molar refraction, ΔR , on the mole fraction of 1-butanol at room temperature (303.15 K) are presented in Table 3.4 and the so-obtained ΔR data are graphically represented in Figure 3.3.

It has been observed that the refractive index deviation shows positive values for the entire mole fraction. And such values are due to the electronic perturbation of the individual molecules during mixing

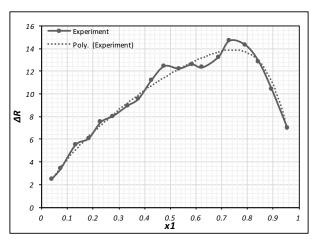


Figure 3.3: The refractive index deviation at 303.15 K (-), Redlich–Kister polynomial fit

and therefore depend very much on the nature of the mixing molecules [16]. It may be noted that The binary system is characterized by a slightly asymmetrical ΔR - x_1 curve, shifted towards lower mole fractions of acetyl acetone, over the entire concentration range (Figure 3.3).



Table 3.3: Molar mass, molar volume and excess	s Gibbs' free energy of activation at 298.15, 303.15 and 308.15, 323.15 $\rm K$
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Mole fraction	ΔR	Exces	Excess free energy ΔG^{*E}			
X_i	303.15 K	298.15 K	$303.15 { m K}$	$308.15 { m K}$		
0.00000	-0.0025865	0.00011	0.00025	3E-05		
0.039880	2.4540616	-0.54981	-0.63011	-0.26936		
0.075000	3.3751363	-0.71922	-0.83069	-0.34474		
0.132832	5.4672796	-1.10054	-1.1696	-0.57124		
0.183000	6.0229021	-1.18408	-1.2428	-0.66611		
0.230442	7.4710448	-1.33984	-1.3961	-0.71464		
0.278000	8.0025028	-1.47962	-1.42309	-0.80373		
0.334141	8.9101204	-1.67651	-1.45045	-0.89311		
0.377000	9.5215262	-1.58568	-1.45708	-1.02302		
0.427724	11.178044	-1.51419	-1.52399	-1.14306		
0.477655	12.411187	-1.55719	-1.54752	-1.09906		
0.534178	12.190016	-1.57046	-1.4623	-1.15706		
0.585000	12.560864	-1.52046	-1.39931	-1.16862		
0.625566	12.293477	-1.49635	-1.33916	-1.11365		
0.688000	13.22358	-1.44949	-1.27976	-1.06667		
0.730838	14.654062	-1.49629	-1.29483	-1.08014		
0.793000	14.269919	-1.35486	-1.18735	-1.01227		
0.844434	12.823151	-1.15427	-0.99428	-0.95802		
0.895000	10.371634	-0.93124	-0.78973	-0.77289		
0.955826	6.9711812	-0.56843	-0.49408	-0.54728		
1.00000	-0.0213192	0.00039	0.0001	-0.00036		

3.3. Redlich-Kister Equation

Table 3.4 has been obtained from Redlich-Kister equation 2.8 by using the best-fit values of coefficient A_i . The variations of smoothed values of V^E , $\Delta \eta$, ΔG^{*E} , and Δn , with mole fraction, x_1 , of acetyl acetone. The Redlich–Kister polynomial equation provided a satisfactory correlation of the binary data, Table 3.4.

4. Conclusion

In this paper, the densities, viscosities and reactive index of the binary liquid mixture of acetyl acetone + 1-butanol has been experimentally determined in temperature range (298.15 to 308.15 K) and for the entire composition range. For the densities, the agreement was found between the measured and the literature values. some differences were shown in the viscosity data for the 1-butanol. The excess molar volumes, V^E , deviations in viscosity, $\Delta \eta$, and excess Gibbs' free energy of activation flow, ΔG^{*E} , have been calculated from the experimental values at three temperatures. The sign and magnitude of these quantities have been discussed in terms of the molecular interactions between the mixing components. Negative, V^E , negative $\Delta \eta$, negative ΔG^{*E} and positive ΔR are observed for acetyl acetone and 1-butanol binary mixtures. The excess molar volumes and viscosity deviations are computed and fitted to Redlich-Kister equation.

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Table 3.4: Coefficients of Redlich-Kister equation and standard deviation for excess molar volumes and viscosity deviations

T (K)	Property	A_0	A_1	A_2	A_3	A_4	A_5	σ
	V^E	32.633	-106.9	129.12	-74.0	41.07	-24.48	0.998
298.15	$\Delta \eta$	6 E-07	-4E-05	9E-03	-0.013	0.10	-0.45	0.984
	ΔG^{*E}	41.107	-118.51	148.5	-105.73	47.431	-12.87	0.974
	V^E	-98.580	295.02	-336.95	178.43	-22.58	-19.58	0.998
303.15	$\Delta \eta$	83.160	-263.70	328.9	-204.89	67.65	-11.11	0.986
	ΔG^{*E}	114.23	-351.69	430.55	-266.63	90.51	-16.99	0.983
	ΔR	-617.66	1675.9	-1827.9	1005.1	-299.46	65.11	0.982
	V^E	-284.98	864.64	-1010.6	568.55	-139.19	-4.38	0.994
308.15	$\Delta \eta$	37.810	-113.31	131.17	-74.03	22.20	-3.86	0.989
	ΔG^{*E}	114.23	-351.69	430.55	-266.63	90.51	16.99	0.983

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