

2,5-Dimethylfuran for Internal Combustion Engines: Potential and Development

Danh Chan Nguyen, and Van Huong Dong

Abstract—Transportation is facing two major challenges: renewable energy supplies and clean transport. On the other hand, the pressure from environmental pollution has led regulators around the world to come up with laws that have forced the auto and petrochemical industries to develop new technologies that reduce emissions and improve the quality of life. fuel economy. 2,5-Dimethylfuran (DMF), which is considered to be a new generation of promising alternative fuels, has the potential to reduce global fossil fuel shortage and air pollution problems. This article introduces the technical specifications and emissions of 2,5-Dimethylfuran (DMF), DMF production and application potential, developing DMF as alternative fuel sources in the World and in Vietnam

Index Terms—2,5-Dimethylfuran; Potential; Alternative Fuel.

I. INTRODUCTION

Energy shortages and environmental pollution are two major issues that have often been discussed at international scientific forums and seminars in recent years [1],[2]. Known as the "blood of the industries," petrol is viewed as a pillar of human civilization, however, the demand for petrol is so great and the emissions of it are toxic [3],[4]. This fossil fuel has reduced the overall development of the transport industry. A series of new technology trials to reduce emissions and increase the efficiency of automotive engine combustion are being proposed by researchers worldwide [5],[6]. In addition, alternative uses of alternative fuels are also an effective way to reduce environmental pollution and address energy shortages [7],[8].

New carbon-based fuels must meet the three minimum requirements. First, the carbon must be taken from the atmosphere and the fuel is derived from the photosynthetic product. Second, the fuel must be suitable for efficient combustion in internal combustion engines without altering the structure too many engines. Third, fuel must be efficiently produced and thrifty. Over the last 40 years, with the development of alternative fuels, bioethanol and bio-butanol have been extensively studied. Although ethanol, a renewable fuel used in internal combustion engines, is currently being produced in large quantities, ethanol fuel is facing many constraints such as low energy density, water absorption from atmosphere and high volatility [9],[10]. For every 10g of ethanol produced, 9.6 g of biomass is released as carbon dioxide and released into the environment during fermentation [11],[12]. The stability of the mixture between ethanol and diesel is simply not possible, but with the aid of

additives, this process can be accomplished. However, ethanol has not been used in diesel engines on a larger scale. For these reasons, ethanol is considered the first biofuel [13],[14].

Researchers have been trying to find a new generation of renewable fuels in the past 10 years [15]. Butanol fermented from sugar, starch, or lignin converted from plants has attracted much attention. Butanol can be used as fuel in internal combustion engines due to its properties such as: (1) butanol has a longer carbon chain than methanol or ethanol, with about 21.6% of it containing oxygen. The heat capacity of butanol is much lower than that of methanol and ethanol. (2) If no additive is used, butanol can be mixed with diesel or petrol in an appropriate proportion and can be used directly on gasoline or diesel engines, and phase separation will not occur. (3) The octane value of butanol is close to gasoline, and the calorific value of butanol is only about 9% lower than gasoline and diesel [16]. The results of many studies have shown the superiority of butanol as a fuel for internal combustion engines [17], but when used on a large scale, it is unsuitable for the cost of production output too high.

It has recently been discovered that DMF can be considered as a potential alternative fuel for internal combustion engines, as DMF has the potential for mass production. Joseph B. Binder and Ronald T. Raines [18] presented a method of converting lignocellulosic biomass, a rich material, into DMF. Summarized the methods of DMF production, and concluded that recent advances have improved the efficiency of DMF conversion from biomass. In fact, DMF has long been used as a solvent in the perfume and pharmaceutical industries. In addition, DMF has the appropriate physical and chemical properties to become an optimal alternative fuel. The physical and chemical properties of DMF, ethanol, n-butanol, gasoline and diesel are listed in Table I. These properties have shown that DMF has overcome the limitations of ethanol and n-butanol.

In Table I, it can be concluded that DMF has a higher calorific value than ethanol and n-butanol. The higher boiling point of DMF than ethanol can help to minimize the phenomenon of vapor barrier in the feeder. On the other hand, the lower boiling point of DMF over n-butanol would benefit from cold startup at low ambient temperatures. Compared to ethanol and n-butanol, less DMF absorbs water from the air resulting in less deterioration in storage. DMF and gasoline have similar viscosity, which facilitates the setting of DMF injection pressure in the fuel system, and protects the movement of the engine fuel system components. The research octane (RON) of DMF is about 119 higher than gasoline.

Compared to ethanol and n-butanol, DMF has many

Published on December 25, 2018.

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advantages. However, the direct use of DMF in the engine still has some potential problems:

TABLE I: COMPARISON OF PHYSICAL AND CHEMICAL PROPERTIES OF DMF, ALCOHOL AND FOSSIL FUELS [15]-[20]

Properties	DMF	Ethanol	1-Butanol	Gasoline	Diesel
Formula	C ₆ H ₈ O	C ₂ H ₆ O	C ₄ H ₁₀ O		
Molecular mass (g/mol)	96.13	46.07	74.12	100-105	
Oxygen content (%)	16.67	34.78	21.6	0	-
Hydrogen content (%)	8.32	13.02	13.49		
Carbon content (%)	75.01	52.2	64.91		
Air/fuel	10.72	8.95	11.2	14.7	14.3
Density (kg/m ³ , 20 °C)	889.7	790.9	810	744.6	820
Laten heat (kJ/kg tır 25 °C)	389.1	919.6	707.9	351	270-301
Lower Heating value (MJ/kg)	33.7	26.9	33.2	42.9	42.5
Boiling point (°C)	93	77.3	117.25	27-225	180-370
Water dissolute (wt% , 20 °C)	0.26		7.7		
Octane number	119	110	98	90-100	-
Cetane number	9	8	25	10-15	40-45
Surface tension (mN/m)	25.9	22.3	24.6	20.0	
Viscosity (cSt, 20 °C)	0.57	1.5	3.6	0.37-0.44	
Self-ignition temperature	286	434	385	420	246

(1) DMF has higher energy density than ethanol and butanol, but lower than gasoline and diesel. To achieve the same engine efficiency, the use of DMF as a fuel for gasoline or diesel engines requires increasing fuel consumption, which results in the fuel supply system having to operate at a higher intensity.

(2) The molecular structure of DMF, consisting of C-C double bond and enol ring, is likely to generate large amounts of soot during combustion.

(3) The toxicity and impact of DMF on the environment and its combustion products should be further studied.

II. RECENT ADVANCES IN THE PREPARATION OF DMF

The global energy shortage and environmental pollution have put pressure on scientists to develop a new generation of technologies that can synthesize cheap biofuels from renewable biomass [19]. Second generation biofuels must be manufactured using sustainable chemical products and produced through modern and developed chemical technologies such as pyrolysis, Fischer Tropsch synthesis or process catalysis. All of them can create complex molecules or convert materials into sustainable biofuels

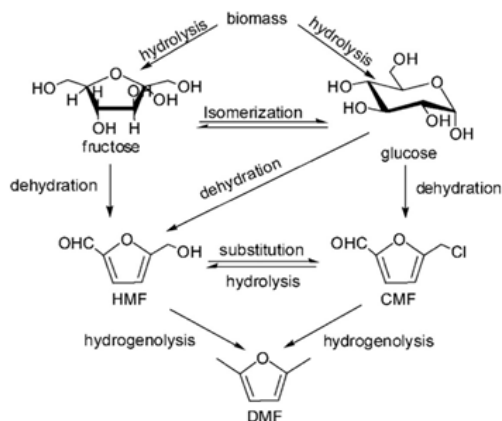


Fig. 1. Diagram of steps to prepare DMF from biomass

More than 75% of the biomass, like corn, trees and grass, contains carbohydrates (such as starch and cellulose). Carbohydrates usually exist in the form of polymer chains consisting of thousands of units (glucose or fructose). And each unit has six carbon atoms and one oxygen atom, which means that biomass molecules contain more than 100 carbon atoms. In contrast, fuel for internal combustion engines typically has molecules of 5 to 15 carbon atoms [20]-[23]. Therefore, the main challenges in converting biomass to biofuels are to break down (hydrolyze) the molecules and remove the oxygen atoms. So far, the main method of using biomass to prepare DMF consists of two ways, as shown in Fig. 1. The first method is to pre-biomass and reduce it to glucose or fructose before triple Oxygen is removed from glucose or fructose by separating water selectively to form 5-hydroxymethylfurfural (HMF). Finally, HMF can be converted to DMF by hydrogenolysis. The second route is to biomass, including cellulose or glucose, to 5-chloromethyl furfural (CMF) by first splitting water. Then transfer CMF to DMF by hydrogenolysis. By providing sufficient biomass and low cost of raw materials, DMF's modulation and yield will directly determine whether it can be used as a substitute for a wide-range car engine. Therefore, in recent years, many researchers have focused on biomass such as glucose, fructose and starch as the raw material for conversion to HMF or CMF using two corresponding paths and improving transfer efficiency, modified by different catalysts and solutions.

A. Convert biomass to DMF with HMF as an intermediate

As an important intermediary in the conversion of biomass to DMF, HMF has been extensively studied on conversion efficiency. Recent achievements of extensive studies are listed in Table II.

TABLE II: HMF MODULATION SOLUTIONS

Solution	Material	Catalysis	T(°C)	Time	Efficiency (%)	Ref
DMA-10wt% NaBR	Fructose	H ₂ SO ₄ , 6mol%	100	2h	93	25
[EMIM][Cl]	Fructose	CrCl ₂ , 6mol %	100	3h	68	26
DMSO	Fructose	FePW ₁₂ O ₄₀ , 0,02 g	120	2h	97	27
DMSO	Fructose	H-BEA zeolit, 0.02 g	120	2h	97	29
DMSO	Fructose	Ambelyst-15-P, 0.02g	120	2h	100	28
[BMIM][Cl]	Glucose and Fructose	NHC-CrCl ₂ , 9mol %	100	6h	81	29
[BMIM][BF ₄]	Glucose	SnCl ₄ , 10 mol %	100	3h	60	30
[BMIM][Cl]	Glucose and Di-polysaccharides	CrCl ₃ , 10 mol %	100	1h	67	31
			120	5min	67	32
CPL-LiCl	Glucose	CrCl ₂ , 6mol %	100	3h	58.7	33
[BMIM][Cl]	Glucose and cellulose	CrCl ₃ , 9 mol %	-	1min	91	33
[BMIM][Cl]	Cellulose	CuCl ₂ và CrCl ₂ , 6mol %	80-120	1h	55.4	34
[OMIM][Cl]-HCL	Starch	CrCl ₂	120	2h	73	35

In 2007, Roman et al. [24] first proposed a promising chemical reaction to produce DMF from fructose. The diagram is shown in Fig. 2 [25]. HCl is used as a catalyst, and fructose is converted to HMF with HCl as a catalyst in 35% NaCl solution. HMF is then extracted from the 1-butanol solution. Finally, HMF is converted to DMF by a copper-ruthenium catalyst (CuRu/C). This method has a high conversion efficiency (62-70%) and no by-product. In addition, the catalyst can be re-used. But butanol, used as an extractant, still needs to be studied for large-scale production. Zhao [26] tested the effectiveness of a series of catalysts to convert sugar into 5-hydroxymethylfurfural (HMF) and found a low-energy method (Fig. 3). Among the various metal halides in 1-alkyl-3-methylimidazolium chloride, chromium (II) chloride is the only high performance, with a yield of nearly 70% in conversion from glucose to HMF. The entire 3-hour process can lead to low productivity. Phosphor-molybdic acid, a new catalyst, can improve conversion efficiency by up to 99% from glucose conversion to HMF even higher by 99% in ionic liquids with a two-step approach. After this step, 44-47% of HMF is converted to DMF by hydrogenation with the presence of Pd/C as a catalyst and acetonitrile as an additive. It should be noted that in the hydrogenation reaction, HMF was first transformed into MF before migration to MFA and finally DFF through hydrogenation. Shimizu [27] changed the pressure inside the reactor and presented two simple ways to increase HMF productivity through the separation of fructose water in the presence of various solid acid catalysts. One method is to remove the water from the reaction mixture with light release at 0.97-105 Pa and increase the yield of HMF with heteropoly acid, zeolite and acid as a catalyst. Another method is to reduce the molecular size of the resin (Amberlyst-15).

The method of converting sugars selectively to HMF in the NHC-Cr / ionic system, as shown in Fig. 3 [28]. The conversion efficiency of fructose to HMF can be at least 96% and 81% for glucose. Lewis SnCl₄ acid system and [EMim] BF₄ system are the best catalysts for converting glucose into HMF [30],[31],[35]. Even if the glucose concentration is higher than 26%, this method can still achieve high efficiency. Appropriate results are obtained

with the use of fructose, sucrose, cellobiose, inulin and starch. This highly efficient, low cost, non-harmful and reusable catalytic system has high application potential.

To simplify the transition, a one-step method for converting fructose to DMF with the presence of HCOOH, H₂SO₄, Pd/C and THF in solution [36]. In a chain of transformations, fructose was first converted to HMF, followed by FMF, HMMF, FMMF and finally DMF. DMF is extracted with diethyl ether with an approximate yield of approximately 51%. In terms of the long reaction time of the above methods, Qi et al. Proposed a method for efficiently converting glucose into 5-HMF in the presence of CrCl₃ as a catalyst using 1-butyl -3-methyl imidazolium chloride as a solvent and a microwave oven. 5-HMF 71% yield achieved in 30 seconds when converting 96% glucose in a microwave oven to 140 ° C. Fructose, sucrose, cellobiose and cellulose were tested, and 5-HMF yield of 54% was obtained during a 10-minute reaction when cellulose was used as a material at 150 ° C. Chen et al. [29] reported that caprolactam (CPL) containing lithium chloride (LiCl) was a preferred solvent that allowed the synthesis of the base chemicals to produce HMF from pure glucose. Meth halide in CPL-LiCl was used as a catalyst, in which CrCl, CrCl 2, SnCl 4 and SnCl 2 were found to be significantly effective, resulting in a yield of 55-67% from glucose to HMF.

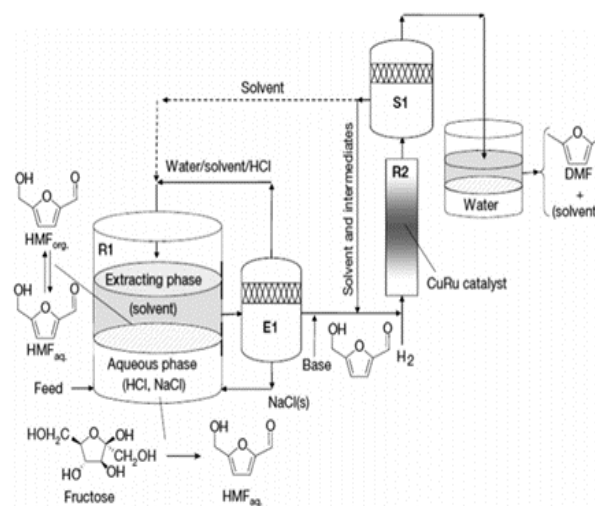


Fig. 2. Schematic of fructose conversion to DMF

In 2009, a number of studies have been conducted to convert lignocellulosic (corn, cellulose), glucose and fructose into HMF with different catalysts in different ionic liquids. The efficiency of each type in different circumstances can be attained about 48%, 54%, 81% and 92% (shown in Fig. 4). Chlorine ions were then removed from the crude HMF raw material by ion-exclusion chromatography. Finally, low purity HMF is converted to DMF by hydrogenation in 1: butanol with Cu: Ru / C as the catalyst. Total output is 49%. In addition, untreated straw as a raw material was used by the authors to convert to DMF in one step, yielding a yield of 9%. Despite the low conversion efficiency, the research has expanded the raw material used to produce DMF from straw.

To improve the conversion efficiency from cellulose to DMF, Li et al. [32] proposed a way to convert cellulose to HMF without separating sugar. This method provides a whole new way to transform the abundant and cheap

cellulosic biomass through a fermentation process into biofuels and bio-based products. HMF was obtained at 91% and 61% yields were isolated in ionic solution under microwave radiation in the presence of some CrCl₃ catalysts. To further simplify the process, a single-step catalytic process in which cellulose is used is rapidly fragmented and results in glucose being converted to HMF in mild conditions [33]. A pair of metal chlorides (CuCl₂ and CrCl₂) dissolved in 1-ethyl-3-methylimidazolium Cl solution (80-120 ° C). Purity of 96% between recoverable products (with an HMF yield of 51-59%). In relatively light conditions (1400C, 1 atm) and when no additional acids were used as catalysts in biomass conversion, cellulose dissolved in some ionic liquids (ILs) was converted into Water-reducible sugars are high in total, reducing sugars (up to 97%), or have been converted directly into highly biochemical HMF substrates (up to 89%) when CrCl₂ is added [35]-[37].

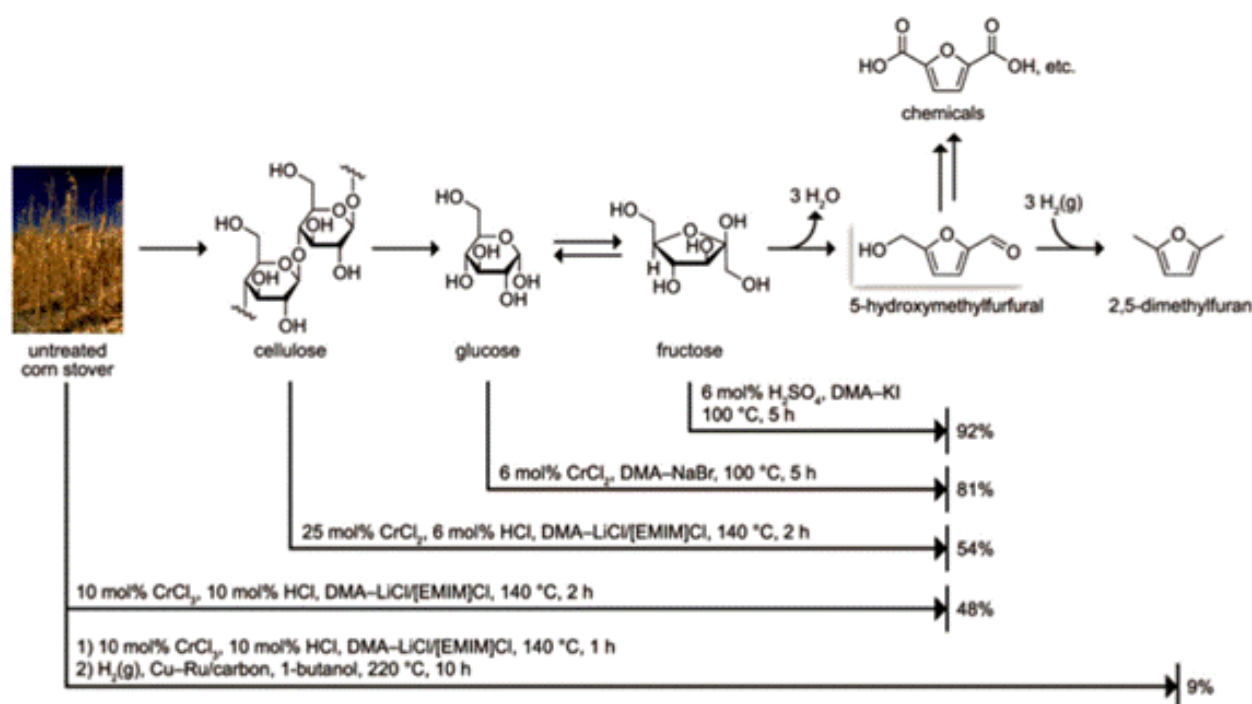


Fig. 3. Comparison of DMF performance through different catalysts

B. Conversion starch to DMF

Starch, as an abundant resource, can be used in the production of long-lasting, diverse chemical intermediates such as 5-hydroxymethylfurfural (HMF). Simple process for the production of HMF from starch in ionic solutions using 1-octyl-3-methylimidazolium chloride (OMIM) as a solution and CrCl₂ as a catalyst [34]. The addition of HCl and CrCl₂ significantly affected the performance of HMF. Eight kinds of starch sources (corn, wheat, rice, potato, sweet potato, cassava, acor and kudzu starch) were tested for HMF synthesis. When dissolving cassava starch with 0.5M HCl, the highest yield of HMF (73.0 ± 3.8% wt).

Effect of various parameters on conversion of carbohydrate and biomass to biofuel with 5-bromomethyl furfural (BMF) neutralization value [38]. The optimum conditions for avoiding the use of lithium salts, making the method economically more environmentally friendly than

other methods. The different types of wood that have the potential to be BMF's raw materials and furfural production have also been estimated. In addition, a simple and effective trick to convert 5-hydroxymethylfurfural (HMF) into BMF or 5-chloromethylfurfural (CMF) has been reported by the authors.

C. Transforming cellulose and starch into DMF using CMF as an intermediate

In 2008, Mascal [39] dissolved cellulose and glucose in concentrated HCl prior to the use of 1,2- 2-ethylchloride to extract CMF. The CMF is then converted to DMF with Pd / C as the catalyst, as shown in Fig. 4. The author recently presented a new but feasible method to become a ground biomass, 5-chloromethyl furfural (CMF) [40]. They say that not only glucose, sucrose and cellulose but also rough biomass (such as cotton, wood, maize, straw and scrap) can be converted into CMFs with high productivity. The authors

predict that CMF may emerge as an intermediate in converting carbohydrate-containing materials into useful organic materials. Mascall et al showed not only a shift from CMF to biofuel 5-ethoxymethylfurfural (EMF) or ethyll- evulinate (EL) in ethanol, but also studied the conversion technology to convert CMF to HMF or LA.

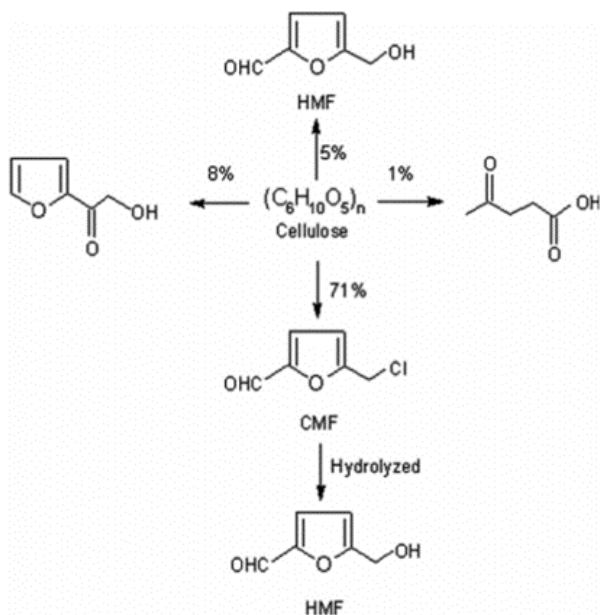


Fig. 4. Flowchart describes the process of converting cellulose to HMF

Compared with ethanol, DMF is a better renewable fuel to replace gasoline in the future. Chemical methods for biomass including glucose, fructose, starch, and cellulose are converted to DMF with high efficiency. However, the DMF synthesized from these methods remains one limited number [27]. Although the HMF yield may be higher than 90% at 1200C for 10 minutes, the preparation of solid acid catalysts is complex. And the method proposed by Gen Yong et al. [19] may convert fructose to HMF with an even higher yield of 95% and 80% with glucose, but requires the reaction mixture to be kept at 100 ° C in 6 hours. And HMF needs to be extracted three times. It is necessary to have a deeper study to find a method with reasonable energy input and significant output efficiency [41]. It should be noted that the use of non-food biomass for DMF production can avoid food and land issues. The progress of Mascall and his colleagues transforms cellulose into DMF. Regarding long-term reaction times and the use of neutralizing HCL, the development of a more efficient catalyst and new solvent is needed. In order to make mass production possible, efficient separation and filtration technology is essential.

III. CONCLUSION

DMF has the potential to become an alternative biofuel with many advantages, such as high energy density, high oxygen content, high octane content, boiling point, insolubility, and the ability to mix with gasoline and diesel. . In addition, DMF has the advantage of being able to expand the material resources in the production process. The methods of producing DMF are usually one-step or two-step methods.

When used with DMF in place of gasoline, the burning time may be shorter due to its higher octane rating. However, under high loads, startup time too early may result in the engine being knocked. By adjusting the MBT / KL-MBT ignition timing, DMF's fuel consumption is greater than that of gasoline due to lower energy density than gasoline. Higher temperatures lead to higher NOx emissions during combustion of DMF than gasoline. DMF has slightly lower HC emissions but emits CO much higher than gasoline. When using mixtures of DMF and diesel, low oxygen content and boiling point can help reduce particle emissions while reducing low temperature combustion based on the EGR ratio at the same time. Although DMF is said to be the alternative fuel, there are still a number of issues that need to be addressed: c. The compatibility of DMF fuel systems with the abnormal emission mechanisms of DMF.

Although there have been many studies and applications of DMF in internal combustion engines, their adaptability and efficiency compared with traditional fuels still require extensive research and experimentation to allow DMF to replace fuel transmission in the future.

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