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脂肪酸酯的自由体积理论黏度模型

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摘要：在对文献中发表的 22 种脂肪酸甲酯和 16 种脂肪酸乙酯的液相黏度实验数据进行收集的基础上，建立了脂肪酸酯类物质的自由体积理论黏度模型，并得到了模型的参数。结果表明，对脂肪酸甲酯而言，实验值和计算值的最大偏差小于 5.5%（其中脂肪酸链的碳原子数与不饱和键数之比为 20∶1和 22∶1的除外），对脂肪酸乙酯，最大偏差小于 4.0%（其中脂肪酸链的碳原子数与不饱和键数之比为 18∶1和 18∶2的除外）。分析发现，模型参数与酯类相对分子质量之间存在一定的规律，获得了其定量关系，使得所建立的模型具有了预测同类物质黏度的功能。将预测模型计算结果和实验值进行比较发现，对 38 种脂肪酸酯类，黏度总体平均相对偏差小于 1%，可以满足实际工程的需要。

关键词：黏度模型；脂肪酸甲酯；脂肪酸乙酯；自由体积理论
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Free-Volume Theory Viscosity Model of Fatty Acid Esters

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Abstract: The experimental data of viscosity of 22 fatty acid methyl esters and 16 fatty acid ethyl esters are collected from literatures, the free-volume theory based viscosity model of these esters is established and the coefficients in the model are obtained. Investigation shows that the coefficients in the model exhibit regular trends along with the molecular weight of the esters. The relationship between the coefficients and the molecular weight is obtained, and the predictive viscosity values using the relationship are compared with the literature data. Results show that the maximum relative deviation between the calculated viscosity values and those from literatures is lower than 5.5% for fatty acid methyl esters (except for C20∶1 and C22∶1) and 4.0% for fatty acid ethyl esters (except for C18∶1 and C18∶2). For these 38 esters, the overall average relative deviation is less than 1%, indicating that this predictive model could meet the requirements of practical engineering.

Keywords: viscosity model; fatty acid methyl ester; fatty acid ethyl ester; free-volume theory

由于能源危机和环境污染的日趋严重，具有可再生性、绿色环保的生物柴油作为一种新型的替代燃料成为了国内外研究的热点[1-2]。生物柴油的主要成分是脂肪酸酯类，包括脂肪酸甲酯和脂肪酸乙酯。脂肪酸酯类物质的黏度对于生物柴油在工程中的应用和相关科学研究具有十分重要的作用，国内

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外许多学者针对其黏度已经展开了相关的研究[3]。

Allal 等基于自由体积概念，提出了一个新的黏度模型[4-5]。该模型已经广泛应用于烷烃、离子液体、制冷剂、醇、二氧化碳以及水等多种物质的黏度计算，然而在脂肪酸酯类物质黏度计算中的应用还比较少。目前，仅有 Oliveira 等利用变形的自由体积理论计算了 14 种甲酯和 10 种乙酯的黏度，其数据来源比较单一，且预测精度较差[6]。鉴于此，本文在对文献黏度和密度实验数据进行收集整理的基础上，建立了 38 种脂肪酸酯类物质的自由体积理论黏度模型，并对模型系数和酯类相对分子质量的关系进行详细研究，得到了其定量的关系式。

1 自由体积黏度理论

流体的黏度主要由两部分组成，即稀薄状态黏度项和过余黏度项。因此，自由体积黏度理论模型的表达式为

$$\eta = \eta_0 + \frac{\rho l(\alpha\rho + PM/\rho)}{\sqrt{3RTM}}\exp\left[B\left(\frac{\alpha\rho + PM/\rho}{RT}\right)^{3/2}\right] \tag{1}$$

式中：η_0 为稀薄状态黏度项；ρ 是流体的密度；M 是物质的相对分子质量；R 是理想气体常数；P 是压力；未知量 l、α、B 由实验数据回归得到。

η_0 的计算采用 Riesco 等提出的关联式[7]

$$\eta_0 = 0.083\ 867\ \sqrt{MT}\ \frac{f_\eta}{\Omega_\eta} \tag{2}$$

其中 Ω_η 是碰撞积分

$$\Omega_\eta = \pi\sigma^2\Omega_\eta^* \tag{3}$$

$$\begin{aligned}\Omega_\eta^* = {} & 1.161\ 45T^{*-0.148\ 74} + \\ & 0.524\ 87\exp(-0.773\ 2T^*) + \\ & 2.161\ 78\exp(-2.437\ 87T^*) - \\ & 6.435\times10^{-4}T^{*0.148\ 74}\sin(18.032\ 3T^{*-0.768\ 30} - \\ & 7.273\ 71)\end{aligned} \tag{4}$$

f_η 是高阶校正因子，可表示为

$$f_\eta = 1 + \frac{3}{49}(4E^* - 3.5)^2 \tag{5}$$

$$\begin{aligned}E^* = {} & [1.115\ 21T^{*-0.147\ 96} + \\ & 0.448\ 44\exp(-0.995\ 48T^*) + \\ & 2.300\ 09\exp(-3.060\ 31T^*) + \\ & 4.565\times10^{-4}T^{*0.147\ 96}\sin(38.586\ 8T^{*-0.694\ 03} - \\ & 2.563\ 75)/\Omega_\eta^*]\end{aligned} \tag{6}$$

式(3)(4)中 σ 和 T^* 可表示为

$$\sigma = 0.809V_c^{1/3} \tag{7}$$

$$T^* = T/\varepsilon \tag{8}$$

$$\varepsilon = T_c/1.259\ 3 \tag{9}$$

式中：T^* 是无量纲温度；ε 是能量参数；σ 是长度比例参数；V_c 是临界体积；T_c 是临界温度。V_c、T_c 按文献[8]取值。

2 计算结果与讨论

对文献中发表的 38 种脂肪酸酯类(包含 22 种脂肪酸甲酯和 16 种脂肪酸乙酯)的液相黏度和密度实验数据进行了全面的收集，密度共计 550 个数据点[9-31]，黏度共计 484 个数据点[9,17-18,20-21,24,27,32-40]，所有实验数据均为常压下数据。

由于建立黏度模型时需要用到密度值，本文将流体密度 ρ 作为温度 T 与参考密度 ρ_{ref} 的函数

$$\rho = \rho_{ref}\sum_{i=0}^{3}a_i(T/298.15)^i \tag{10}$$

其中系数 $a_0\sim a_3$ 通过对酯类密度实验数据进行回归得到，ρ_{ref} 为每种酯类在 298.15 K 时的密度，对于熔点较高的脂肪酸酯，参考密度为熔点以上最低测试温度时的密度。

根据收集到的黏度实验数据以及密度方程，建立了脂肪酸甲酯和乙酯的自由体积理论黏度模型，所得到的模型系数如表 1 所示。

图 1 和图 2 给出了所建立的黏度模型计算值与实验值的偏差分布。从图 1 中可以看出，对于脂肪酸甲酯，除 C20∶1和 C22∶1以外，实验值与计算值偏差均处在 5.5% 以内，而对于脂肪酸乙酯，除了 C18∶1和 C18∶2以外，偏差均处在 4.0% 以内，说明本文建立的黏度模型具有较高的计算精度。

通过对表 1 中的系数进行分析发现，在一定范围内模型参数 l、α、B 与酯类相对分子质量之间存在一定的规律性。研究表明，对于 C1～C6 的脂肪酸酯类而言，有如下关系

$$Y = p_0 + p_1M + p_2/M + p_3M^2 + p_4/M^2 + p_5M^3 \tag{11}$$

对于 C7～C24 的脂肪酸酯类(C18∶3除外)，有

$$Y = p_0 + p_1M + p_2M^{2.5} + p_3M/\ln M + p_4/M^2 \tag{12}$$

式中：Y 分别代表 l、α 和 B；M 为酯类相对分子质量；$p_0\sim p_5$ 通过对表 1 中的系数回归得到，其结果列于表 2。

表 3 给出了根据式(1)、式(11)(12)推算得到的两个黏度值各自与实验值的相对偏差(分别对应表中的初始值、预测值)。同时，为了比较，表 3 还给出了文献[6]的计算结果。从表 3 中可以看出，所建立

表 1　自由体积理论模型参数

$Cx:y$	M	$l/10^{-8}$	$\alpha/10^{-1}$	$B/10^{2}$	$Cx:y$	M	$l/10^{-8}$	$\alpha/10^{-1}$	$B/10^{2}$
^{M}C1:0	0.060	5.752	0.750	0.803	^{M}C18:1	0.297	3.162	2.074	1.498
^{M}C2:0	0.074	6.889	0.624	1.698	^{M}C20:1	0.325	2.662	2.457	1.293
^{M}C3:0	0.088	7.028	0.522	2.969	^{M}C22:1	0.353	1.577	4.828	0.543
^{M}C4:0	0.102	7.671	0.451	4.740	^{E}C1:0	0.074	2.944	1.396	0.692
^{M}C5:0	0.116	8.748	0.344	7.484	^{E}C2:0	0.088	4.720	0.924	1.385
^{M}C6:0	0.130	9.733	0.203	14.650	^{E}C3:0	0.102	6.355	0.665	2.549
^{M}C7:0	0.144	8.864	0.159	19.620	^{E}C4:0	0.116	7.501	0.553	3.826
^{M}C8:0	0.158	9.213	0.319	9.914	^{E}C5:0	0.130	5.802	0.450	6.713
^{M}C10:0	0.186	6.996	0.425	8.368	^{E}C6:0	0.144	8.402	0.377	7.646
^{M}C12:0	0.214	5.789	0.530	7.207	^{E}C8:0	0.172	8.136	0.264	12.480
^{M}C14:0	0.242	5.967	0.571	6.790	^{E}C10:0	0.200	7.257	0.304	11.610
^{M}C16:0	0.270	5.269	1.187	2.925	^{E}C12:0	0.228	6.207	0.399	9.509
^{M}C18:0	0.299	4.147	1.915	1.687	^{E}C14:0	0.256	5.345	0.574	6.895
^{M}C20:0	0.327	3.787	2.799	1.021	^{E}C16:0	0.284	5.864	0.757	5.029
^{M}C22:0	0.355	3.808	3.180	0.881	^{E}C18:3	0.306	9.099	0.566	5.260
^{M}C24:0	0.383	3.795	3.419	0.828	^{E}C18:2	0.309	5.874	0.892	3.763
^{M}C16:1	0.268	5.277	0.783	4.531	^{E}C18:1	0.311	4.391	1.215	2.948
^{M}C18:3	0.292	6.998	0.993	2.939	^{E}C18:0	0.313	3.954	2.392	1.237
^{M}C18:2	0.294	4.386	1.710	1.669	^{E}C20:0	0.341	3.899	2.819	1.019

注：$Cx:y$ 中 x 代表脂肪酸链的碳原子数，y 代表脂肪酸链中不饱和键数；上标 M 代表甲酯，E 代表乙酯。

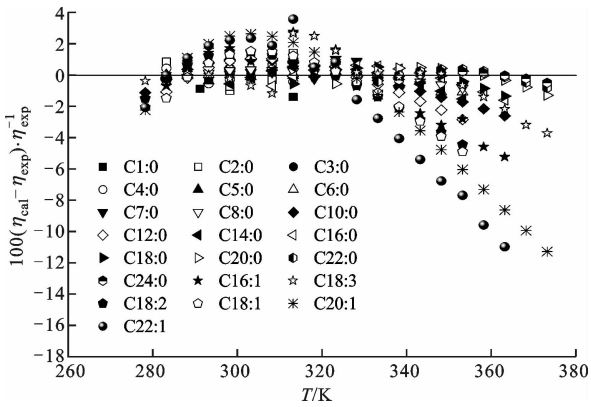

图 1　脂肪酸甲酯黏度偏差分布

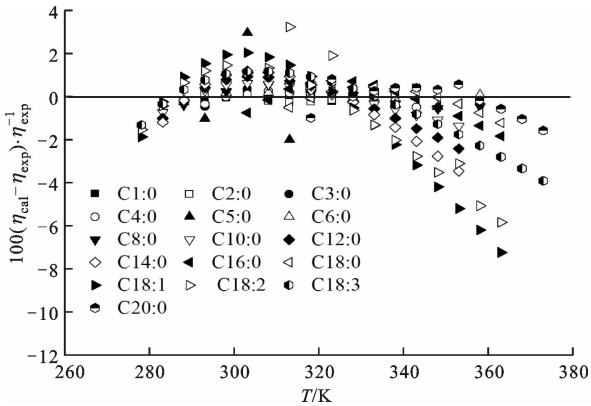

图 2　脂肪酸乙酯黏度偏差分布

的预测模型计算精度与直接回归的精度相当，且明显高于文献[6]的模型计算精度。对于脂肪酸甲酯和脂肪酸乙酯预测黏度的总体平均偏差分别为 1% 和 0.84%，具有较高的预测精度，可以用来推算其他同类物质的黏度。需要指出的是，对于顺-11-二十烯酸甲酯(C20:1)和芥酸甲酯(C22:1)两种脂肪酸甲酯而言，其黏度偏差较大(最大偏差超过 10%)，对比文献中 VTF 方程关联结果和实验数据[21]，发现偏差较大的实验点均出现在 70°以上的温度点，因此对这两种物质在高温时的黏度实验测试还需进一步研究。

3　结　论

在对 38 种脂肪酸酯类物质黏度文献数据整理的基础上，结合 Riesco 稀薄气体黏度推算法与Allal 自由体积理论模型对酯类黏度进行了研究，建立了其黏度模型，同时获得了基于酯类相对分子质量推算模型参数的运算法则。研究结果表明，在研究的温度范围内，预测值与实验值的总体平均相对偏差在 1%以内，说明本文建立的模型对常压下脂肪酸甲酯和乙酯的黏度推算是适用的。

表 2 参数关联结果

Cx:y	参数	p_0	p_1	p_2	p_3	p_4	p_5
^{M}C1:0～^{M}C6:0	l	-1.59×10^{-6}	2.22×10^{-6}	1.41×10^{-7}	6.85×10^{-5}	-3.68×10^{-9}	-2.86×10^{-4}
	a	-1.44×10^{1}	1.56×10^{2}	6.56×10^{-1}	-8.18×10^{2}	-1.15×10^{-2}	1.66×10^{3}
	B	-5.02×10^{5}	5.92×10^{6}	2.11×10^{4}	-3.44×10^{7}	-3.49×10^{2}	7.98×10^{7}
^{M}C7:0～^{M}C14:0	l	-1.91×10^{-6}	2.15×10^{-4}	1.42×10^{-3}	5.31×10^{-4}	-1.25×10^{-8}	
	a	-2.74×10^{1}	1.95×10^{2}	1.22×10^{3}	4.65×10^{2}	-6.43×10^{-3}	
	B	1.44×10^{5}	-1.20×10^{7}	-7.72×10^{7}	-2.92×10^{7}	5.88×10^{2}	
^{M}C16:0～^{M}C24:0	l	3.75×10^{-8}	1.77×10^{-6}	2.19×10^{-5}	6.83×10^{-6}	8.38×10^{-9}	
	a	-5.98×10^{1}	1.01×10^{2}	-1.36×10^{3}	-3.48×10^{2}	9.02×10^{-1}	
	B	2.22×10^{4}	-2.51×10^{4}	6.61×10^{5}	1.77×10^{5}	-2.63×10^{2}	
^{M}C16:1、^{M}C18:1、	l	3.75×10^{-8}	1.77×10^{-6}	2.19×10^{-5}	6.83×10^{-6}	8.38×10^{-9}	
^{M}C18:2、^{M}C20:1、	a	-2.37×10^{3}	1.23×10^{3}	-7.85×10^{4}	-2.20×10^{4}	3.39×10^{1}	
^{M}C22:1	B	6.08×10^{5}	-1.32×10^{5}	2.19×10^{7}	6.24×10^{6}	-8.38×10^{3}	
^{E}C1:0～^{E}C6:0	l	-3.39×10^{-4}	3.32×10^{-3}	1.71×10^{-5}	-1.61×10^{-2}	-3.39×10^{-7}	3.06×10^{-2}
	a	-6.03×10^{1}	5.70×10^{2}	3.13×10^{0}	-2.65×10^{3}	-6.29×10^{-2}	4.84×10^{3}
	B	1.90×10^{6}	-1.85×10^{7}	-9.66×10^{4}	8.89×10^{7}	1.94×10^{3}	-1.68×10^{8}
^{M}C8:0～^{E}C16:0	l	4.93×10^{-7}	-3.70×10^{-5}	-2.40×10^{-4}	-9.02×10^{-5}	2.33×10^{-9}	
	a	2.95×10^{-1}	3.10×10^{1}	2.38×10^{2}	8.50×10^{1}	-6.27×10^{-3}	
	B	7.52×10^{3}	-3.39×10^{5}	-2.15×10^{6}	-8.06×10^{5}	-1.06×10^{1}	
^{E}C18:0、^{E}C18:1、	l	-1.97×10^{-2}	2.37×10^{-2}	-5.21×10^{-1}	-1.40×10^{-1}	3.06×10^{-4}	
^{E}C18:2、^{E}C18:3、	a	4.50×10^{5}	-4.23×10^{5}	1.30×10^{7}	3.57×10^{6}	-6.81×10^{3}	
^{E}C20:0	B	-7.67×10^{8}	7.42×10^{8}	-2.19×10^{10}	-6.01×10^{9}	1.16×10^{7}	

表 3 模型初始偏差和预测偏差

Cx:y	δ_{MRD}/%		δ_{ARD}/%			Cx:y	δ_{MRD}/%		δ_{ARD}/%		
	初始	预测	初始	预测	文献[6]		初始	预测	初始	预测	文献[6]
^{M}C1:0	1.39	2.24	0.83	1.09		^{M}C20:1	11.25	11.33	3.64	3.65	4.00
^{M}C2:0	1.42	1.31	0.61	0.63		^{M}C22:1	10.95	10.93	3.62	3.62	
^{M}C3:0	0.08	0.14	0.07	0.09		总体			0.96	1.00	4.07
^{M}C4:0	0.53	0.60	0.37	0.36		^{E}C1:0	0.21	0.25	0.12	0.11	
^{M}C5:0	0.40	0.51	0.19	0.26		^{E}C2:0	0.43	0.45	0.16	0.16	
^{M}C6:0	1.07	1.37	0.38	0.50		^{E}C3:0	0.49	0.49	0.20	0.20	
^{M}C7:0	1.04	1.34	0.46	0.49		^{E}C4:0	0.48	0.42	0.18	0.20	
^{M}C8:0	0.82	1.16	0.30	0.48	0.48	^{E}C5:0	2.97	3.09	1.99	1.96	
^{M}C10:0	2.60	2.88	0.95	0.97	2.10	^{E}C6:0	0.45	0.41	0.24	0.24	
^{M}C12:0	2.82	2.96	1.01	1.01	7.00	^{E}C8:0	0.56	0.80	0.35	0.36	2.20
^{M}C14:0	1.52	1.27	0.53	0.58	4.90	^{E}C10:0	1.37	1.71	0.54	0.63	2.30
^{M}C16:0	1.59	1.50	0.57	0.58	4.90	^{E}C12:0	2.41	2.73	0.88	0.91	5.00
^{M}C18:0	1.33	1.35	0.48	0.48	7.90	^{E}C14:0	3.46	3.39	1.19	1.19	8.30
^{M}C20:0	1.28	1.33	0.45	0.45	5.10	^{E}C16:0	1.83	1.95	0.67	0.68	5.00
^{M}C22:0	0.81	0.84	0.35	0.35	6.20	^{E}C18:3	3.98	3.74	1.31	1.31	3.70
^{M}C24:0	0.48	0.42	0.21	0.23	5.50	^{E}C18:2	5.83	5.69	2.09	2.08	4.60
^{M}C16:1	5.22	5.19	1.98	1.98		^{E}C18:1	7.23	7.35	2.39	2.40	5.20
^{M}C18:3	3.70	1.05			0.70	^{E}C18:0	1.21	1.24	0.42	0.42	4.80
^{M}C18:2	4.42	4.50	1.53	1.53	1.30	^{E}C20:0	0.60	0.59	0.60	0.59	6.30
^{M}C18:1	4.89	4.91	1.62		2.80	总体			0.59	0.84	4.74

注：δ_{MRD}为最大相对偏差；δ_{ARD}为平均相对偏差。

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