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Investigations on the Thermophysical Properties of Binary Systems of Fatty Acid Esters + Dimethyl Carbonate

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Abstract

Carbonate esters are used as additives in biodiesel to improve its thermophysical properties for better applicability. In this work, the experimental densities, dynamic viscosities and refractive indices for binary mixtures of dimethyl carbonate and four fatty acid esters (methyl decanoate, ethyl decanoate, methyl laurate, ethyl laurate) have been measured and presented over the complete concentration range at temperatures range from 293.15 to 323.15 K under normal pressure. The excess molar volumes and the viscosity deviations of the four binary mixtures were calculated with the measured data and correlated with the Redlich-Kister equation. The excess Gibbs energy of activation was also calculated. From the values of excess properties of the binary mixtures and its deviations indicate that the changes of molecular interactions in the mixtures. The experimental results could be useful for the study of biodiesel-carbonate blended fuels.

1 Introduction

Thermophysical properties of liquid mixtures have important practical application values in many engineering fields, especially in fuel systems [1, 2]. The density of fuel can have a significant effect on the quality of fuel injection in engines, since the injection systems, pumps, and injectors must deliver precisely adjusted amounts of fuel to provide proper combustion [3]. Viscosity is the most important property of biodiesel since it affects the fluidity of the fuel and injection behavior [4, 5]. Viscosity affects the operation of fuel injection equipment, especially at low temperatures. If the viscosity is too high that may lead to less accurate operation of the fuel injectors and poorer atomization of the fuel spray [6]. Therefore,

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Thermophysical properties of blended biodiesel are important to design transfer heat and mass equipment and to interpret the kind of interactions that occur in the studied blends.

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the essential physical properties of fuels such as density and viscosity of the blends are the important physicochemical characteristics of fuel related to fuel storing, transportation and utilization. These properties of blended fuels should be investigated that can provide important information for predictive and diagnostic engine combustion models.

Biodiesels are methyl or ethyl esters of long chain fatty acids that can be produced from renewable and biodegradable resources. It is biologically degradable that is less hazardous for the environment. The advantageous of biodiesel are that it has higher cetane number, higher combustion efficiency, contains no sulfur, safe in handling, non-toxic, etc. [7, 8]. Biodiesels are also easy to mix with normal diesel fuel. However, besides these advantages, biodiesels also have some disadvantages for using in engine such as high viscosity, low volatility and low heating value, which may lead to problems in pumping, atomization of fuel systems [9, 10]. Therefore, it is generally less appropriate for use in lower temperature. It is comparatively less inflammable than the normal diesel. One solution to alleviate this problem is to use add oxygenated compounds to a biodiesel to form blended fuels, examples are ethers, alcohols, esters, glycols, acetates, and carbonates [11–13]. Dimethyl carbonate (DMC) has been a green solvent with versatile applications due to its environmental-friendly attributes. It also be can an oxygenated fuel or additive for fuels and has a higher oxygen content than alcohols and excellent compatibility with biodiesel and diesel [14, 15]. DMC is considered as a promising additive for fuel because of its high oxygen content, no carbon–carbon atomic bonds, and excellent solubility. DMC additive can improve the contact of fuel with oxygen during combustion and emission performances. Adding DMC to fuels can solve the problem of hypoxia caused by local fuel-rich zones that can appear in the combustion process [16, 17]. Furthermore, DMC is insoluble in water, which reduces the pollution of groundwater. Razzaq et al. investigated the effect of palm biodiesel blends with DMC on compression ignition engines [14]. Ren et al. investigated the densities, surface tensions, and viscosities of the blends of diesel + DMC [18]. Liu et al. investigated the effect of DMC on density and viscosity of n-butanol as alternative to gasoline [19]. However, thermodynamics properties of fatty acid ethyl esters with DMC are still scarce in the literature.

In order to investigate the effects of the thermodynamics properties of biodiesel blended with DMC, the densities, dynamic viscosities and refractive indices of four binary mixtures of DMC and four fatty acid esters (methyl decanoate, ethyl decanoate, methyl laurate, ethyl laurate) have been measured in the temperature range from 293.15 to 323.15 K with an interval of 5 K and $p = 0.1$ MPa in the present work. The excess properties and deviations of the tested mixtures are calculated and fitted to the Redlich-Kister equation. These experimental results are expected to supplement the information for the DMC-biodiesel blending fuels.

2 Experimental Section

2.1 Chemicals Used

The samples of dimethyl carbonate, ethyl decanoate, methyl decanoate, ethyl laurate and methyl laurate were supplied by Aladdin Chemistry Co., Ltd., Shanghai, China. The reagents in this work were used without further purification. Table 1 lists the detailed information about the five samples of pure compounds.

Table 1 Specifications of the chemicals in this work

Compound	Source	CAS	Provided mass fraction purity	Purification
Dimethyl carbonate	Aladdin	616-38-6	≥ 0.990	None
Ethyl decanoate	Aladdin	110-38-3	≥ 0.990	None
Ethyl laurate	Aladdin	106-33-2	≥ 0.990	None
Methyl decanoate	Aladdin	110-42-9	≥ 0.990	None
Methyl laurate	Aladdin	111-82-0	≥ 0.990	None

2.2 Method

In this work, the samples were prepared by the weighing method with an analytical balance (Mettler Toledo AL204) and the stated precision is 0.0001 g. The combined standard uncertainty of the mole fraction is estimated to be 0.0002.

Densities (ρ) of pure components and their mixtures were determined with a DMA 4500 M densimeter (Anton Paar) with digital vibrating tube at $T = $ (293.15 to 323.15) K and atmospheric pressure. The densitometer can automatically balance the temperature and is stable with a precision of 0.03 K. The densimeter can do self-checks and be calibrated with dried air and double-distilled water before determination. The uncertainty of densities of mixtures is related to the purity of pure compounds and measurement accuracy (± 0.00005 g·cm^{-3}). The relative standard uncertainty of ρ is 0.0005.

The dynamic viscosities (η) of all samples at $T = $ (293.15 to 323.15) K and atmospheric pressure were determined using an Anton-Paar AMVn viscometer in which a stainless steel ball rolls under gravity down a thermostated glass capillary. The temperature of the measurement cell is stable within ± 0.01 K. The calibration of the viscometer was carried out with freshly twice distilled water. The relative uncertainty of η is 0.010 [20].

Refractive indices (n_D) of all samples at $T = $ 293.15, 303.15, 313.15 and 323.15 K and atmospheric pressure were determined using a WAY-2 S refractometer (Shenguang). The measurement temperatures were controlled by circulating water bath with an accuracy of ± 0.02 K. The refractometer was calibrated with double-distilled water at each temperature. The presented refractive index values were the average values of three measurements. The uncertainty of the refractive indices is 0.0005.

2.3 Experimental Results

Densities (ρ), viscosities (η) and refractive indices (n_D) have been measured for the pure components and four binary mixtures (DMC + methyl decanoate), (DMC + ethyl decanoate), (DMC + methyl laurate) and (DMC + ethyl laurate) in the temperature range from 293.15 to 323.15 K. The experimental values of ρ, η and n_D of the binary liquid mixtures were provided in Tables 2, 3 and 4, respectively.

Table 2 Densities (ρ) for the binary mixtures of (DMC + methyl decanoate), (DMC + ethyl decanoate), (DMC + methyl laurate) and (DMC + ethyl laurate) at T = (293.15 − 323.15) K and p = 0.1 MPa[a]

x_1	ρ/(g·cm^{-3})						
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
DMC (1) + methyl decanoate (2)							
0.0000	0.8722	0.8681	0.8640	0.8599	0.8558	0.8517	0.8476
0.1013	0.8800	0.8758	0.8716	0.8674	0.8631	0.8589	0.8547
0.1993	0.8885	0.8842	0.8799	0.8755	0.8712	0.8669	0.8625
0.3003	0.8988	0.8943	0.8899	0.8854	0.8809	0.8765	0.8720
0.4003	0.9106	0.9061	0.9014	0.8968	0.8922	0.8876	0.8829
0.5004	0.9247	0.9200	0.9152	0.9104	0.9055	0.9007	0.8959
0.6005	0.9418	0.9369	0.9318	0.9268	0.9218	0.9168	0.9116
0.7004	0.9628	0.9576	0.9523	0.9470	0.9417	0.9363	0.9310
0.8001	0.9891	0.9836	0.9780	0.9723	0.9667	0.9610	0.9553
0.9000	1.0234	1.0174	1.0113	1.0052	0.9992	0.9930	0.9869
1.0000	1.0697	1.0632	1.0566	1.0499	1.0432	1.0365	1.0298
DMC (1) + ethyl decanoate (2)							
0.0000	0.8641	0.8601	0.8560	0.8519	0.8479	0.8438	0.8397
0.1001	0.8714	0.8673	0.8631	0.8589	0.8548	0.8506	0.8464
0.2000	0.8799	0.8756	0.8713	0.8670	0.8628	0.8585	0.8542
0.3000	0.8897	0.8854	0.8809	0.8765	0.8721	0.8677	0.8633
0.3999	0.9014	0.8969	0.8923	0.8878	0.8832	0.8786	0.8740
0.4999	0.9155	0.9108	0.9060	0.9013	0.8965	0.8917	0.8869
0.6002	0.9325	0.9276	0.9226	0.9176	0.9127	0.9076	0.9026
0.7000	0.9539	0.9487	0.9435	0.9382	0.9330	0.9277	0.9224
0.8008	0.9819	0.9763	0.9708	0.9651	0.9595	0.9539	0.9483
0.8998	1.0183	1.0123	1.0063	1.0003	0.9942	0.9881	0.9820
1.0000	1.0697	1.0632	1.0566	1.0499	1.0432	1.0365	1.0298
DMC (1) + methyl laurate (2)							
0.0000	0.8693	0.8654	0.8615	0.8576	0.8536	0.8497	0.8458
0.1001	0.8760	0.8720	0.8680	0.8640	0.8600	0.8559	0.8519
0.2000	0.8837	0.8796	0.8755	0.8714	0.8672	0.8631	0.8589
0.2932	0.8921	0.8879	0.8836	0.8794	0.8751	0.8709	0.8666
0.3999	0.9036	0.8992	0.8948	0.8904	0.8860	0.8816	0.8772
0.5001	0.9168	0.9122	0.9076	0.9030	0.8984	0.8938	0.8892
0.5998	0.9329	0.9281	0.9233	0.9185	0.9137	0.9088	0.9040
0.6995	0.9533	0.9482	0.9432	0.9381	0.9330	0.9278	0.9227
0.8002	0.9804	0.9750	0.9695	0.9641	0.9586	0.9531	0.9476
0.8997	1.0168	1.0109	1.0050	0.9990	0.9930	0.9870	0.9810
1.0000	1.0697	1.0632	1.0566	1.0499	1.0432	1.0365	1.0298
DMC (1) + ethyl laurate (2)							
0.0000	0.8621	0.8582	0.8543	0.8504	0.8465	0.8426	0.8387
0.1002	0.8686	0.8646	0.8606	0.8566	0.8526	0.8486	0.8446
0.1999	0.8760	0.8719	0.8678	0.8638	0.8596	0.8555	0.8514
0.2999	0.8849	0.8807	0.8765	0.8722	0.8680	0.8638	0.8595
0.4000	0.8955	0.8912	0.8868	0.8825	0.8781	0.8737	0.8693

Table 2 (continued)

x_1	$\rho/(\text{g·cm}^{-3})$						
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
0.4999	0.9085	0.9040	0.8994	0.8949	0.8904	0.8858	0.8812
0.5999	0.9248	0.9200	0.9153	0.9105	0.9057	0.9009	0.8961
0.7000	0.9457	0.9407	0.9356	0.9306	0.9255	0.9204	0.9153
0.8000	0.9733	0.9680	0.9626	0.9572	0.9517	0.9462	0.9408
0.8988	1.0116	1.0057	0.9998	0.9939	0.9880	0.9820	0.9760
1.0000	1.0697	1.0632	1.0566	1.0499	1.0432	1.0365	1.0298

[a]Standard uncertainties u are $u(p) = 0.20$ kPa, $u(T) = 0.03$ K, $u(x_1) = 0.0002$, $u_r(\rho) = 0.0010$.

3 Results and Discussion

The experimental and literature [21–43] values of ρ, η and n_D for the pure components are listed in Table 5. In order to compare the experimental and literature data, the absolute average relative deviations (AARD) are calculated according to Eq. 1. The results are also listed in Table 5.

$$\text{AARD} = \frac{100}{n} \sum_{i=1}^{n} \frac{|Y_{\text{lit}} - Y_{\text{exp}}|}{Y_{\text{lit}}} \tag{1}$$

In the above equation, Y_{lit} is the value in the literature, and Y_{exp} is the measured data in this work.

The excess molar volumes (V_m^E) and viscosity deviations ($\Delta\eta$) for four binary systems of DMC + fatty acid esters are calculated from the experimental data according to the following relations.

$$V_m^E = (M_1 x_1 + M_2 x_2)/\rho_m - (M_1 x_1/\rho_1 + M_2 x_2/\rho_2) \tag{2}$$

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \tag{3}$$

where x_1 stands for the mole fractions of DMC in a binary mixture and x_2 is the mole fractions of fatty acid ethyl ester; M_1 and M_2 stand for the molar masses of DMC and a fatty acid ethyl ester; ρ_m, ρ_1 and ρ_2 are the densities of the binary mixture, DMC and a fatty acid ethyl ester, respectively. η, η_1 and η_2 represent the dynamic viscosity of the measured viscosity of the mixture, DMC and a fatty acid ethyl ester, respectively.

The excess properties of the binary mixed solution will show the difference between the properties of the solution itself and that of the hypothetical solution under ideal conditions [25]. Figure 1 shows the trends of V_m^E for the four binary systems of DMC with methyl decanoate, ethyl decanoate, methyl laurate and ethyl laurate, respectively. The solid lines in Fig. 1 are the calculated results from Eq. 4. It can be observed that the V_m^E of the four binary mixed solutions are positive over the entire molar fraction range and the V_m^E increases with temperature. At the same temperature, the excess volume of the binary mixed solution first increased with the increase of the mole fraction of DMC, and then decreased after reaching the maximum. The maximum value appears at about x_1(the mole fraction of DMC) ≈ 0.6. The positive deviation for the V_m^E of the binary system is thought

Table 3 Dynamic viscosities (η) for the binary mixtures of (DMC + methyl decanoate), (DMC + ethyl decanoate), (DMC + methyl laurate) and (DMC + ethyl laurate) at $T = (293.15 - 323.15)$ K and $p = 0.1$ MPa[a]

x_{DMC}	η/(mPa·s)						
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
DMC (1) + methyl decanoate (2)							
0.0000	2.125	1.906	1.735	1.588	1.454	1.342	1.240
0.1013	1.963	1.765	1.610	1.475	1.354	1.251	1.159
0.1993	1.804	1.625	1.489	1.366	1.256	1.165	1.081
0.3003	1.644	1.485	1.365	1.255	1.156	1.076	0.999
0.4003	1.484	1.348	1.240	1.143	1.057	0.985	0.917
0.5004	1.330	1.210	1.114	1.029	0.956	0.895	0.834
0.6005	1.175	1.070	0.991	0.919	0.857	0.805	0.750
0.7004	1.027	0.938	0.871	0.811	0.759	0.715	0.669
0.8001	0.881	0.814	0.761	0.709	0.665	0.631	0.593
0.9000	0.748	0.694	0.653	0.610	0.576	0.549	0.518
1.0000	0.637	0.592	0.556	0.521	0.496	0.474	0.446
DMC (1) + ethyl decanoate (2)							
0.0000	2.330	2.122	1.923	1.744	1.598	1.475	1.358
0.1001	2.148	1.958	1.779	1.616	1.483	1.373	1.265
0.2000	1.962	1.791	1.629	1.485	1.367	1.267	1.171
0.3000	1.778	1.624	1.484	1.356	1.250	1.161	1.078
0.3999	1.597	1.464	1.339	1.228	1.133	1.055	0.983
0.4999	1.417	1.301	1.198	1.101	1.022	0.954	0.887
0.6002	1.243	1.148	1.060	0.978	0.910	0.851	0.794
0.7000	1.080	0.999	0.927	0.858	0.801	0.752	0.702
0.8008	0.919	0.856	0.796	0.739	0.692	0.653	0.614
0.8998	0.773	0.720	0.673	0.628	0.592	0.561	0.528
1.0000	0.637	0.592	0.556	0.521	0.496	0.474	0.446
	DMC (1) + methyl laurate (2)						
0.0000	3.153	2.787	2.504	2.257	2.046	1.870	1.707
0.1001	2.844	2.523	2.274	2.057	1.874	1.711	1.575
0.2000	2.542	2.269	2.053	1.868	1.705	1.560	1.443
0.2932	2.274	2.038	1.854	1.688	1.546	1.425	1.319
0.3999	1.988	1.789	1.634	1.490	1.371	1.269	1.177
0.5001	1.735	1.565	1.432	1.316	1.213	1.127	1.049
0.5998	1.489	1.350	1.235	1.142	1.057	0.981	0.920
0.6995	1.243	1.133	1.050	0.977	0.903	0.843	0.791
0.8002	1.014	0.932	0.869	0.811	0.757	0.710	0.667
0.8997	0.810	0.752	0.705	0.659	0.618	0.584	0.551
1.0000	0.637	0.592	0.556	0.521	0.496	0.474	0.446
x_{DMC}	DMC (1) + ethyl laurate (2)						
0.0000	3.370	3.011	2.698	2.443	2.211	2.023	1.836
0.1002	3.062	2.744	2.460	2.232	2.026	1.859	1.690
0.1999	2.731	2.463	2.219	2.016	1.834	1.693	1.539
0.2999	2.422	2.179	1.982	1.803	1.649	1.527	1.390
0.4000	2.123	1.920	1.748	1.594	1.463	1.363	1.247

Table 3 (continued)

x_{DMC}	$\eta/(mPa \cdot s)$						
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
0.4999	1.845	1.674	1.532	1.396	1.289	1.203	1.103
0.5999	1.578	1.435	1.318	1.209	1.118	1.050	0.963
0.7000	1.313	1.206	1.116	1.024	0.951	0.896	0.828
0.8000	1.064	0.983	0.913	0.844	0.786	0.744	0.690
0.8988	0.836	0.777	0.724	0.677	0.636	0.604	0.561
1.0000	0.637	0.592	0.556	0.521	0.496	0.474	0.446

[a]Standard uncertainties u are $u\,(p) = 0.20$ kPa, $u\,(T) = 0.03$ K, $u\,(x_1) = 0.0002$, and $u_r\,(\eta) = 0.010$.

to be caused by the expansion of the volume of the solution, as a result of the expansive effects associated with the breaking of the homo-molecular interactions [29, 44]. DMC is a polar molecule. Due to the presence of polar C=O bonds and C–O bonds in DMC molecules, DMC molecules exhibit dipole–dipole interactions. Fatty acid ethyl or methyl esters are generally considered to be nonpolar molecules due to the presence of predominantly nonpolar hydrocarbon chains in their structures. The polar ester functional group contributes some degree of polarity but the overall molecule is still relatively nonpolar. This means that during mixing the fatty acid ester molecules with DMC can weaken the dipole–dipole interactions in the case of DMC. The positive values of V_m^E are the result of a combination of the disruption of the dipole–dipole interactions between molecules and structural differences. An increase in system temperature leads to an increase in the volumes of the component molecules and the excess volume value increases with the increasing temperature. Figure 2 shows a comparison of V_m^E for (DMC + methyl caprate, DMC + ethyl caprate, DMC + methyl laurate, DMC + ethyl laurate) at 293.15 K. Comparing the mixture of two fatty acid methyl esters and fatty acid ethyl esters with DMC, it is found that the excess volume of the binary system composed of DMC and fatty acid methyl esters is smaller than that of the binary system composed of DMC and fatty acid ethyl esters at the same temperature and composition. At the same temperature and composition, the V_m^E of fatty acid ester mixtures increases with the increase of carbon chain. This indicates that the values of V_m^E for the binary systems are affected by the carbon chain length of fatty acid ethyl esters, and the increase of chain length of esters leads to the increase of the values of V_m^E. In addition, the V_m^E of the binary mixture of (DMC + ethyl ester) is slightly larger than that of (DMC + methyl ester).

Figure 3 displays the dependence of viscosity deviations as the molar fraction of DMC. A negative excess viscosity is observed for all binary systems from the curves of $\Delta\eta - x_1$ for entire composition range. The minima of the ($\Delta\eta - x_1$) curve for all the four binary mixtures occur at $x_1 \approx 0.5$–0.7. Moreover, the values of $\Delta\eta$ become less negative with increasing temperatures, which means the absolute values of viscosity deviation decrease. The negative values of the viscosity deviations indicate that attractive forces among molecules are stronger than the repulsion forces [45]. A comparison of the viscosity deviation of the studied mixtures as a function of the mole fraction of DMC at 293.15 K is exhibited in Fig. 4. It indicates that the values of $\Delta\eta$ become more negative with increasing alkyl ester chain length. The chain length of ester affects the viscosity deviations of the binary mixtures. The viscosity deviations for two systems of DMC + methyl or ethyl dodecanoate are

Table 4 Refractive indices (n_D) for the binary mixtures of (DMC + methyl decanoate), (DMC + ethyl decanoate), (DMC + methyl laurate) and (DMC + ethyl laurate) at T = (293.15 – 323.15) K and $p = 0.1$ MPa[a]

x_1	n_D				x_1	n_D			
	293.15 K	303.15 K	313.15 K	323.15 K		293.15 K	303.15 K	313.15 K	323.15 K
DMC(1) + methyl decanoate(2)					**DMC (1) + ethyl decanoate (2)**				
0.0000	1.4258	1.4214	1.4165	1.4123	0.0000	1.4255	1.4214	1.4164	1.4122
0.1013	1.4233	1.4185	1.4139	1.4101	0.1001	1.4236	1.4192	1.4144	1.4098
0.1993	1.4215	1.4167	1.412	1.4079	0.2000	1.4214	1.4168	1.4120	1.4080
0.3003	1.4183	1.4138	1.4091	1.4048	0.3000	1.4183	1.414	1.4090	1.4050
0.4003	1.4149	1.4098	1.4054	1.4015	0.3999	1.4150	1.4106	1.40595	1.4014
0.5004	1.4107	1.4056	1.4011	1.3965	0.4999	1.4105	1.4059	1.4014	1.3975
0.6005	1.405	1.3996	1.3948	1.3922	0.6002	1.4058	1.4009	1.3959	1.3921
0.7004	1.3985	1.3934	1.38835	1.3858	0.7000	1.399	1.3946	1.3896	1.3859
0.8001	1.3910	1.3857	1.3808	1.3774	0.8008	1.3911	1.3863	1.3818	1.3772
0.9000	1.3811	1.376	1.3714	1.3676	0.8998	1.3815	1.3771	1.3716	1.3673
1.0000	1.3692	1.3641	1.3594	1.3543	1.0000	1.3692	1.3641	1.3594	1.3543
DMC (1) + methyl laurate (2)					**DMC (1) + ethyl laurate (2)**				
0.0000	1.4323	1.4280	1.4233	1.4191	0.0000	1.4314	1.4274	1.4229	1.4186
0.1001	1.4302	1.4255	1.4211	1.4167	0.1002	1.4291	1.4249	1.4204	1.4165
0.2000	1.4275	1.4228	1.4183	1.414	0.1999	1.4264	1.4223	1.4177	1.4135
0.2932	1.4243	1.41945	1.4147	1.4106	0.2999	1.4232	1.4193	1.4147	1.4103
0.3999	1.4202	1.4154	1.4103	1.4067	0.4000	1.4195	1.4159	1.4116	1.4070
0.5001	1.4156	1.4106	1.4064	1.4024	0.4999	1.4149	1.4110	1.4071	1.4027
0.5998	1.4099	1.4053	1.4007	1.3967	0.5999	1.4096	1.4055	1.4013	1.3972
0.6995	1.4036	1.39845	1.3937	1.3896	0.7000	1.4038	1.3987	1.3944	1.3897
0.8002	1.3942	1.3898	1.3850	1.3806	0.8000	1.395	1.3904	1.3858	1.3812
0.8997	1.3834	1.3790	1.3743	1.3696	0.8988	1.3838	1.3793	1.3747	1.3700
1.0000	1.3692	1.3641	1.3594	1.3543	1.0000	1.3692	1.3641	1.3594	1.3543

[a]Standard uncertainties u are $u(x_1)$ = 0.0002, $u(T)$ = 0.02 K, $u(p)$ = 0.20 kPa and $u(n_D)$ = 0.0005.

Table 5 Densities and dynamic viscosities of pure compounds in this work compared with literature data at corresponding temperatures and 0.1 MPa[a]

Component	T/(K)	ρ/(g·cm⁻³)			η/(mPa·s)			n_D		
		Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%
Dimethyl carbonate	293.15	1.0697	1.06965 [21], 1.0698 [22]	0.007	0.637	0.629 [23], 0.624 [24]	1.677	1.3692	1.3689 [25], 1.36835 [26]	0.042
	298.15	1.0632	1.06311 [21], 1.0632 [22]	0.004	0.592	0.590 [23], 0.585 [24]	0.768			
	303.15	1.0566	1.05635 [21], 1.0565 [22]	0.016	0.556	0.555 [23], 0.550 [24]	0.635	1.3641	1.36414 [26]	0.003
	308.15	1.0499	1.0498 [22], 1.0496 [27]	0.019	0.521	0.519 [24]	0.385			
	313.15	1.0432	1.04310 [21], 1.0430 [22]	0.014	0.496	0.495 [23], 0.491 [24]	0.610	1.3594	1.35932 [26], 1.3604 [25]	0.0039
	318.15	1.0365	1.036590 [24]	0.009	0.474	0.475 [23]	0.210			
	323.15	1.0298	1.02959 [28], 1.029843 [24]	0.012	0.446	0.446 [23], 0.442 [24]	0.452	1.3543	1.3546 [29]	0.022

Table 5 (continued)

Component	T/(K)	ρ/(g·cm^{-3})			η/(mPa·s)			n_D		
		Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%
Methyl decanoate	293.15	0.8722	0.8723 [30], 0.8721 [31]	0.011	2.125	2.1360 [30]	0.515	1.4258	1.4258 [33]	0
	298.15	0.8681	0.8682 [30],	0.011	1.906	1.9335 [30], 1.915 [32]	0.946			
	303.15	0.8640	0.8641 [30], 0.8636 [31]	0.028	1.735	1.7601 [30], 1.739 [32]	0.828	1.4214	1.4216 [33]	0.014
	308.15	0.8599	0.8600 [30]	0.012	1.588	1.6091 [30], 1.589 [32]	0.687			
	313.15	0.8558	0.8560 [30], 0.8559 [31]	0.017	1.454	1.4773 [30], 1.458 [32]	0.925	1.4165	1.4173 [33]	0.056
	318.15	0.8517	0.8519 [30]	0.023	1.342	1.3613 [30], 1.341 [32]	0.746			
	323.15	0.8476	0.8478 [30], 0.8477 [31]	0.018	1.240	1.2589 [30], 1.240 [32]	0.751	1.4122		

Table 5 (continued)

Component	T/(K)	$\rho/(\text{g·cm}^{-3})$			$\eta/(\text{mPa·s})$			n_D		
		Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%
Ethyl decanoate	293.15	0.8641	0.8643 [30], 0.86400 [34]	0.017	2.330	2.3263 [30], 2.34 [35]	0.293	1.4255	1.4254 [36], 1.425771 [37]	0.013
	298.15	0.8601	0.8602 [30], 0.859995 [34]	0.012	2.122	2.1029 [30]	0.908			
	303.15	0.8560	0.8562 [30], 0.85589 [34]	0.018	1.923	1.9111 [30]	0.623	1.4214	1.4215 [36], 1.421546 [37]	0.009
	308.15	0.8519	0.8521 [30], 0.85182 [34]	0.016	1.744	1.7453 [30]	0.074			
	313.15	0.8479	0.8480 [30],	0.012	1.598	1.6000 [30], 1.59 [35]	0.314	1.4164	1.4173 [36], 1.417290 [37]	0.063
	318.15	0.8438	0.8439 [30], 0.84367 [34]	0.014	1.475	1.4729 [30]	0.143			
	323.15	0.8397	0.8398 [30]	0.012	1.358	1.3599 [30]	0.140	1.4121	1.412965 [37]	0.061

Table 5 (continued)

Component	$T/(\text{K})$	$\rho/(\text{g·cm}^{-3})$			$\eta/(\text{mPa·s})$			n_D		
		Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%
Methyl laurate	293.15	0.8693	0.8693 [38], 0.86916 [39]	0.008	3.153	3.1519 [40]	0.035	1.4323	1.43208 [39], 1.43209 [21]	0.015
	298.15	0.8654	0.86527 [32], 0.86523 [39]	0.017	2.787	2.7895 [40], 2.780 [32]	0.171			
	303.15	0.8615	0.86133 [32], 0.8614 [38]	0.016	2.504	2.4907 [40], 2.495 [32]	0.447	1.4280	1.42786 [39], 1.4277 [41]	0.015
	308.15	0.8576	0.85740 [32], 0.85737 [39]	0.025	2.257	2.2437 [40], 2.250 [32]	0.452			
	313.15	0.8536	0.85347 [32], 0.8535 [38]	0.013	2.046	2.0346 [40], 2.041 [32]	0.403	1.4233	1.42401 [41], 1.4235 [33],	0.032
	318.15	0.8497	0.84954 [32], 0.84951 [39]	0.021	1.870	1.8624 [40], 1.862 [32]	0.419			
	323.15	0.8458	0.84560 [32], 0.8458 [38]	0.012	1.707	1.7023 [40], 1.705 [32]	0.197	1.4191	1.41943 [39]	0.023

Table 5 (continued)

Component	T/(K)	ρ/(g·cm⁻³)			η/(mPa·s)			n_D		
		Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%	Expt.	Lit.	AARD/%
Ethyl laurate	293.15	0.8621	0.8622 [30], 0.86234 [42]	0.020	3.370	3.3797 [30], 3.3803 [33]	0.296	1.4314	1.4315 [36], 1.431689 [37]	0.014
	298.15	0.8582	0.85825 [30]	0.005	3.011	3.0152 [30], 3.015 [31]	0.136			
	303.15	0.8543	0.85436 [30], 0.85428 [43]	0.005	2.698	2.7073 [30], 2.704 [31]	0.283	1.4274	1.4275 [36], 1.427545 [37]	0.009
	308.15	0.8504	0.8505 [30]	0.012	2.443	2.4455 [30], 2.439 [31]	0.133			
	313.15	0.8465	0.8466 [30], 0.84677 [42]	0.022	2.211	2.2198 [30], 2.213 [31]	0.243	1.4229	1.4235 [36], 1.423352 [37]	0.037
	318.15	0.8426	0.8427 [30]	0.012	2.023	2.0240 [30], 2.016 [31]	0.198			
	323.15	0.8387	0.8388 [30], 0.83890 [43]	0.018	1.836	1.8531 [30], 1.846 [31]	0.732	1.4186	1.419149 [37]	0.039

[a]Standard uncertainties u are u (p) = 1.0 kPa, u (T) = 0.03 K for density experiments, u (T) = 0.01 K for viscosity experiments, u_r $(ρ)$ = 0.001, u_r $(η)$ = 0.010.
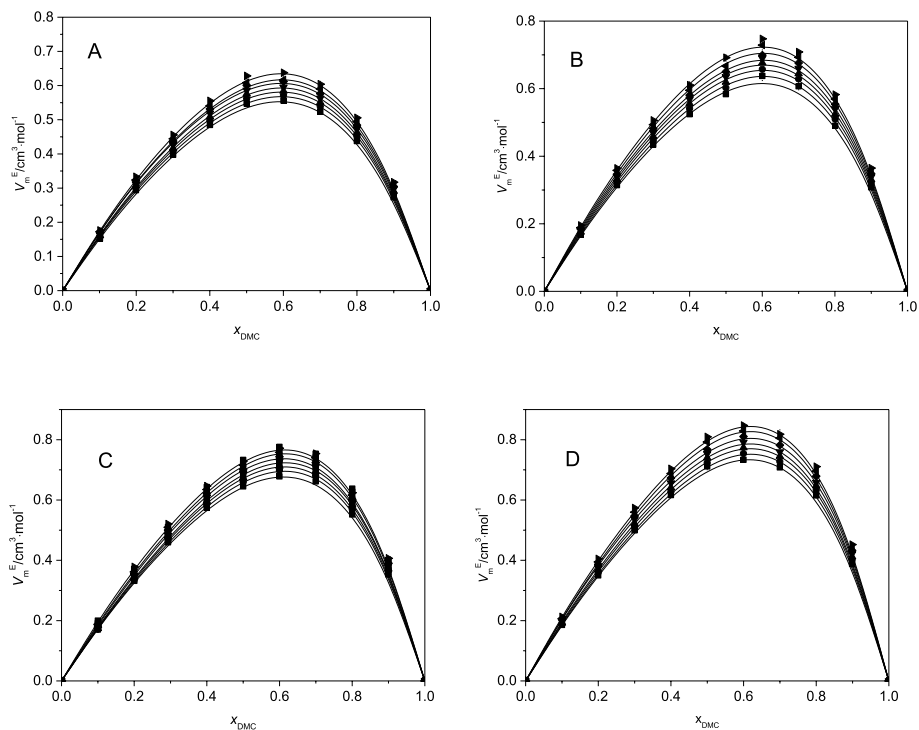
Fig. 1 Excess molar volumes for binary systems as a function of mole fraction of DMC (**a**, DMC + methyl decanoate; **b**, DMC + ethyl decanoate; **c**, DMC + methyl laurate; **d**, DMC + ethyl laurate) at $T =$ (293.15 − 323.15) K and pressure $p = 0.1$ MPa: Filled Square, 293.15 K; Filled Circle, 298.15 K; Filled Triangle, 303.15 K; Filled Down-Pointing Triangle , 308.15 K; Filled Lozenge, 313.15 K; Filled Left-Pointing Triangle, 318.15 K; Filled Right-Pointing Triangle, 323.15 K; Solid Line, Redlich-Kister equation correlations

Fig. 2 Comparison of excess molar volumes of DMC + fatty acid ethyl esters at $T = 293.15$ K: Filled Square, DMC + methyl decanoate; Filled Circle, DMC + ethyl decanoate; Filled Triangle, DMC + methyl laurat; Filled Down-Pointing Triangle, DMC + ethyl laurate; Solid Line, Redlich-Kister equation correlations
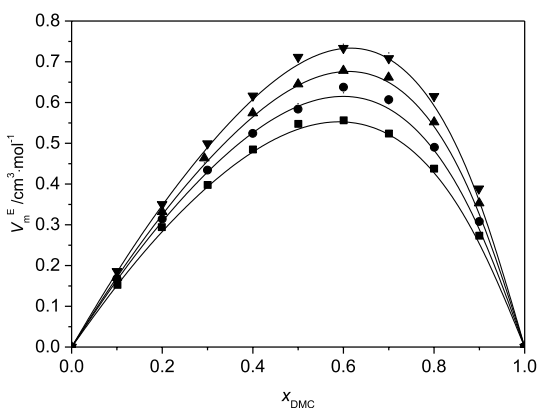
higher than those of DMC + methyl or ethyl decanoate. There is not enough difference of viscosity deviation between the mixture of DMC + methyl and ethyl esters of fatty acids with the same carbon atom number.
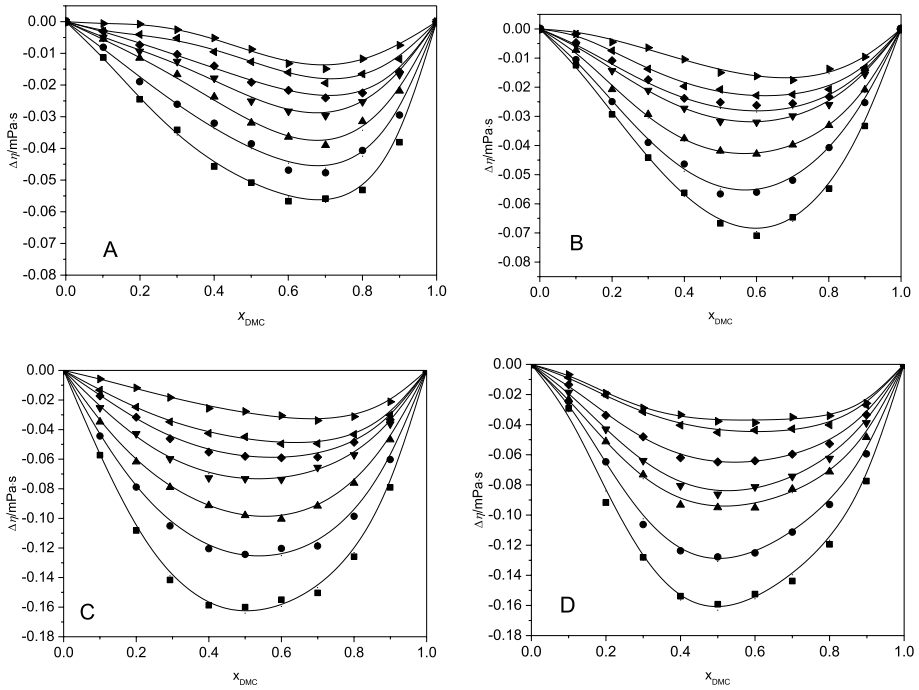
Fig. 3 Curves of viscosity deviations ($\Delta\eta$): **A**, DMC + methyl decanoate; **B**, DMC + ethyl decanoate; **C**, DMC + methyl laurate; **D**, DMC + ethyl laurate at $T = (293.15 - 323.15)$ K and pressure $p = 0.1$ MPa: Filled Square, 293.15 K; Filled Circle, 298.15 K; Filled Triangle, 303.15 K; Filled Down-Pointing Triangle, 308.15 K; Filled Lozenge, 313.15 K; Filled Left-Pointing Triangle, 318.15 K; Filled Right-Pointing Triangle, 323.15 K; Redlich-Kister equation correlations

Fig. 4 Viscosity deviations as a function of mole fraction of DMC for four binary systems at 293.15 K: Filled Square, DMC + methyl decanoate; Filled Circle, DMC + ethyl decanoate; Filled Triangle, DMC + methyl laurate; Filled Down-Pointing Triangle, DMC + ethyl laurate; Solid Lines, Redlich-Kister equation correlations
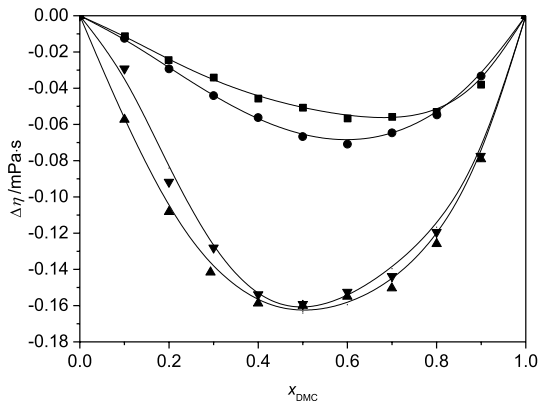
Excess values and deviations (V_m^E, $\Delta\eta$) of binary mixtures are all fitted to the Redlich-Kister polynomial equation.

Fig. 5 Excess Gibbs energies of activation vs. composition of DMC for the binary systems at 293.15 K: Filled Square, DMC + methyl decanoate; Filled Circle, DMC + ethyl decanoate; Filled Triangle, DMC + methyl laurate; Filled Down-Pointing Triangle, DMC + ethyl laurate; Solid Line, Redlich-Kister equation correlat ions
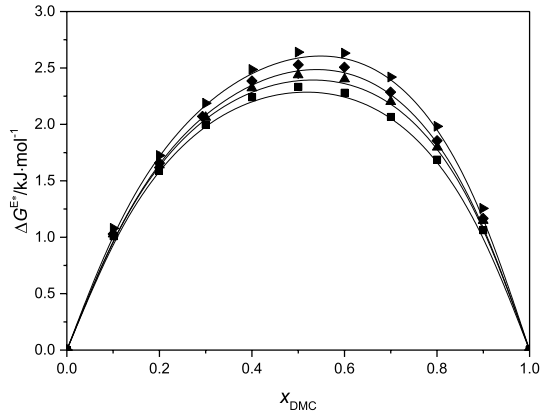
$$Y = x_1\left(1 - x_1\right) \sum_{i=1}^{k} A_i(2x_1 - 1)^{i-1} \tag{4}$$

where x_i and A_i are molar fraction and the polynomial coefficients of the Redlich-Kister equation; k is the number of coefficients. A_i are obtained by using the linear least-squares fitting method. The values correlated from Eq. 5 are used to identify the standard deviation of the experimental and theoretical values. The standard deviation (σ) is calculated by the following equation:

$$\sigma = \sum \left(Y_{\text{exp}} - Y_{\text{cal}}\right)^2 /(n - k).^{1/2} \tag{5}$$

where Y_{exp} and Y_{cal} are the experimental and calculated value of excess properties or deviations. n is the number of datum points. The values of the fitting parameters (A_i) along with the standard deviations (σ) for V_m^E and $\Delta\eta$ are all summarized in Tables S1 and S2 in the Supporting Information.

On the basis of the theory of absolute reaction rates, the excess Gibbs energy (ΔG^{E*}) of activation of viscous flow can be calculated by using Eq. 6:

$$\Delta G^{E*} = RT\left[\ln\left(\eta V/\eta_2 V_2\right) - x_1 \ln(\eta_1 V_1/\eta_2 V_2)\right] \tag{6}$$

In Eq. 6, V, V_1 and V_2 are the molar volumes of the binary mixture, DMC and a fatty acid ethyl ester, respectively. The sign of the values of ΔG^{E*} can be considered as a reliable criterion for detecting or excluding the presence of interaction between unlike molecules. The variations of ΔG^{E*} with x_{DMC} for four binary mixtures at 293.15 K are presented in Fig. 5. The ΔG^{E*} has been found to be positive for four tested systems under investigation over the entire range of mole fractions of DMC. The order of positive (ΔG^{E*}) values is as follows: (DMC + ethyl laurate) > (DMC + methyl laurate) > (DMC + ethyl decanoate) > (DMC + methyl decanoate). ΔG^{E*} indicates the degree of intermolecular interaction and the transport behavior of the mixture [2]. Volumetric and viscometric effects both affect the values of ΔG^{E*}. Positive deviations of ΔG^{E*} are the result of specific interactions between unlike molecules [46, 24].
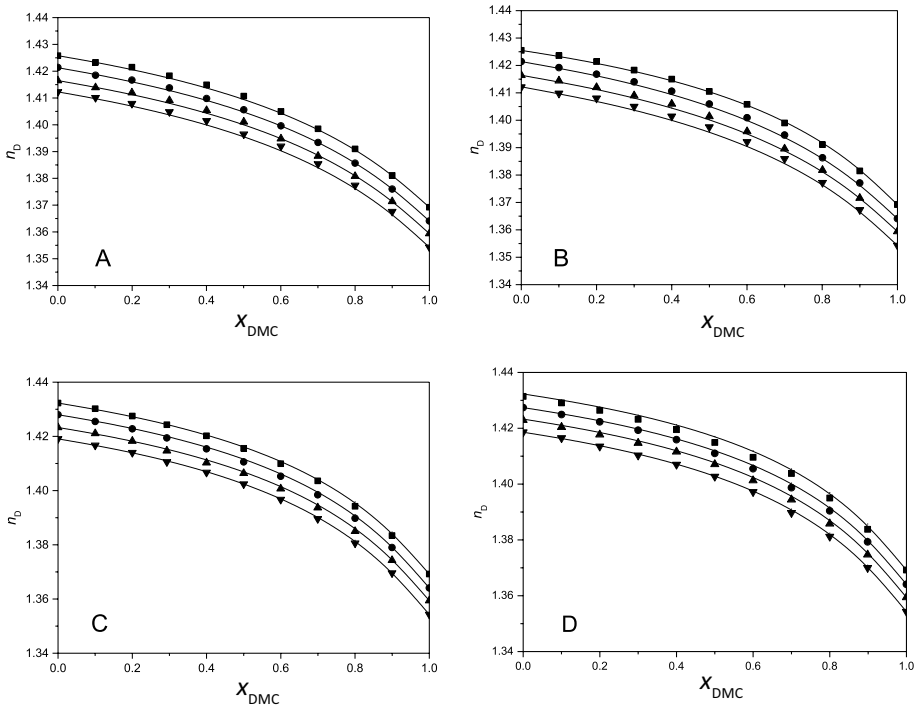
Fig. 6 Variation of n_D for the binary systems: A, DMC + methyl decanoate, B, DMC + ethyl decanoate, C, DMC + methyl laurate, D, DMC + ethyl laurate: Filled Square, 293.15 K; Filled Circle, 303.15 K; Filled Triangle, 313.15 K; Filled Down-Pointing Triangle, 323.15 K; − the correlated results of Lorentz-Lorenz equation

The experimental n_D values are analyzed using the theoretical methods. The Dale-Gladstone equation (Eq. 7) Arago-Biot equation (Eq. 8) and Lorentz-Lorenz equation (Eq. 9) were used to predicting the refractive index for binary systems.

$$n_D - 1 = \phi_1\left(n_{D1} - 1\right) + \phi_2(n_{D2} - 1) \tag{7}$$

$$n_D^2 = \phi_1 n_{D1}^2 + \phi_2 n_{D2}^2 \tag{8}$$

$$\frac{n_D^2 - 1}{n_D^2 + 2} = \phi_1 \frac{n_{D1}^2 - 1}{n_{D1}^2 + 2} + \phi_2 \frac{n_{D2}^2 - 1}{n_{D2}^2 + 2} \tag{9}$$

where n_D the refractive index of the mixture, φ_i the volume fractions of component $_i$, and n_{Di} is the refractive index of pure component i, respectively. The solid line in Fig. 6 is the results correlated from Lorentz-Lorenz equation. The standard deviations of values of three theoretical methods for binary systems are listed at Table 6.

Table 6 Standard deviation of refractive index calculated by Arago-Biot equation (σ_{A-B}), Dale–Gladstone relations (σ_{D-G}), Lorentz-Lorenz relations (σ_{L-L})

Binary systems	T/K	σ_{D-G}	σ_{A-B}	σ_{L-L}
DMC + methyl decanoate	293.15	0.0005	0.0005	0.0006
	303.15	0.0006	0.0006	0.0006
	313.15	0.0007	0.0007	0.0006
	323.15	0.0011	0.0009	0.0010
DMC + ethyl decanoate	293.15	0.0006	0.0007	0.0005
	303.15	0.0007	0.0005	0.0009
	313.15	0.0008	0.0007	0.0005
	323.15	0.0011	0.0005	0.0007
DMC + methyl laurate	293.15	0.0009	0.0010	0.0006
	303.15	0.0009	0.0011	0.0006
	313.15	0.0009	0.0011	0.0007
	323.15	0.0006	0.0008	0.0003
DMC + ethyl laurate	293.15	0.0018	0.0020	0.0015
	303.15	0.0011	0.0013	0.0008
	313.15	0.0016	0.0018	0.0013
	323.15	0.0008	0.0010	0.0005

4 Conclusions

New experimental data for densities, viscosities and refractive indices of binary systems of (DMC + methyl decanoate), (DMC + ethyl decanoate), (DMC + methyl laurate) and (DMC + ethyl laurate) have been determined over the temperatures ranging from 293.15 to 323.15 K at atmospheric pressure. The densities, viscosities and refractive indices of the binary mixture all decrease with the increase of temperature. The excess molar volume, viscosity deviations of the studied systems have been calculated based on the experimental data of density and dynamic viscosity and compared. The calculated excess properties were also fitted to the Redlich–Kister equation. The excess mole volumes are positive within the measured range, and the viscosity deviations are negative. The excess Gibbs energy of activation was also calculated that presented positive values. Investigating the thermophysical and excess properties of fatty acid methyl and ethyl esters and their mixtures will assist in researching the surrogate mixtures of biodiesels using as a model system to investigate the combustion characteristic of fuels.

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Author Contributions XG and DL wrote and revised the main manuscript text. DL prepared Figs. 1, 2, 3, 4, 5 and 6. MZ and BH investigated and collected the data. All authors reviewed the manuscript.

Data Availability All data generated or analysed during this study are included in this published article.

Declarations

Competing Interests The authors declare no competing interests.

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