Journal of Molecular Liquids 340 (2021) 117231

Contents lists available at ScienceDirect

Journal of Molecular Liquids

journal homepage: www.elsevier.com/locate/molliq

Soybean oil-based monoacylglycerol synthesis using bio-compatible amino acid ionic liquid as a catalyst at low temperature

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ARTICLE INFO

Article history:
Received 11 July 2021
Revised 3 August 2021
Accepted 8 August 2021
Available online 11 August 2021

Keywords:
Monoacylglycerol
Soybean oil
Glycerolysis
Amino acid-based ionic liquid
[TMA][Arg]
Response surface methodology

ABSTRACT

Monoacylglycerol (MAG) from edible oil is one kind of green and safe food additive. This project mainly focused on a nontoxic and biodegradable catalyst, amino acid-based ionic liquids (AAILs), for the glycerolysis of soybean oil triacylglycerol (TAG) to prepare MAG at low temperature. Seven AAILs, from arginine, lysine, histidine, tryptophan, or glutamic acid and choline hydroxide, tetramethylhydroxide ammonium, or tetrabutylammonium hydroxide, were successfully synthesized in this work. The effects of substrate ratio, AAIL load, temperature, and reaction time on AAIL-catalyzed the glycerolysis of soybean oil were evaluated. Moreover, the activation energy of MAG formation was explored and reaction mechanism was also proposed. Finally, reaction conditions were optimized by response surface methodology. Results showed that, among all tested AAILs, [TMA][Arg] prepared by arginine (Arg) and tetramethylhydroxide ammonium (TMAOH) showed the outstanding catalytic performance for the glycerolysis of soybean oil. The maximum MAG yield (64.89 ± 1.26%) and TAG conversion (86.31 ± 1.68%) were achieved under the optimized conditions: 105 °C, 0.5 h, [TMA][Arg] load 10%, and substrate molar ratio 1:2 (soybean oil/glycerol). Moreover, the activation energies of MAG formation and TAG conversion were 44.22 kJ/mol and 37.58 kJ/mol, respectively.

1. Introduction

Monoacylglycerol (MAG), popular in food, pharmaceutical, and cosmetic industries, is a safe food additive [1,2]. In food industry, MAG is commonly used as an emulsifier [3], which thanks to its' excellent emulsifying properties. For instance, adding MAG into dairy products and beverages can prevent the sinking of protein and the floating of oil, and maintain the stability of the product. However, the content of MAG in nature is low (<5% in edible oil). To meet the abundant applications and large demands in industry, MAG is usually obtained by the esterification of fatty acids with glycerol [4,5], and hydrolysis [6,7] or alcoholysis [8–10] of fats and oils. With the high value-added utilization of glycerol and atom effective utilization, MAG synthesis by the glycerolysis of triacylglycerol (TAG) has drawn great attention [11,12].

Recently, inorganic alkali metal hydroxides or oxides (e.g., KOH or CaO), bio-enzyme [13,14], or ionic liquids (ILs) [15] have been used as catalysts for MAG synthesis by the glycerolysis of TAG. Unsatisfactorily, high temperature (e.g., 260 °C) was usually employed during the alkaline-catalyzed glycerolysis, which resulted in more energy requirement, undesired odor and taste formation, soap formation, and separation and purification difficulty [16]. In order to overcome these issues, enzymes have been successfully used in glycerolysis process [14]. However, high cost of enzyme and great mass transfer issues limited their applications in industry [17]. Compared with the traditional catalysts, the applications of ionic liquids as catalysts in chemical synthesis have been attracted more attention in recent years.

Ionic liquids (ILs) consisting entirely of ions are molten salts at room temperature [18]. Featured by favorable physicochemical properties, such as non-volatility, good thermal stability, low melting temperature, and high decomposition temperature, ILs have been successfully used in some reactions as solvents and promising catalysts [19–21]. For example, a MAG yield of ~69% can be obtained by the glycerolysis of soybean oil in the presence of 1-Butyl-3-methylimidazolium imidazolide ([Bmim]Im) at 200 °C [16]. However, ILs containing aromatic cations, such as imidazolium- and pyridinium-based ILs, have the disadvantages

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Abbreviations: AAILs, amino acid-based ionic liquids; MAG, monoacylglycerol; DAG, diacylglycerol; TAG, triacylglycerol; GC, gas chromatography; [Ch][OH], choline hydroxide; [TBA][OH], tetrabutylammonium hydroxide 30-hydrate; [TMA][OH], tetramethylammonium hydroxide pentahydrate.

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https://doi.org/10.1016/j.molliq.2021.117231
0167-7322/

of expensive cost, complex preparation, toxicity, and non-biodegradable [22–24]. Therefore, the development of the nontoxic, biodegradable and simple preparation IL has been very popular.

Compared with these catalysts, amino acid-based ionic liquids (AAILs), composed of renewable biomaterial amino acid, have the merits of simple preparation, nontoxic, and biodegradable [25,26], which have evoked considerable interest as an efficient catalyst [23,27] or green solvent [28]. AAILs have been used to prepare biodiesel and showed the best performance for the transesterification [27]. However, no information about AAIL as a catalyst for the glycerolysis of edible oil to prepare food additive MAG was available.

The main purpose of this work is to develop an eco-friendly catalyst-AAIL with high catalytic activity at low temperature to prepare soybean oil-based MAG by glycerolysis. First, seven AAILs were synthesized by a two-step strategy. The effect of AAIL structure on the glycerolysis of soybean oil TAG was evaluated. The effects of reaction parameters (temperature, substrate ratio, and AAIL load) on the glycerolysis were investigated and optimized by the response surface methodology (RSM). Furthermore, reaction mechanism of the glycerolysis was also proposed.

2. Material and methods

2.1. Materials

Refined soybean oil was purchased from a local supermarket. Physicochemical properties and the fatty acid composition of soybean oil were shown in Tables S1 and S2. Glycerol (G) was from Tianjin Kermel Chemical Reagent Co. Ltd. (Tianjin, China). Choline hydroxide ([Ch][OH]), tetrabutylammonium hydroxide 30-hydrate ([TBA][OH]), and tetramethylammonium hydroxide pentahydrate ([TMA][OH]) were obtained from Sigma-Aldrich (St. Louis, MO., USA). Arginine (Arg), lysine (Lys), histidine (His), tryptophan (Try), and glutamic acid (Glu) were from Shanghai Macklin Biochemical Co. Ltd. (Shanghai, China). For GC analysis, standard glyceryl monooleate, 1,3-diolein, and glyceryl trioleate were also purchased from Sigma-Aldrich (St. Louis, MO., USA).

2.2. Synthesis of AAIL

The AAILs were synthesized according to the previous report with some modifications [26]. In a typical preparation, 10 mL of an aqueous solution of 0.05 mol quaternary ammonium hydroxide ([Ch][OH], [TBA][OH] or [TMA][OH]) was added dropwise into 100 mL aqueous solution of amino acid with a 1:1.1 M ratio of quaternary ammonium hydroxide and amino acid, and stirred at room temperature and 250 rpm for 48 h. After neutralized, vacuum distillation at 80 °C was employed to remove water. The solvent mixture was dissolved in acetonitrile/methanol (7:3) to precipitate unreacted amino acid, and the precipitate was removed by vacuum filtration. Then, the solvent in AAIL was removed by the evaporation under vacuum at 60 °C. Finally, the AAIL were further dried in a vacuum oven at 90 °C for 0.5 h and stored in a desiccator. The water content of each AAIL was<0.08%.

2.3. Glycerolysis procedure

The glycerolysis of soybean oil were carried out in 25 mL round-bottom flasks with a magnetic stirrer bar in an oil bath at 300 rpm. The heterogeneous reaction mixtures consisted of soybean oil, glycerol, and AAIL. The effects of molar ratio of soybean oil and glycerol (2:1, 1:1, 1:2, and 1:4), reaction temperature (80 °C, 90 °C, 100 °C, 110 °C, and 120 °C), and AAIL load (3%, 5%, 8%, 10%, and 12%, w/w, based on oil mass) were investigated. Reaction mixture (0.10 mL) was withdrawn from glycerolysis system periodically for gas chromatography (GC) analysis.

2.4. Determination of acylglycerides by GC.

According to the previous reported method [14], sample analysis was performed by GC (Agilent 7890B, USA). Standards (monoolein, 1,3-diolein, and triolein) were used for the quantification of the products. Gas chromatography of TAG, DAG, and MAG was shown in Fig. S1.

The calculations for TAG conversion and MAG yield were as follows:

$$TAG\ conversion\ (\%) = \frac{n_{MAG} + 2 \times (n_{DAG} - n_{DAG_0})}{3 \times n_{TAG} + 2 \times n_{DAG+}n_{MAG}} \times 100 \quad (1)$$

$$MAG\ yield\ (\%) = \frac{n_{MAG}}{3 \times n_{TAG} + 2 \times n_{DAG+}n_{MAG}} \times 100 \quad (2)$$

where n_{TAG}, n_{DAG}, and n_{MAG} are the molar contents of TAG, diacylglycerol (DAG), and MAG, respectively; n_{DAG_0} is the initial DAG content of the glycerolysis system.

2.5. Response surface design

Design expert 10.0 software was used to perform the response surface design and analysis. According to the principle of Box-Behnken design and the results of single factor experiments, one RSM design with three factors and three levels was used. MAG yield (Y_1) was selected as the response. The interaction effect of reaction temperature (A), substrate molar ratio (B), and [TMA][Arg] loading (C) on the glycerolysis was explored, and the glycerolysis conditions were optimized.

3. Results and discussion

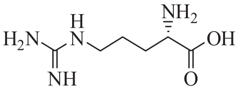
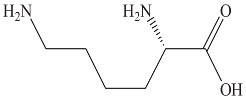
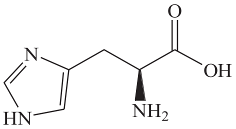
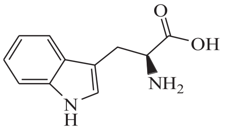
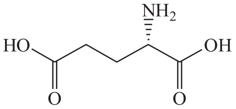
3.1. Effect of different AAILs on the glycerolysis of soybean oil

Seven AAILs ([CH][Arg], [CH][Lys], [CH][His], [CH][Trp], [CH][Glu], [TMA][Arg], and [TBA][Arg]) prepared by amino acid and quaternary ammonium hydroxide were used as catalysts for the glycerolysis of soybean oil, and their catalytic activity were shown in Table 1 and Fig. 1. As shown in Table 1, among all tested AAILs with cholinium as cation, [CH][Arg] showed the best catalytic activity for the glycerolysis of soybean oil. And TAG conversion (73.54 ± 1.20%) catalyzed by [CH][Arg] was 12 times of other catalysts (less than 6%). These results were ascribed to the presence of guanidine group in [CH][Arg]. Guanidine group (pKa = 13.8 [29], Fig. S2) in [CH][Arg] can effectively be combined with H^+ dissociated from glycerol to form a relatively stable protonated nitrogen atom in the =NH group, which can enhance the glycerolysis reaction. In fact, the outstanding performances of guanidine-based catalysts were also found in other reactions [30–32].

Fig. 1 shows the effect of the cation of AAIL on catalytic activity and glycerolysis selectivity. The reactions reached equilibrium within 30 and 60 min under the presence of [TMA][Arg] and [TBA][Arg], respectively. However, when [CH][Arg] was used as a catalyst, TAG conversion and MAG yield all increased after 4 h. Compared with other AAILs, [TMA][Arg] showed the most effective performance by obtaining high TAG conversion (64.63 ± 2.13%) and MAG yield (54.85 ± 1.59%) in a short time (30 min). These results were mainly attributed to the electronic and steric hindrance effects of alkyl in cations ($[TMA]^+$, $[CH]^+$, and $[TBA]^+$). Therefore, [TMA][Arg] was selected and used for the next glycerolysis of soybean oil.

Table 1

Glyceride content of the reaction catalyzed by AAILs with cholinium as the cation. Glycerolysis conditions: soybean oil/glycerol molar ratio 1:2, 100 °C, 12 h, and 10% catalyst load (based on soybean oil weight).

Ionic liquids	Structure of anion	Properties of side chains of the amino acid	Isoelectric point of parent amino acid	Content (%)		
				MAG	DAG	TAG
[CH][Arg]		Basic	10.76	59.64 ± 1.23	18.56 ± 0.59	21.80 ± 1.01
[CH][Lys]		Basic	9.66	5.81 ± 0.96	4.94 ± 1.26	93.25 ± 1.20
[CH][His]		Basic	7.60	3.95 ± 1.15	2.51 ± 0.69	94.54 ± 1.45
[CH][Trp]		Neutral	5.89	2.18 ± 1.03	2.66 ± 1.02	95.16 ± 2.21
[CH][Glu]		Acidic	3.15	2.22 ± 089	2.69 ± 0.39	95.09 ± 1.76

It is worth noting that Arg could not catalyze the glycerolysis process by itself (Fig. 1), which indicated that the formations of AAILs enabled Arg to have the catalytic activities for the glycerolysis. Similar finding can also be reported in biodiesel preparation [26]. Meanwhile, the catalytic activity of each component ($[TMA]^+$ or $[TBA]^+$) has also been improved by the formation of AAILs with non-catalytic Arg. These AAILs have also been used as phase transfer catalysts in transesterification or intensification [33,34].

Compared with the traditional catalysts, for the glycerolysis, [TMA][Arg] had the advantages of low temperature, high reaction rate, high MAG yield, and low cost. Especially, compared with enzyme catalyst, [TMA][Arg] had the advantages of fast reaction rate and low cost. For examples, the glycerolysis of soybean oil catalyzed by [TMA][Arg] reached equilibrium within 0.5 h, which was great shorter than the enzymatic glycerolysis (3–8 h) [35,36]. The prices of the components (arginine and tetramethylammonium hydroxide pentahydrate) of AAIL [TMA][Arg] are 0.08 $/g and 0.13 $/g, respectively, which were great cheaper than the immobilized enzyme Novozym 435 (2.48 $/g).

3.2. Effect of reaction temperature

Generally, elevating temperature is conducive to the thermodynamics reaction. To investigate the effect of temperature on the glycerolysis catalyzed by [TMA][Arg], the experiments were performed under the following reaction conditions: molar ratio of 1:2 soybean oil/glycerol and 10% [TMA][Arg] load (Fig. 2). Results showed that the glycerolysis rates of soybean oil were accelerated by increasing temperature in the range of 80–120 °C (Fig. 2C). Besides, TAG conversion and MAG yield also increased when the temperature increased from 80 °C to 100 °C. The maximum TAG conversion (81.11 ± 2.26%, Fig. 2B) and MAG yield (64.85 ± 1.26%, Fig. 2A) were achieved at 100 °C. Furthermore, when reaction temperature was higher than 100 °C, TAG conversion and MAG yield did not increase. Therefore, the optimum temperature catalyzed by [TMA][Arg] was 100 °C, which was great lower than MAG preparation using ionic liquid [Bmim]Im as a catalyst (200 °C) [16], and also lower than that of traditional alkane catalyst (>200 °C) [15].

According to the previous reports [37,38], it is difficult to obtain a MAG yield of 60% catalyzed by inorganic basic catalysts, such as KOH, $Ca(OH)_2$, NaOH, and MgO, even though at high temperature (220–240 °C). Meanwhile, high temperature was responsible for poor product quality, greater energy consumption, and the formation of glycerol polymerize [39]. Compared with these traditional catalysts, [TMA][Arg] exhibited a higher activity for the glycerolysis and higher MAG yield (~65%) at lower temperatures (100 °C). These phenomena were related to the lower activate temperature of [TMA][Arg], which was confirmed by the low activation energies (Ea) of TAG conversion (37.58 kJ/mol) and MAG formation (44.22 kJ/mol) in the presence of [TMA][Arg] (Fig. 2C).

3.3. Effect of substrate ratio

The glycerolysis of soybean oil catalyzed by alkaline catalyst is beneficial to the formation of MAG [16,37,38]. In this work, more glycerol was used to achieve the complete conversion of soybean oil and high MAG yield. In final MAG product, excessive glycerol and AAIL [TMA][Arg] can be removed by washing with water. As shown in Fig. 3, with the increase of glycerol amount, MAG yield and TAG conversion significantly increased, and reached the maximum (64.85 ± 2.13% and 86.11 ± 1.65%, respectively) at 1:2 M ratio of soybean oil and glycerol, because the increase in the amount of glycerol can produce more glycerol anion, which is conducive to the glycerolysis of soybean oil and intermediate DAG to produce more MAG. However, a further increase of glycerol (soybean oil/glycerol molar ratio at 1:4) did not enhance TAG conversion or MAG yield. On the contrary, it decreased the positive glycerolysis rate, which was due to its high viscosity and great mass transfer limitation.

3.4. Effect of catalyst [TMA][Arg] load

The effect of [TMA][Arg] load on the glycerolysis reaction was shown in Fig. 4. The glycerolysis rates increased continuously with

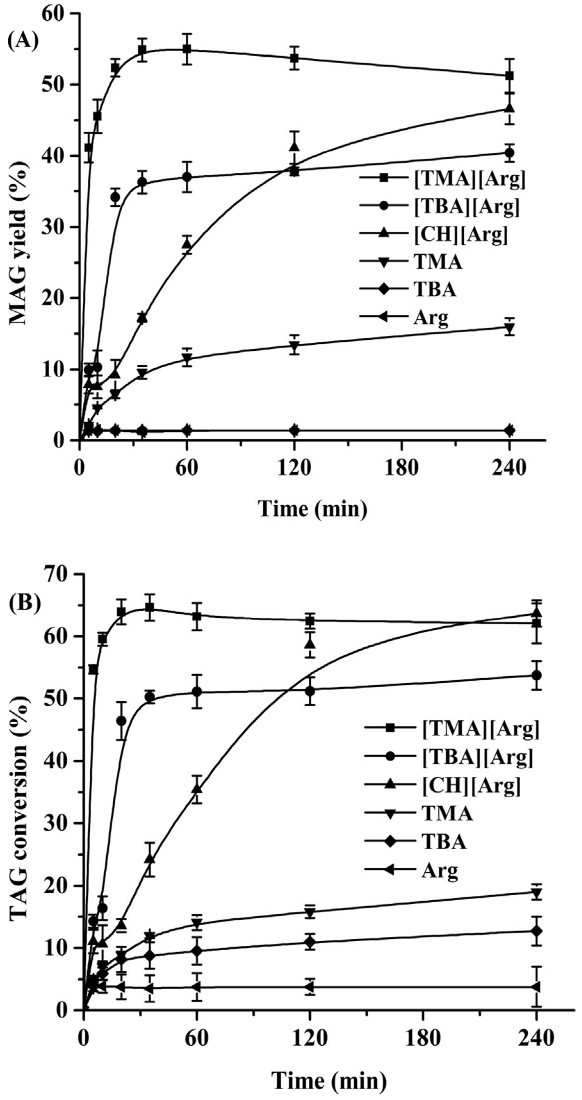


Fig. 1. Catalytic activity of synthesized AAILs with selected Arg and their substrates for the glycerolysis process: (A) MAG yield, (B) TAG conversion. Glycerolysis conditions: soybean oil/glycerol molar ratio 1:2, 100 °C, and 10% catalyst load (based on soybean oil weight).

the increase of [TMA][Arg] load in the range of 4–12%. When 10% [TMA][Arg] load was used, the maximum TAG conversion and MAG yield were 86.11 ± 1.65% and 64.84 ± 1.56%, respectively. These were ascribed to the increasing amount of catalytic active sites with high [TMA][Arg] load, which was favorable for the glycerolysis reaction [40]. In addition, [TMA][Arg] is a liquid catalyst, which can overcome the mass transfer limitation. With a further increase of [TMA][Arg] load (>10%), the time to reaction equilibrium shortened. However, TAG conversion and MAG yield had no obvious change. Therefore, 10% catalyst load was enough for this glycerolysis.

3.5. The glycerolysis mechanism catalyzed by [TMA][Arg]

According to the GC results, TAG, DAG, and MAG are the main substances in the product of [TMA][Arg]-catalyzed the glycerolysis of soybean oil. According to Fig. 5, extending reaction time can

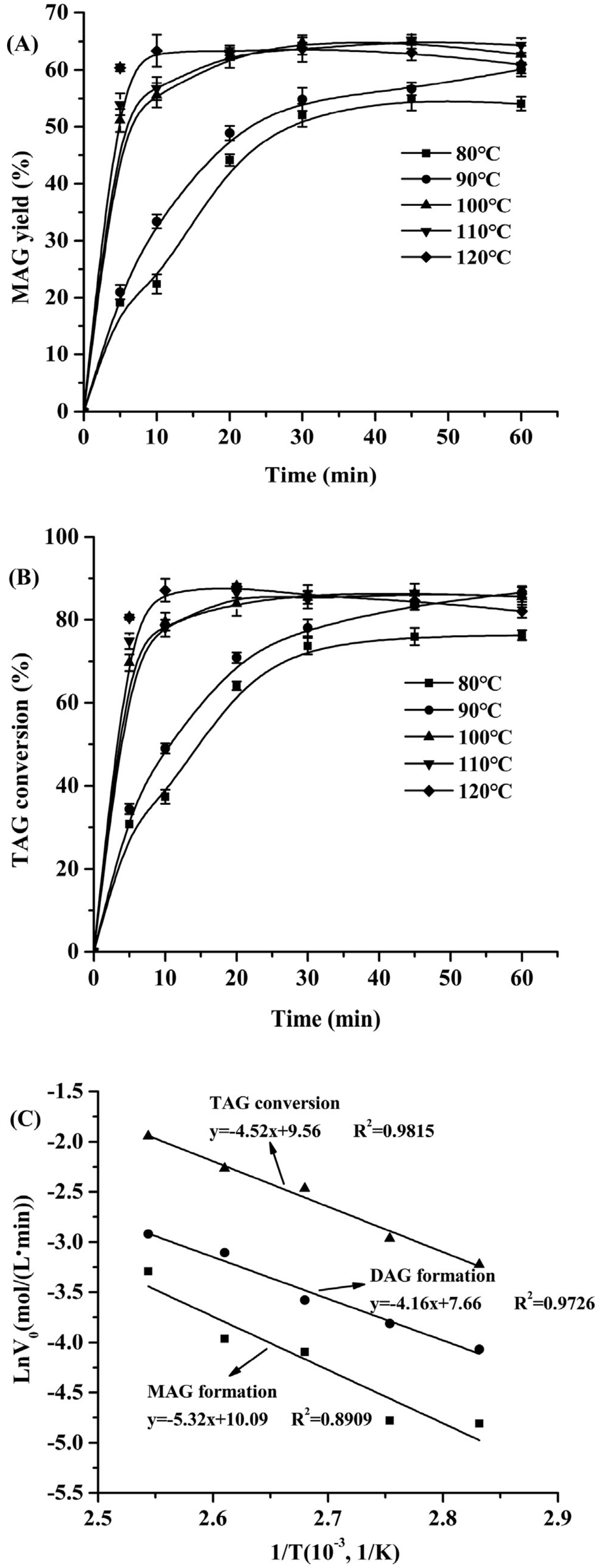


Fig. 2. Effect of reaction temperature on the process: (A) MAG yield, (B) TAG conversion. (C) Relationship between the initial glycerolysis rate and temperature. Glycerolysis conditions: soybean oil/glycerol molar ratio 1:2 and 10% [TMA][Arg] load (based on soybean oil weight).

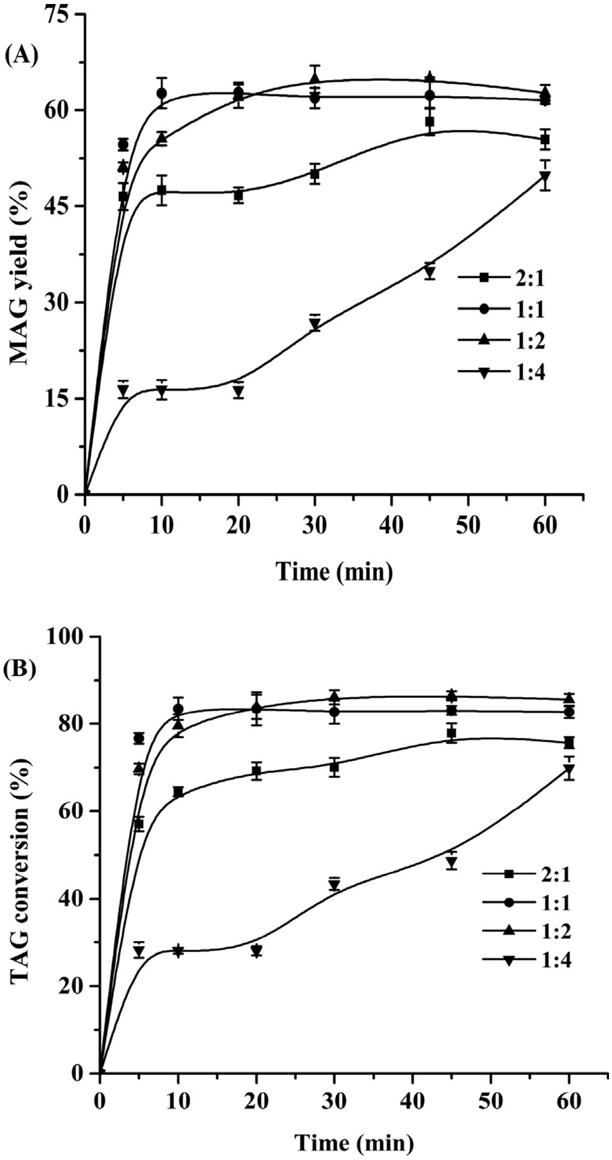


Fig. 3. Effect of soybean oil/glycerol molar ratio on the process: (A) MAG yield, (B) TAG conversion. Glycerolysis conditions: 100 °C and 10% [TMA][Arg] load (based on soybean oil weight).

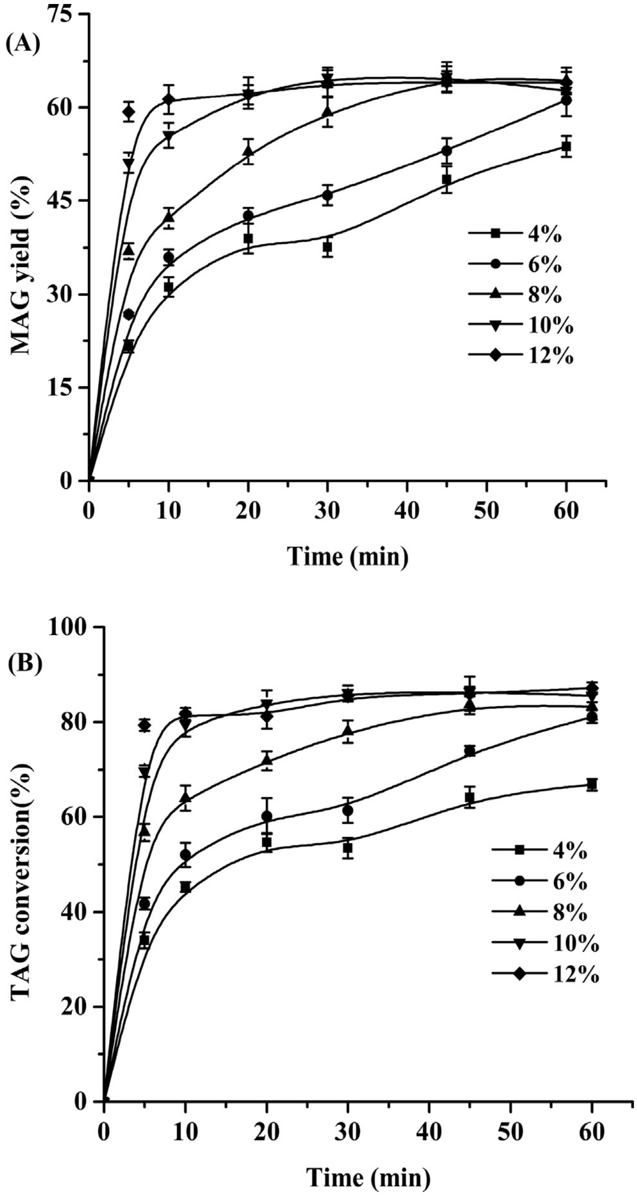


Fig. 4. Effect of [TMA][Arg] load on glycerolysis process: (A) MAG yield, (B) TAG conversion. Glycerolysis conditions: soybean oil/glycerol molar ratio 1:2 at 100 °C.

improve TAG conversion and the formations of MAG and DAG. Before the reaction reached equilibrium (<0.5 h), TAG conversion, MAG and DAG contents in the product increased with reaction process, which indicated that DAG formation rate was faster than its consumption rate. The reaction reached equilibrium at 0.5 h. Meanwhile, TAG conversion, DAG and MAG contents in the product were 86.11 ± 2.12%, 21.27 ± 2.26%, and 64.84 ± 2.26%, respectively. When the glycerolysis time was longer than 0.5 h, the contents of DAG and MAG showed no obvious change.

According to Fig. 5 and the reports of the alkaline-catalyzed transesterification [2,16,26], the reaction mechanism of the glycerolysis of soybean oil with glycerol to produce MAG catalyzed by [TMA][Arg] was proposed, and the possible reaction procedure was illustrated in Scheme 1. Firstly, the guanidine group of Arg^- was obtained a proton from the hydroxyl group of glycerol, and then glycerol was activated to form a glycerol anion (G^-). The carbonyl carbon with positive charge of TAG was attacked by the nucleophiles (G^-) to produce an intermediate [16,41]. After electron was transferred and the bond was ruptured, the intermediate was split into diacylglycerol anions (DAG^-) and MAG. Then DAG^- was converted into DAG by associating with the protonated guanidinium group. And then DAG was reacted with G to produce MAG in the same way. Meanwhile, the hydrophilic amino moieties favor the shift of reaction equilibrium towards to MAG formation by hydrogen-bond in the presence of [TMA][Arg].

3.6. Model fitting and response surface analysis

Response surface design was carried out to optimize the effect of experimental factors (substrate ratio, temperature, and [TMA][Arg] load), and the results were shown in Table 2. The model of MAG formation was evaluated by multiple regression analysis, and the results were shown in Table 3 and Fig. 6. By fitting the experimental data in Table 2, the quadratic polynomial regression equation of MAG yield was obtained as follows:

$$\begin{aligned} MAG\ yield\ (\%) = {} & 64.47 + 4.45A - 10.80B + 2.31C - 1.15AB \\ & - 0.73AC - 0.50BC - 5.11A^2 - 24.70B^2 \\ & - 2.97C^2 \end{aligned} \quad (3)$$

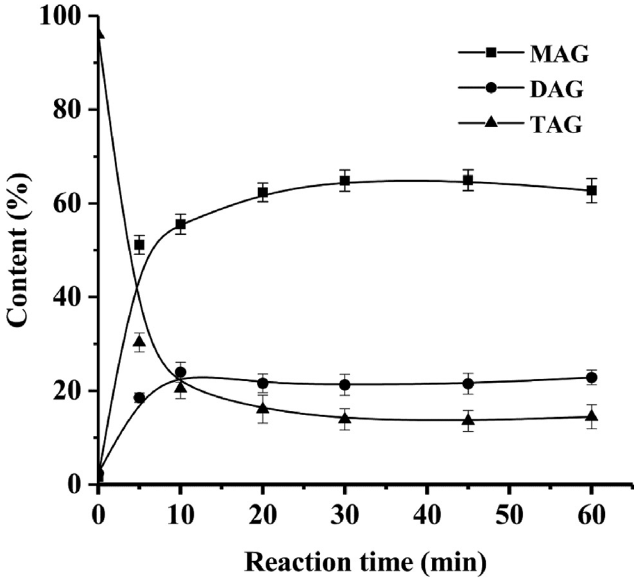


Fig. 5. Changes of components in the product with reaction process. Glycerolysis conditions: 10% [TMA][Arg] load, soybean oil/glycerol molar ratio 1:2 and 100 °C.

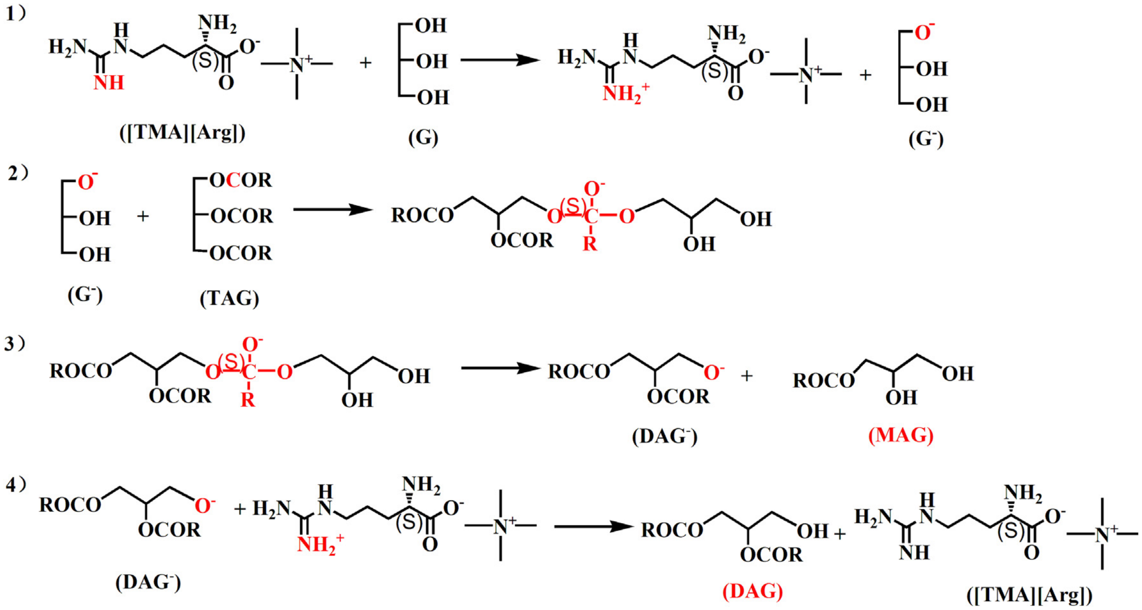
Table 3 suggested that the model of MAG yield ($P < 0.0001$) was extremely significant. The correlation coefficient (R^2) was 0.995, which indicated that the regression equation had a good fitting and could predict the actual result of the glycerolysis. In the prediction model of MAG yield (Table 3), the effects of three factors and their quadratic terms were significant, but the interaction was not significant. According to Table 3 and Fig. 6, the effects of three factors on MAG yield decreased in the order of substrate ratio > glycerolysis temperature > [TMA][Arg] load. The optimized conditions was obtained as: 105 °C, [TMA][Arg] load 10%, and substrate ratio 1:2. Under these optimized conditions, the maximum MAG yield was 64.89 ± 1.26%, which was higher than those of NaOH, KOH, $Ca(OH)_2$, or MgO at 240 °C (40–60%) [16].

Table 2
Response surface design and results of MAG yield.

Trials	A: Temperature (°C)	B: Substrate ratio (mol/mol)	C: [TMA][Arg] (%)	MAG yield (%)
1	0 (100)	0 (1:2)	0 (10)	64.84 ± 0.91
2	1 (120)	1 (1:3.5)	0 (10)	26.83 ± 0.61
3	−1 (80)	0 (1:2)	1 (12)	55.00 ± 1.14
4	0 (100)	−1 (2:1)	1 (12)	50.08 ± 1.27
5	0 (100)	1 (1:3.5)	1 (12)	26.84 ± 1.64
6	−1 (80)	1 (1:3.5)	0 (10)	21.54 ± 1.10
7	0 (100)	−1 (2:1)	−1 (8)	45.78 ± 1.40
8	1 (120)	0 (1:2)	−1 (8)	59.27 ± 0.44
9	−1 (80)	−1 (2:1)	0 (10)	40.20 ± 0.95
10	0 (100)	1 (1:3.5)	−1 (8)	24.53 ± 1.92
11	0 (100)	0 (1:2)	0 (10)	64.45 ± 1.69
12	1 (120)	0 (1:2)	1 (12)	63.75 ± 1.44
13	0 (100)	0 (1:2)	0 (10)	63.97 ± 1.72
14	0 (100)	0 (1:2)	0 (10)	64.54 ± 0.93
15	0 (100)	0 (1:2)	0 (10)	64.56 ± 2.19
16	−1 (80)	0 (1:2)	−1 (8)	47.58 ± 0.69
17	1 (120)	−1 (2:1)	0 (10)	50.08 ± 1.41

Table 3
ANOVA analysis of glycerolysis variables on MAG yield.

Source	Sum of squares	df	Mean square	F-value	p-value
Model	3975.68	9	441.74	376.50	<0.0001
A-Temperature	158.60	1	158.60	135.17	<0.0001
B-Substrate ratio	932.98	1	932.98	795.18	<0.0001
C-[TMA][Arg]	42.82	1	42.82	36.49	0.0005
AB	5.25	1	5.25	4.47	0.0722
AC	2.16	1	2.16	1.84	0.2172
BC	0.99	1	0.99	0.84	0.3887
A^2	109.83	1	109.83	93.61	<0.0001
B^2	2369.09	1	2569.09	2189.64	<0.0001
C^2	37.02	1	37.02	31.55	0.0008
Residual	8.21	7	1.17		
Lack of Fit	7.81	3	2.60	25.76	0.0045
Pure Error	0.40	4	0.10		
Total	3983.89	16			
$R^2 = 0.995$					



Scheme 1. Reaction mechanism of the glycerolysis catalyzed by [TMA][Arg].

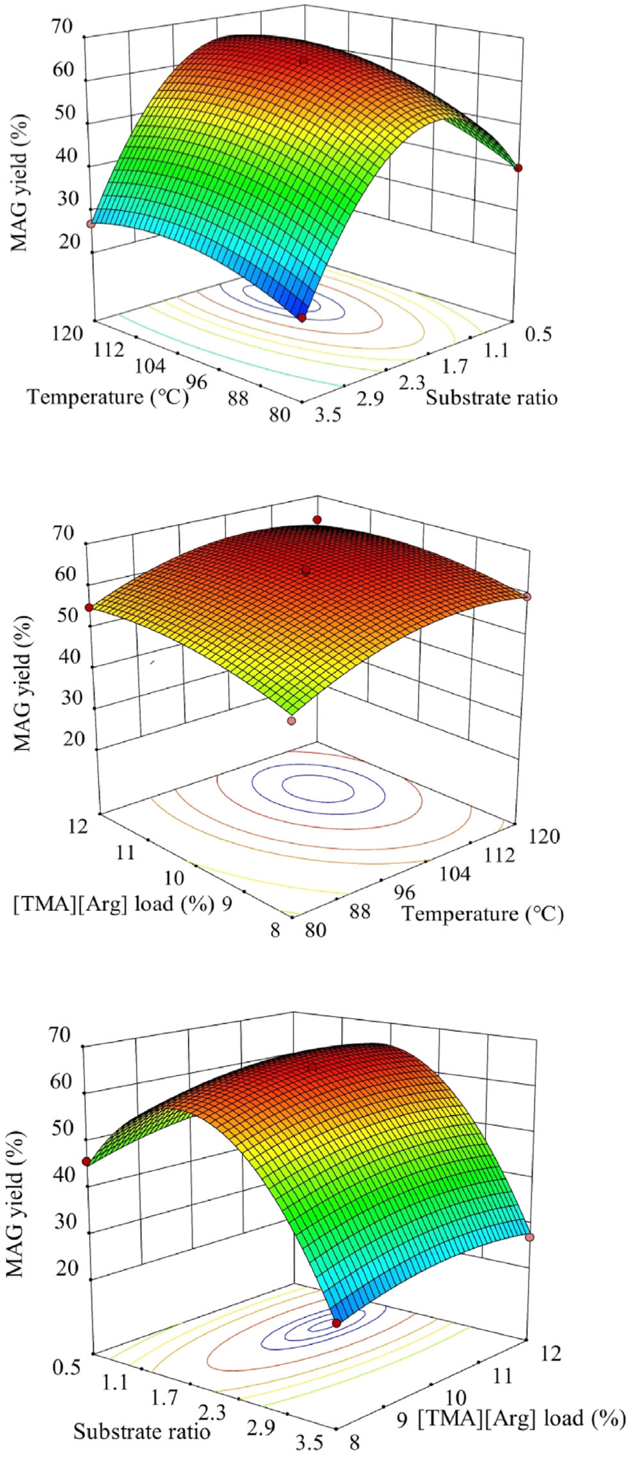


Fig. 6. Model graphs: interaction effect of [TMA][Arg] load, substrate rate, and temperature on MAG yield.

4. Conclusions

In this work, a bio-compatible AAIL [TMA][Arg] was successfully developed as a novel catalyst for MAG synthesis by glycerolysis of soybean oil. Compared with the traditional catalysts, [TMA][Arg] had the advantages of low temperature, high reaction rate, high MAG yield, and low cost. The effects of reaction factors on MAG yield was substrate ratio > glycerolysis temperature > [TMA][Arg] load. High TAG conversion (86.31 ± 1.68%) and MAG yield (64.89 ± 1.26%) were achieved under the optimal conditions of 1:2 M ratio of soybean oil and glycerol and 10% catalyst load at 105 °C for 0.5 h. The activation energies of MAG formation and TAG conversion were 44.22 kJ/mol and 37.58 kJ/mol, respectively. Besides these, green sources of raw materials and high catalytic performance made [TMA][Arg] have great potential in glycerolysis procedure.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The authors sincerely acknowledge the financial support from National Natural Science Foundation of China (32072255).

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.molliq.2021.117231.

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