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1	Investigation on cold start issues of methanol engines and its
2	improvement from the perspective of droplet evaporation
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18 Abstract

The cold start issue of methanol engines limits their wide application in cold 19 seasons and regions. To explore the underlying mechanisms and propose effective 20 improvement measures, the evaporation characteristics of methanol at 243-303 K were 21 22 first investigated by the single droplet method in this study. The effects of ambient temperature, initial diameter, fuel temperature and intake air flow velocity on 23 evaporation of methanol droplets were quantitatively analyzed and their guidance for 24 improving the cold start performance of methanol engines were discussed. The results 25 26 revealed that the evaporation process of methanol droplets in low temperature and humid environment showed two-stage feature due to its hygroscopicity, including pure 27 methanol evaporation and water-dominated evaporation of methanol-water mixture, but 28 29 it could be regarded as the evaporation of a pseudo single component and the same was true for other binary mixtures. The existence of water in methanol droplets not only led 30 to their incomplete evaporation at temperatures below 263 K, but also caused the linear 31 32 change of their evaporation rates with temperature, which was different from the exponential change of pure methanol evaporation rate with temperature. Increasing the 33 ambient temperature from 243-283 K to 293 K was optimal for promoting methanol 34 evaporation. Reducing droplet diameter inhibited the water absorption of methanol 35 droplets, thus enhancing their evaporation rates. Increasing fuel temperature could not 36 promote droplet evaporation, but mainly influenced water absorption of methanol 37 droplets. The promotion effect of air flow on methanol evaporation became weaker with 38 the increase of intake air flow velocity, especially when it exceeded 3 m/s. The findings 39

- 40 of this study suggest that multiple methods should be combined to improve the cold
- 41 start performance of methanol engines.
- 42 *Keywords:* Cold start issues; Droplet evaporation; Low temperature; Initial diameter;
- 43 Fuel temperature; Intake air flow velocity

44	Hi	ghl	lights	5
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45 •	Methanol droplets could not completely evaporate at temperatures below 263 K
46	due to its hygroscopicity.
47 •	Raising ambient temperature from 243-283 K to 293 K was optimal for promoting
48	evaporation.
49 ●	The evaporation of binary mixtures could be regarded as the evaporation of a
50	pseudo single component.
51 •	The water absorption of methanol droplets was inhibited by reducing droplet
52	diameter.
53 •	Increasing fuel temperature could not promote droplet evaporation.
54	



55 Graphical abstract

Nomenclature

Abbreviations

PM	Particulate matters	BMEP	Brake mean effective pressure
NO _x	Nitrogen oxides	BTE	Brake thermal efficiency
UHC	Unburned hydrocarbons	BSFC	Brake specific fuel consumption
CO ₂	Carbon dioxides	IMEP	Indicated mean effective pressure
СО	Carbon monoxide	CVC	Constant volume chamber

SI Spark ignition

Symbols

d	droplet diameter
d_0	initial droplet diameter
d_1	droplet diameter when evaporation is completed
σ	normalized squared diameter when evaporation is completed
τ_1	time of the first stage
τ_2	time of the second stage
τ _i	ideal normalized droplet lifetime
τ_{a}	actual normalized droplet lifetime
ti	ideal droplet lifetime
ta	actual droplet lifetime
K_1	average evaporation rate of the first stage
K_2	average evaporation rate of the second stage

- K_a average evaporation rate of methanol droplets
- K_w average evaporation rate of water droplets
- K_c calculated value of average evaporation rate of methanol droplets
- K_p evaporation rate constant of the pseudo single component
- P₁ proportion of the first stage
- P₂ proportion of the second stage
- P_{K1} improved percent of K_1
- P_{K2} improved percent of K_2
- P_{Ka} improved percent of K_a
- $P_{\tau} \qquad \text{ increased percent from } \tau_i \text{ to } \tau_a$
- $P_{\tau i}$ relative change (increased percent or decreased percent) of τ_i
- $P_{\tau a}$ relative change (increased percent or decreased percent) of τ_a
- η_{K1} improvement efficiency of K_1
- η_{K2} improvement efficiency of K_2
- η_{Ka} improvement efficiency of K_a
- ΔT Value of increased ambient temperature
- $\Delta T'$ Value of increased fuel temperature
- V intake air flow velocity
- T_d droplet temperature
- RH relative humidity
- AH absolute humidity

59 **1 Introduction**

Automobiles are still predominately powered by traditional hydrocarbon fuels 60 nowadays, inevitably leading to significant harmful pollutants such as particulate 61 matters (PM), nitrogen oxides (NO_x) and unburned hydrocarbons (UHC) [1]. Further, 62 the production of carbon dioxides (CO_2) also exacerbates the greenhouse effect [2]. The 63 urgent need to achieve the carbon neutralization goal drives the engine industry to 64 search for green, low-carbon and renewable alternative fuels [3]. Methanol has received 65 extensive attention due to its unique advantages [4]. On the one hand, methanol can be 66 67 produced by both fossil fuels (e.g., coal, biomass, natural gas and coke oven gas [5]) and the synthesis of CO₂ captured in the air and hydrogen obtained by electrolysis of 68 water from renewable energies [6], so as to realize the green cycle of CO_2 to fuel. On 69 70 the other hand, methanol has a high flame speed [7], and its low C/H ratio produces less CO₂ than gasoline. The high oxygen content reduces the formation of carbon monoxide 71 (CO) [8]. The high latent heat of vaporization and more ternary molecules in the 72 73 combustion products lead to lower in-cylinder temperature [9], not only reducing NO_x emissions and heat loss, but also providing better anti-knock performance [10]. The 74 high-octane number allows higher compression ratios [11], thereby improving engine 75 thermal efficiency [12]. Furthermore, methanol is liquid at standard temperature and 76 pressure [13], making it easier to store and transport [14]. 77

Many scholars have studied the performance of methanol-fueled engines. Xu et al.
[15] experimentally and numerically studied the engine performance and emission
characteristics of methanol and iso-octane in a heavy-duty direct-injection compression

81	ignition engine operating in partially premixed combustion (PPC) regimes. The results
82	showed that the lower CO and NO _x emissions as well as much higher combustion
83	efficiency with a similar power output were achieved for methanol as compared to iso-
84	octane. Celik et al. [16] comparatively investigated the performance and emission
85	characteristics of methanol and gasoline in a single-cylinder engine at different
86	compression ratios. They found that methanol produced less CO, CO_2 and NO_x
87	emissions than gasoline at compression ratio of 6, with insignificant loss of power.
88	Knock occurred in gasoline mode but not in methanol mode when the compression ratio
89	increased from 6 to 10. The power and brake thermal efficiency increased by 14% and
90	36%, respectively, while the CO, CO_2 and NO_x emissions decreased by 37%, 30% and
91	22%, respectively. Balki et al. [17] studied the effects of compression ratio on the
92	performance of a single-cylinder low-power spark ignition (SI) engine fueled with pure
93	methanol and unleaded gasoline. The results showed that methanol had higher brake
94	mean effective pressure (BMEP), cylinder pressure, brake thermal efficiency (BTE)
95	and brake specific fuel consumption (BSFC), and lower UHC, CO and NO _x emissions
96	than that of gasoline at compression ratios of 8.5-9.5. When the compression ratio
97	increased from 8.0 to 8.5, the BMEP, BTE and BSFC of methanol increased by 10.5%,
98	4.5% and 30.2% respectively. Zhu et al. [18] studied the performance of a pure
99	methanol engine modified from a multi-cylinder port fuel injection SI heavy-duty
100	natural gas engine. The results showed that the pure methanol engine reached a peak
101	BTE of 41.4%, which was 3% higher than the original natural gas engine. NO _x and CO
102	emissions were reduced, while UHC emissions were increased. Meanwhile, the

coefficient of variation of the indicated mean effective pressure (IMEP) remained
below 2% at EGR rate of 31%. In summary, the above studies showed that methanol
engines have great advantages in energy saving and emission reduction. Combined with
green production, methanol engines have broad application prospects.

However, methanol evaporates slowly at low ambient temperatures due to its low 107 vapor pressure and high latent heat of vaporization [10], resulting in cold start issues of 108 methanol engines. Li et al. [19] concluded that methanol engines cannot start reliably 109 without auxiliary devices under an ambient temperature below 289 K even when a large 110 111 amount of methanol is injected, which limits the use of methanol engines in cold seasons and regions. Therefore, to promote the application of methanol engines, it is of 112 great significance to explore the evaporation characteristics of methanol at low 113 114 temperatures and find ways to improve its evaporation rate.

In real engines, the fuel is injected into combustion chamber through high pressure 115 injectors and evaporates in the form of spray droplets to produce an ignitable mixture 116 117 before combustion and power output [20]. As a result, the engine performance is closely related to the evaporation of droplets. Thus, it is a common and necessary method to 118 investigate the fuel evaporation characteristics from the perspective of a single droplet 119 [21]. Many scholars have used this method to study the evaporation characteristics of 120 methanol fuel. Yuan et al. [22] studied the evaporation characteristics of methanol 121 droplets at ambient temperatures of 323-473 K. They found that methanol droplets 122 evaporated linearly when the ambient temperature was higher than 373 K, while the 123 evaporation process of methanol droplets was divided into two linear evaporation 124

stages at temperatures below 373 K, which was caused by the hygroscopicity of 125 methanol. Wang et al. [23] studied the evaporation characteristics of methanol droplets 126 127 at ambient temperatures of 473-1087 K and reported a similar conclusion: the normalized square diameter of methanol droplets decreased linearly with time. The 128 evaporation of methanol droplets in different humidity environments at an ambient 129 temperature of 297 K was investigated by Law [24]. The results showed that methanol 130 droplets evaporated linearly when the humidity was 0%. As the humidity increased, the 131 droplet normalized diameter curve became a downwards concave and the droplet 132 lifetime prolonged. Hegseth et al. [25] studied the evaporation characteristics of 133 methanol droplets at an atmospheric pressure and an ambient temperature of 293 K. 134 They found a convection driven by surface tension gradient inside the droplets during 135 136 evaporation. Kosasih et al. [26] investigated the heat and mass transfer of methanol droplets at ambient temperatures of 308-328 K and air flow rates of 0.1-0.7 m/s. 137 However, the ambient temperatures in above studies were all over 293 K. There is 138 139 a research gap in the evaporation of methanol droplets at temperatures below 293 K, which is of great significance for the cold start of methanol engines. Depending on the 140 seasons and regions, the start-up temperature of an engine usually changes between 243 141 and 303 K. Therefore, this study investigated the evaporation characteristics of 142 methanol droplets at a wide range of ambient temperatures between 243-303 K. The 143 effects of ambient temperature, initial droplet diameter, fuel temperature and intake air 144 flow velocity on the evaporation of methanol droplets were quantitatively analyzed, 145 and their implications for designing methanol engines were discussed. The findings of 146

this research are expected to provide great theoretical and practical guidance for
tackling the cold start issues of methanol engines and thus promote their applications
in cold seasons and regions.

150

151 **2 Material and methods**

152 **2.1 Experimental setup**

Fig. 1 shows the schematic of the experimental setup for the evaporation of 153 methanol droplets at low temperatures. It consisted of a droplet generation and transport 154 155 system, a temperature control system, a fuel heating system, an inlet air flow system and an image acquisition system. In the droplet generation and transport system, as the 156 stepper motor I runs, a given volume of liquid was squeezed out by a microliter syringe 157 158 and then a droplet with the specific diameter was formed on the K-type thermocouple with the diameter of 0.08 mm, which was sent to the constant volume chamber (CVC) 159 by stepper motor III. According to the conclusion of Rehman et al. [27], the effect of 160 thermocouple was ignorable when the ratio of the square of thermocouple diameter to 161 the square of initial droplet diameter was less than 0.01. Considering that the diameter 162 of the thermocouple was 0.08 mm and the diameters of the droplets in the experiment 163 exceeded 0.8 mm, the effect of heat conduction by thermocouple on droplet evaporation 164 could be negligible. The temperature control was achieved by the environmental 165 chamber which could change between 233 and 353 K. Three K-type thermocouples 166 were evenly arranged in the CVC to ensure uniform ambient temperature. The accuracy 167 of temperature control was ± 1 K. The fuel heating system was used to change the 168

methanol temperature before the evaporation, which was reached by the heating plate 169 pasted on the baffle below the droplet. Since the heating plate was exposed to the cold 170 171 environment, a large amount of heat generated by the heating plate was dissipated to the environment and only a small part of the heat was used to improve the fuel 172 temperature, which ultimately led to a smaller increase in fuel temperature. The 173 174 maximum value of increased fuel temperature was only 8.5 K in this experiment. After the baffle was removed by the stepper motor II, the droplets with different fuel 175 temperatures were sent to the CVC. The inlet air flow system mainly included an intake 176 port manufactured by three-dimensional printing, a flowmeter (1.5% accuracy) and a 177 blower, which could vary the intake air flow velocity. In the image acquisition system, 178 a LED lamp, an industrial camera (MV-CS060-10GM) and a tele-macro lens (Nikon 179 180 Micro-ED 200 mm f/4) were combined to get the droplet images with the resolution of 1072×1548 pixels. Due to the slow evaporation of methanol droplets at low 181 temperatures, the sampling frequency was changed between 1 and 18 frames per second 182 183 (fps).



The evaporation characteristics of methanol droplets with various initial diameters were first investigated at ambient temperatures of 243-303 K. Then the fuel temperature and intake air flow velocity were increased to study their promotion effects on the evaporation of methanol droplets at 273 K. The specific experimental conditions were presented in **Table 1**.

193	Table 1 Experimental conditions for droplet evaporation.		
	Ambient pressure (atm)	1	
	Ambient temperature (K)	243-303	
	Initial diameter (mm)	0.816-1.604	
	Value of increased fuel temperature (K)	0-8.5	
	Intake air flow velocity (m/s)	0-3.20	

2.3 Data processing 195

Droplet diameter was a key parameter in the evaporation process, which was 196 obtained by a MATLAB code. The procedures of image processing were shown in Fig. 197 2. Firstly, a region of 300*300 with the droplet at the center was cut out from the raw 198 image (1072*1548). Secondly, the image was binarized by a proper threshold. Then 199 thermocouple wire on both sides of the droplet was removed. Finally, the droplet 200 boundary was extracted from the image and the pixel number (N) of the droplet was 201 obtained. The droplet diameter (d) was calculated by Eq. (1) where l was the image 202 spatial resolution: 203



205

206

Fig. 2. The procedures of image processing.

Droplet lifetime and evaporation rate were also significant parameters to evaluate 207 the evaporation, which were described in Fig. 3. The evaporation process of methanol 208 droplets included two linear stages and the demarcation point between them was 209

210 determined by the maximum value of the second derivative of the concave part of droplet diameter curve. The average evaporation rates of the first and second stages 211 212 were defined as K₁ and K₂, respectively which were obtained by linear fitting for each stage. The times of two stages were defined as τ_1 and τ_2 , respectively, and their sum 213 was the actual normalized lifetime τ_a . The ideal normalized lifetime, τ_i , was defined by 214 Eq. (2), where σ was the corresponding normalized squared diameter at the end of 215 evaporation, which represented the lifetime of methanol droplets without the 216 disturbance of water vapor. The difference between τ_a and τ_i indicated the effect of water 217 218 vapor on the evaporation of methanol droplets. In addition, the average evaporation rate of methanol droplets in the entire evaporation process, K_a, was calculated by the Eq. 219 (3), where d_0 was the initial droplet diameter, d_1 was the droplet diameter when the 220 221 evaporation was completed and t was the evaporation time.

222
$$\tau_i = (1 - \sigma)/K_1$$
 (2)

$$K_a = (d_0^2 - d_1^2)/t \tag{3}$$



224

223

Fig. 3. The parameters characterizing the evaporation of methanol droplets.

226

227 2.4 Experimental errors and uncertainties

The errors of droplet characteristic parameters were mainly caused by the random error of experiment and the error of droplet boundary identification. Experiment was repeated five times to minimize the random error (δ) which was calculated by Eq. (4):

231
$$\delta = \sqrt{(x_1 - \bar{x})^2 + ((x_2 - \bar{x})^2) + \dots + ((x_n - \bar{x})^2)/(n - 1)}$$
(4)

where $x_1, x_2 \cdots x_n$ were the values of repeated experiments, \bar{x} was the average value, and *n* was the number of experiment.

Regarding the error of droplet boundary identification, if the experimental result *R* satisfied Eq. (5) [28], [29] [30], the relative uncertainty of *R* was calculated by Eq. (6):

237
$$R = Y_1^{a_1} * Y_2^{a_2} * \dots * Y_n^{a_n}$$
(5)

238
$$\frac{\Delta R}{R} = \sqrt{(a_1 * \frac{\Delta Y_1}{Y_1})^2 + (a_2 * \frac{\Delta Y_2}{Y_2})^2 + \dots + (a_n * \frac{\Delta Y_n}{Y_n})^2}$$
(6)

where Y_i was the variable, and ΔR and ΔY_i were the uncertainties of R and Y_i , respectively. Assuming that a pixel was a square, then the droplet area could be calculated by Eq. (7) where L was the side length of pixel, D_0 was the diameter of alundum tube for calibration, M was the pixel number of alundum tube diameter, and N was the pixel number of the droplet.

244
$$S = L^2 * N = \left(\frac{D_0}{M}\right)^2 * N = D_0^2 * M^{-2} * N$$
(7)

Assuming that the droplet was a sphere, then the relative uncertainty of square of droplet diameter could be calculated by Eq. (8) where ΔN , ΔD_0 and ΔM were the uncertainties of the pixel number of the droplet, the diameter and pixel number of 248 alundum tube, respectively. In this study, the droplet diameter varied from 0.816 mm to 1.604 mm and D_0 was 2.5 mm (measured by a vernier caliper whose accuracy was 0.02 249 mm, so ΔD_0 was 0.02 mm). It was assumed that there was an error of two pixels when 250 251 measuring alundum tube diameter, so ΔM was 2. According to Morin et al. [31], the pixels on the droplet boundary were regarded as errors. For the droplet with diameter 252 of 0.816 mm, M, N and ΔN were 290, 6512 and 260, respectively, so $\Delta D^2/D^2$ was 253 4.52%. For the droplet with diameter of 1.604 mm, M, N and ΔN were 290, 25149 and 254 562, respectively, so $\Delta D^2 / D^2$ was 3.08%. Overall, it could be concluded that the 255 uncertainty of the droplet diameter was less than 5%. 256

257
$$\frac{\Delta D^2}{D^2} = \frac{\Delta S}{S} = \sqrt{\left(2 * \frac{\Delta D_0}{D_0}\right)^2 + \left(-2 * \frac{\Delta M}{M}\right)^2 + \left(\frac{\Delta N}{N}\right)^2}$$
(8)

258

259 **3. Results and discussion**

260 **3.1** Evaporation characteristics of methanol droplets at low temperatures

Fig. 4(a) plots the normalized squared diameters and temperatures of methanol 261 droplets at temperatures of 243-303 K. The droplet diameter and the time are 262 normalized to eliminate the effect of different initial diameters between repeated 263 experiments caused by surface tension [32]. As shown in Fig. 4(a), the evaporation 264 process of methanol droplets at 253-303 K is divided into two stages by the two linear 265 parts of the diameter curves and the two quasi-steady parts of the droplet temperature 266 curves, which is similar to the evaporation of binary mixture. It is because the first stage 267 is the evaporation of pure methanol and the second stage is the water-dominated 268 evaporation of methanol-water mixture [33]. The d^2 law is obeyed in each stage, which 269

is consistent with the evaporation of methanol droplets at ambient temperature of 323-270 373 K [22]. This is also the result of the hygroscopicity of methanol in humid 271 272 environment, because there is always water vapor in the air even when the ambient temperature is extremely low. The normalized squared diameter corresponding to the 273 274 turning point between the first and second stages gradually decreases with the decrease of ambient temperature, which suggests that more methanol is evaporated in the first 275 stage and the concentration of water in the mixture is increased. The reason is that less 276 water vapor is absorbed by the methanol droplets, which is attributed to the reduction 277 278 of number of water molecule in the environment caused by the lower saturation vapor pressure and slower evaporation of water. Fig. 4(b) describes the relative humidity and 279 absolute humidity at various temperatures. It can be seen from Fig. 4(b) that the relative 280 281 humidity is gradually reduced when ambient temperature increases, which is due to the simultaneous increase of the partial pressure and saturation vapor pressure of water 282 vapor in the air. To better evaluate the content of water vapor in the air, absolute 283 humidity is used to represent the mass of water vapor per unit volume of air. According 284 to Fig. 4(b), absolute humidity increases with temperature and reaches the maximum 285 value of 3.76 g/m^3 at 303 K, which is far less than the absolute humidity at temperature 286 of 298 K and relative humidity of 50%. This proves that the content of water vapor in 287 the air is low and its increase is limited when ambient temperature increases from 243 288 K to 303 K, leading to the little change of turning point between the first and second 289 stages. Owing to the relatively low humidity, the droplets are evaporated by over 85 % 290 in the first stage. Surprisingly, the absorbed water can still evaporate at 263 K when it 291

is mixed with methanol. Maybe it is the result of the hydrogen bonding interaction 292 between the hydroxyl of methanol and water [34], which promotes the water 293 evaporation. However, the methanol droplets cannot completely evaporate at 253 K, 294 and even the second stage is missing at 243 K, as presented in Fig. 4(a). The 295 experimental images show that the absorbed water is attached on the thermocouple and 296 basically not evaporated. This is due to the separation of methanol and water caused by 297 the low temperature and reduced concentration of methanol in the mixture, and the 298 residual methanol continues to evaporate, while the water freezes on the thermocouple. 299 300 Then the schematic of evaporation for methanol droplets is concluded in Fig. 4(c). It can be inferred that the freezing phenomenon of water can also occur on the wall once 301 the spray impinges on the engine wall in the cold start process of methanol engines, 302 303 whose effect on engine performance is needed to consider. With the decrease of ambient temperature, the difference in the droplet diameter curves is more obvious, which 304 implies the slower evaporation of methanol at lower temperatures. 305



306



Fig. 4. (a) Temporal evolution of the normalized squared diameters $(d/d_0)^2$ and temperatures (T_d) of methanol droplets at temperatures of 243-303 K. (b) The change of the relative humidity (RH) and absolute humidity (AH) with ambient temperature. (c) The schematic of evaporation for methanol droplets.

The average evaporation rates of the first and second stages, methanol droplets and pure water at various ambient temperatures, K₁, K₂, K_a and K_w are shown in **Fig.** 5. Apparently, the trends of K₁, K₂ and K_w are similar, that is, the average evaporation

319	rate varies exponentially with the ambient temperature, while Ka increases linearly with
320	the increase of ambient temperature. Then the empirical formulas for droplet
321	evaporation of single component and binary mixture at low temperatures can be
322	inferred: $K = A * \exp(-T/B) + C$ and $K = A * T + B$, respectively, where K is the
323	average evaporation rate, T is ambient temperature, A, B and C are both constant, which
324	is significant for droplet evaporation simulation. Meanwhile, another method
325	calculating the average evaporation rate of methanol droplets is proposed: $K_c = K_1 *$
326	$P_1 + K_2 * P_2$, where K _c is the calculated value of the average evaporation rate of
327	methanol droplets, P_1 and P_2 are the proportion of the first and second stages,
328	respectively. The comparison of K_c and K_a is shown in Fig. 5. It is found that K_c is in
329	good agreement with K _a , which verifies the accuracy of the proposed method. And this
330	method can also be used for binary mixture evaporation due to the same two-stage
331	feature of the evaporation process. On the other hand, the reduction from K_1 to K_2 is
332	noticeable (more than 64%), especially at temperature below 273 K, resulting in K_a
333	being less than K ₁ . It can be deduced that the addition of water slows down the
334	evaporation of methanol, while methanol accelerates the water evaporation.
335	Furthermore, K_2 is always larger than K_w , which proves that the second stage is the
336	water-dominated evaporation of methanol-water mixture rather than the evaporation of
337	pure water. The reduced difference between K_2 and K_w is observed with the decrease
338	of temperature. This is due to the increase of water concentration in the methanol-water
339	mixture at lower temperatures.



340

Fig. 5. Variation of the average evaporation rates of the first and second stages (K_1 and K_2), methanol droplets (K_a) and pure water (K_w) with ambient temperature. K_c is the calculated value of K_a by another method.

In the actual methanol engines, the larger injection quantity and higher latent heat 344 345 of vaporization of methanol as compared to gasoline can reduce the combustion chamber temperature [10]. This reduces the evaporation rate of methanol, which is the 346 main reason for the cold start difficulty of methanol engines. Therefore, the promotion 347 348 of methanol evaporation brought by increasing temperature at different initial ambient temperatures is quantitively analyzed. Fig. 6(a) illustrates that the improved percent 349 of the average evaporation rates of the first and second stages and methanol droplets, 350 P_{K1} , P_{K2} and P_{Ka} , both increases with the increased temperature ΔT for each initial 351 ambient temperature, but gradually weakens at higher initial ambient temperature. For 352 instance, P_{K1} is 106.13% at an initial temperature of 243 K, but only 19.41% at an 353 initial temperature of 293 K when ΔT is 10 K, as demonstrated in Fig. 5. On the other 354 hand, the improvement efficiency of the average evaporation rates of the first and 355

second stages and methanol droplets, η_{K1} , η_{K2} and η_{Ka} , are defined in Fig. 6(b) to 356 evaluate the benefit of increasing ambient temperature on promoting the evaporation 357 358 of methanol droplets. It can be observed that both of them are decreased with the increase of initial ambient temperature, and first increase and then decrease with ΔT 359 at each initial ambient temperature. There is a maximum value marked with '*' in Fig. 360 6(b), at which the improvement of evaporation rate brought by an increased 361 temperature of 1 K is maximum. The corresponding ΔT is 50, 40, 30, 20, 10 K for 362 initial ambient temperatures of 243-283 K, respectively. Interestingly, the 363 corresponding actual temperatures are both 293 K. This means that increasing the 364 ambient temperature from original of 243-283 K to 293 K in the cold start process is 365 the way to maximize the benefit of promoting the evaporation by increasing ambient 366 367 temperature for methanol engines.



368



370

371

Fig. 6. (a) Variation of the improved percent (P_{K1} , P_{K2} and P_{Ka}) of K_1 , K_2 and K_a with the increased temperature ΔT . $K^{\Delta T}$ is the K after the ambient temperature increases by ΔT and K^0 is the K at initial ambient temperature. (b) Variation of the improvement efficiency (η_{K1} , η_{K2} and η_{Ka}) of K_1 , K_2 and K_a with the increased temperature ΔT .

Fig. 7 shows the ideal and actual normalized lifetimes, the time of the first and 376 second stages, and their proportion in the entire lifetime. With the increase of ambient 377 temperature, the ideal and actual normalized lifetimes both decrease exponentially and 378 the latter is always greater than the former due to the presence of the second stage 379 caused by water vapor in the environment. The difference between them is related to 380 the ambient temperature and humidity. As shown in Fig. 7, the increased percent from 381 the ideal to the actual normalized lifetime, P_{τ} , has little change at 283-303 K due to 382 similar low humidity, but rise quickly due to the extremely slow evaporation of 383 methanol-water mixture in the second stage when the ambient temperature is below 273 384 K. This can be also proved by the rapidly increased proportion of the second stage with 385

a value of 39.62% at 263 K. Since the methanol droplets cannot completely evaporate
at 253 and 243 K owing to the freezing of absorbed water, their lifetimes and the time
of the first and second stages are missing.



389

Fig. 7. Variations of the ideal and actual droplet normalized lifetimes (τ_i and τ_a), times
of the first and second stages (τ₁ and τ₂), proportion of the first and second stages (P₁
and P₂), and the increased percent of lifetime (P_τ) with ambient temperature.

393

394 3.2 Effect of initial droplet diameter on methanol evaporation

With the decrease of ambient temperature, the size of spray droplets in methanol engines become larger [35], which affects the evaporation of methanol droplets and further the cold start performance. Hence, the effect of initial droplet diameter on the evaporation characteristics of methanol droplets is discussed in this section. **Fig. 8** shows temporal evolution of the normalized squared diameters of methanol droplets with different initial diameters at temperatures of 243-303 K. It is noticed that the pure methanol evaporation of the first stage of methanol droplets has weak dependence on

initial droplet diameter. According to the conclusion of Verwey and Birouk [36], the 402 droplet evaporation in quiescent environment is related to molecular diffusion and 403 404 natural convection. The former is determined by fuel volatility and mainly affected by vapor concentration gradient on the droplet surface, while the latter is proportional to 405 the square of pressure and the cube of droplet diameter. For high volatile droplets, such 406 as methanol, the relative importance of the molecular diffusion appears to govern the 407 entire evaporation process at low temperatures and atmospheric pressure as compared 408 to natural convection, leading to the little effect of initial droplet diameter on pure 409 410 methanol evaporation. Similarly, Verwey and Birouk [37] also found that the change of the evaporation rate of n-heptane with initial droplet size in quiescent environment at 411 room temperature and atmospheric pressure is insignificant due to its high volatility. 412 413 However, the effect of initial droplet diameter on the methanol-water mixture evaporation of the second stage is more obvious as compared to the first stage. 414



415

Fig. 8. Temporal evolution of the normalized squared diameters $(d/d_0)^2$ of methanol 416 droplets with different initial diameters at temperature of 243-303 K.

Fig. 9 demonstrates the average evaporation rates of the first and second stages 418 and methanol droplets with various initial diameters at 243-303 K, K₁, K₂ and K_a. As 419 420 demonstrated in Fig. 9, K₁ is almost a constant and do not change with the initial droplet diameter, while K₂ shows relatively large fluctuation, especially at 293 and 303 K. And 421 K_a also shows a different trend, which decreases with the increase of the initial droplet 422 diameter. This can be explained by the fact that the larger the methanol droplet, the 423 more water vapor it absorbs, and the longer time of the second stage is, which can be 424 validated by the increased proportion of the second stage in Fig. 10(a). Then the lifetime 425 of methanol droplets is prolonged and K_a is reduced. 426



427

428 Fig. 9. Variation of the average evaporation rates of the first and second stages (K₁



temperatures of 243-303 K.

Fig. 10(a) shows the evolution of ideal and actual lifetimes with the square of initial droplet diameter at 243-303 K. As presented in Fig. 10(a), the ideal lifetime of methanol droplets without the disturbance of water vapor increases linearly with the

increase of the square of droplet diameter. And the relation between droplet lifetime t 434 and square of droplet diameter d_0^2 is obtained: $t = A * d_0^2$. According to the classical 435 d^2 law of single component evaporation: $d^2 = d_0^2 - K * t$, where K is evaporation rate 436 constant, A is 1/K. Notably, the same trend occurs in the actual lifetime. This suggests 437 that the actual two-stage and binary component evaporation process of methanol 438 droplets can be regarded as the evaporation of a pseudo single component, whose 439 evaporation rate constant can be acquired by the reciprocal of the linear fitting slope of 440 actual lifetime curves, that is, $K_p = 1/A$, where K_p is the evaporation rate constant of 441 the pseudo single component. The variation of K_p with ambient temperature is shown 442 in Fig. 10(b). It can be found that K_p increases linearly rather than exponentially with 443 the increase of ambient temperature, meaning that the pseudo single component is 444 445 various at each ambient temperature. Base on the above analysis, the lifetime of methanol droplets will be n² times of the original if the initial droplet diameter increases 446 by n times. Due to the larger size of spray droplet during the cold start process of 447 methanol engines, the evaporation of methanol droplets is greatly prolonged, resulting 448 in less combustible mixture of methanol vapor and air in a short time, whose 449 combustion is not enough to start the engine. Therefore, in order to enhance the average 450 evaporation rate of methanol droplets and shorten the entire evaporation time of 451 methanol, the size of spray droplets in the cold start process of methanol engines should 452 be reduced by improving injection pressure or other methods. For example, when the 453 droplet diameter is reduced by half, the droplet lifetime can be reduced to 1/4 of the 454 original according to the above formula, which greatly promotes the complete 455



evaporation of injected methanol in a short time.

Fig. 10. (a) The ideal and actual droplet lifetimes (t_i and t_a) and the proportions of the first and second stages (P1 and P2) under different initial droplet diameter at 243-303 K. (b) The effect of ambient temperature on evaporation rate constant of the pseudo

single component (K_p).

466 **3.3 Effect of fuel temperature on methanol evaporation**

Fuel temperature plays an important role on its physical properties, such as density, 467 468 viscosity and surface tension [38], thus affecting the spray droplets [39]. Consequently, the effect of fuel temperature on the evaporation characteristics of methanol droplets at 469 273 K is investigated to reveal the underlying mechanism, which can provide guidance 470 for practical methanol engine application. Fig. 11 shows the temporal evolution of the 471 normalized squared diameters and temperatures of methanol droplets with various fuel 472 temperatures. As shown in Fig. 11, there is little difference in the evaporation process 473 474 of droplets when methanol temperature changes. However, the relatively large difference can be found for droplet temperature curves. When the droplet is not heated, 475 its temperature rises rapidly first, namely transient heating stage, and then rises slowly, 476 477 reaching quasi-steady state, which is called equilibrium evaporation stage [40]. This occurs again in the late stage of evaporation due to the existence the second stage of 478 methanol droplets. Adversely, the temperature of droplets with higher fuel temperature 479 experiences first a process of rapid decline and then rising again before equilibrium 480 evaporation. The reason may be that the hot droplets releases heat rapidly until it 481 reaches a relatively equilibrium state with the surrounding cold environment due to 482 their enormous temperature difference. Therefore, there is an additional transient 483 cooling stage in the evaporation process of methanol droplets when the fuel temperature 484 rises. And the higher fuel temperature, the longer transient cooling stage and the shorter 485 transient heating stage. The effect of fuel temperature on the first equilibrium 486 evaporation stage is negligible, but mainly reflects in the second equilibrium 487

evaporation stage. Due to the test methanol droplets is in a large space, the effect of heat transfer to the environment on the ambient temperature can be neglected, while the humidity around the droplets is changed. This means that the fuel temperature has little influence on pure methanol evaporation, but mainly affects the evaporation of methanol-water mixture.



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Fig. 11. Temporal evolution of the normalized squared diameters $(d/d_0)^2$ and temperatures (T_d) of methanol droplets with various fuel temperatures at ambient

Fig. 12 illustrates the effect of fuel temperature on the average evaporation rates. As presented in Fig. 12, although the fuel temperature of droplets has increased by 8.5 K, the average evaporation rate of the first stage, K_1 , is almost unchanged and the improved percent of K_1 , P_{K1} , is around 0 % due to considerable droplet temperature rise rate in the first equilibrium evaporation stage according to Fig. 11. On the contrary, the average evaporation rate and its improved percent of the second stage, K_2 and P_{K2} , show remarkable fluctuation when the fuel temperature rises. It is because that the water

absorption in the second stage is influenced by the changed humidity around the methanol droplets. Ultimately, the average evaporation rate of methanol droplets, K_a , gradually decreases with the increase of fuel temperature owing to the increased proportion of the second stage shown in **Fig. 13**.



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Fig. 12. Variation of the average evaporation rate of the first and second stages (K1
and K2) and methanol droplets (Ka) (a) and their improved percent (PK1, PK2 and PKa)

514	(b) with increased fuel temperature $\Delta T'$. $K^{\Delta T'}$ represents the K after the fuel
515	temperature increases by ΔT ', K ⁰ represents the K under initial fuel temperature.
516	Fig. 13 shows the variation of ideal and actual normalized lifetimes and their
517	increased percent with the increased fuel temperature ΔT '. It is found that the trend of
518	ideal normalized lifetime is same as that of K1, because the former is determined by the
519	latter. And the increased percent of ideal normalized lifetime, $P_{\tau i}$, is also around 0 %.
520	However, the actual normalized lifetime gradually rises with the increase of ΔT ', which
521	has a maximum improvement of 14.5 %. It is mainly due to the extension of the second
522	stage, as shown in Fig. 13. According to the above discussion, improving fuel
523	temperature cannot promote droplet evaporation in terms of its effect on evaporation
524	rate and lifetime in this study. However, the droplet evaporation rate may be enhanced
525	if the heat exchange between the fuel and the environment can improve the ambient
526	temperature when the fuel temperature is high enough or the volume of environment is
527	small. More importantly, the viscosity and surface tension of methanol can be reduced
528	when the fuel temperature rises, then the size of spray droplets is reduced in methanol
529	engines [41], which is significant for shortening droplet evaporation time, as discussed
530	in section 3.2.



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Fig. 13. Variation of the ideal and actual normalized droplet lifetimes (τ_i and τ_a) and their increased percent ($P_{\tau i}$ and $P_{\tau a}$) and the proportions of the first and second stages (P_1 and P_2) with the increased fuel temperature $\Delta T'$. $\tau^{\Delta T'}$ is the τ after the fuel temperature increases by $\Delta T'$, τ^0 is the τ under initial fuel temperature.

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537 **3.4 Effect of intake air flow velocity on methanol evaporation**

It is commonly accepted that fuel droplet evaporation can be enhanced by flow. 538 On the one hand, flow can take away the fuel vapor on the droplet surface, which 539 promotes the diffusion of methanol molecules into the environment [42]. Furthermore, 540 the heat transfer between the droplet and the environment is also strengthened [37]. 541 Thus, the effect of intake air flow velocity on the improvement of methanol droplets 542 evaporation at low temperatures is quantitively evaluated at ambient temperature of 273 543 K. Fig. 14 shows temporal evolution of the normalized squared diameters and 544 temperatures of methanol droplets at various intake air flow velocities. It is clear that 545 the two-part feature of methanol droplets evaporation still exists, meaning that the d^2 546

law is not influenced by turbulence, which is consistent with the conclusion of Birouk
and Fabbro [43]. And the evaporation is accelerated as expected. This promotion effect
is more noticeable at low intake air flow velocity, but become weaker at higher velocity.
It can be also seen from the time history of the droplet temperature that temperature rise
rate of methanol droplets gradually increases with the increase of intake air flow
velocity due to the enhanced heat transfer by flow.



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Fig. 14. Temporal evolution of the normalized squared diameters $(d/d_0)^2$ and temperatures (T_d) of methanol droplets at various intake air flow velocities and ambient temperature of 273 K.

Fig. 15 illustrates that average evaporation rates of the first and second stages and methanol droplets, K_1 , K_2 and K_a , both are enhanced with the increase of intake air flow velocity. For example, K_1 is increased by 182 %, K_2 is increased by 145 % and K_a is increased by 163 % at intake air flow velocity of 2.92 m/s as compared to stationary methanol droplets, respectively. However, this enhancement is gradually weakened, because there are limits on the ability of vapor molecules to follow the fluid and the





Fig. 15. Variation of the average evaporation rates of the first and second stages (K_1 and K_2) and methanol droplets (K_a) and their improved percent (P_{K1} , P_{K2} and P_{Ka}) and

579 improvement efficiency (η_{K1} , η_{K2} and η_{Ka}) with intake air flow velocity V. K^V is the K 580 at intake air flow velocity of V, K⁰ is the K in the quiescent state.

According to the Fig. 16, the proportion of the first stage, P_1 , decreases and the 581 proportion of the second stage, P₂, increases as compared to the droplets in the quiescent 582 environment. It indicates that the flow also affects the water absorption of methanol 583 droplets while enhancing the evaporation rate. Moreover, the effect of intake air flow 584 on the lifetime of methanol droplets is shown in Fig. 16. The ideal and actual 585 normalized lifetimes of droplets both decrease exponentially with intake air flow 586 velocity. But their decreased percent, $P_{\tau i}$ and $P_{\tau a}$, are gradually leveling off with the 587 increase of intake air velocity, meaning that degree of the decrease of droplet lifetime 588 gradually decreases, e.g., the ideal and actual normalized lifetimes decrease by 50.44 % 589 590 and 48.07 %, respectively, when the intake air flow velocity increases from 0 m/s to 1.17 m/s, while only 21.56 % and 21.99 %, respectively, when it increases from 1.17 591 m/s to 2.34 m/s. Based on the above analysis, it implies that the intake air velocity 592 should not seemingly continue to increase when it reaches to a certain value. 593 Considering that there is already flow in the cylinder caused by inlet stroke of methanol 594 engines, the improvement of droplet evaporation rate caused by further increasing 595 intake air flow velocity can be limited based on the above analysis. However, from 596 another point of view, the flow is beneficial to spray breakup [45], thereby reducing the 597 droplet size, which can reduce the entire evaporation time of methanol. Besides, the 598 spatial distribution of methanol droplets can be more uniform under the action of flow. 599 This avoids the extreme descent of temperature in the local area caused by the 600

601 concentrated evaporation of methanol to absorb a large amount of heat, then the 602 reduction of evaporation rate of methanol droplets due to the descent of surrounding 603 temperature can be alleviated. Therefore, high intensity turbulence is useful for 604 improving the evaporation of methanol in the cold start process of methanol engines.



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Fig. 16. Variation of the ideal and actual normalized droplet lifetimes (τ_i and τ_a), their decreased percent ($P_{\tau i}$ and $P_{\tau a}$) and the proportions of the first and second stages (P_1

and P₂) with intake air flow velocity V.

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610 **4.** Conclusions

This study investigated the cold start issues of methanol engines and their improvement methods from the perspective of droplet evaporation. A wide range of fuel temperatures of 243-303 K was investigated. In particular, the low temperatures below 293 K were rarely studied in existing studies, which are of great importance for understanding the cold start issues of methanol engines and proposing effective improvement measures. The key conclusions are as follows:

(1) The evaporation process of methanol droplets with hygroscopicity consists of 617 two stages. The first stage is the evaporation of pure methanol and the second stage is 618 619 the water-dominated evaporation of methanol-water mixture, which shows noticeable difference in their evaporation rates. The droplets are evaporated by over 85% in the 620 first stage and freeze in the second stage at temperatures below 263 K. The proportion 621 of the second stage and the normalized droplet lifetime increase greatly when the 622 ambient temperature is below 273 K. The empirical formulas describing the 623 relationship between evaporation rate and ambient temperature for single-component 624 625 and binary-component droplets are obtained. The average evaporation rates of methanol droplets are enhanced by increasing ambient temperature, but this promotion effect 626 becomes weaker at higher initial ambient temperature. Increasing the ambient 627 628 temperature from 243-283 to 293 K has the largest improvement efficiency of evaporation rate. 629

(2) The initial droplet diameter has little effect on the pure methanol evaporation, 630 but mainly influences the evaporation of methanol-water mixture. The increase of 631 absorbed water in methanol droplets leads to the decrease of their evaporation rates 632 with initial droplet diameter. The proportion of the second stage increases with the 633 increase of initial diameter and the droplet lifetime increases linearly with the square of 634 initial diameter. The two-stage and binary-component evaporation process of methanol 635 droplets in humid environment can be regarded as the evaporation of a pseudo single 636 component and the same is true for other binary mixtures. The size of spray droplets 637 should be reduced by improving injection pressure or other methods to shorten the 638

entire evaporation time of methanol, so that more combustible fuel/air mixture can beproduced in a short time for cold start of methanol engines.

(3) The fuel temperature has little effect on the pure methanol evaporation, but 641 mainly influences the evaporation of methanol-water mixture. There is an additional 642 transient cooling stage before the evaporation of methanol droplets when the fuel 643 temperature rises. With the increase of fuel temperature, the average evaporation rate 644 of methanol droplets in humid environment decreases, while both the proportion of the 645 second stage and the normalized droplet lifetime increase. A higher fuel temperature 646 647 promotes methanol evaporation mainly through the reduced droplet size in real methanol engines when the increase of fuel temperature is insufficient. 648

(4) The intake air flow enhances the evaporation rate, but does not affect the two-649 650 stage feature of methanol droplet evaporation. This promotion effect is more noticeable at low intake air flow velocities and becomes weaker at higher velocity, especially when 651 intake air flow velocity exceeds 3 m/s. With the increase of intake air flow velocity, the 652 653 normalized droplet lifetime decreases exponentially, while the proportion of the second stage increases. High intensity turbulence is needed to reduce the size of spray droplets 654 and make the descent of overall ambient temperature in the cylinder relatively small, 655 thus improving cold start performance of methanol engines. 656

To sum up, multiple methods should be combined to improve the cold start performance of methanol engines efficiently. For example, to avoid a significant decrease in droplet evaporation rate caused by the massive heat absorption, the environment in the cylinder should be heated and the intake air flow velocity should be 661 increased. To shorten the evaporation time of methanol, the injection pressure and fuel

temperature can be also increased to reduce the size of spray droplets. In the future

663 work, molecular dynamics simulation method can be applied to investigate the

behaviors of water vapor in the process of methanol evaporation and further understand

the interaction between methanol and water.

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667 Acknowledgements

The assistance from the Geely Royal Engine Components Co., Ltd is greatly

669 appreciated.

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