Review on the physicochemical, spray characteristics and chemical reaction mechanism of F-T diesel-methanol-diesel-biodiesel blended alternative fuels

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Abstract. Alternative fuel is an ideal solution to solve the problem of petroleum resources shortage and diesel engines’ pollution. After analyzing the China’s energy structure and the merit and demerit of liquid alternative fuel, we finally find that the F-T diesel-methanol-diesel-biodiesel blended fuel is the best choice. The paper reviewed the study of the blended fuel home and abroad from the three aspects which contains the study of the miscibility and physicochemical properties, the study of the spray characteristics and the study of the chemical reaction mechanism. Finally, the insufficiency and the development direction of F-T diesel-methanol-diesel-biodiesel blended fuels were proposed in the paper.

1. Introduction
Gasoline engines are widely used in the world due to their advantages such as high energy density[1] and low fuel consumption[2], which have become major delivery vehicles in the fields of transportation and workers and peasants' machinery. However, rapid growth in the number of diesel has increased the consumption of oil resources, which makes China's oil import dependence from 60.86% in 2015 increase to 67.4% in 2017[3]. In addition, due to the effects of fuel and self-combustion, a large amount of particulates and NOx are generated, which seriously pollute the environment and harm human health. Furthermore, with the imminent implementation of the China 6 regulation [4], more stringent requirements have been placed on diesel engines. Alternative fuels are considered to be the most ideal solution to solve the shortage of oil resources and improve diesel emissions.

Up to now, the commonly used alternative fuels mainly include hydrocarbon and oxygenated fuel. Hydrocarbon-based alternative fuels mainly consist of F-T diesel fuel and aviation kerosene. These fuels contain less aromatics and sulfur, inhibit the formation of PAH and sulfate in soot, which can effectively reduce particulate matter without increasing NOx emissions. Oxygenated fuels mainly contain esters, alcohols, and ethers. This type of fuel reduces the amount of carbon-carbon and carbon-hydrogen bonds due to the increase of its own oxygen element, so that the amount of unsaturated molecular carbon at high temperature decreases, and the presence of oxygen can react to produce hydroxyl groups under certain conditions[5][6]. Liquid ethers are mostly added to other fuels as
additives, they are not used as fuel on diesel engines alone. Therefore, they will not be discussed in this paper. The advantages and disadvantages of commonly used alternative fuels are shown in Table 1.

| Fuel               | Advantage                                      | Disadvantage                                      |
|--------------------|-----------------------------------------------|---------------------------------------------------|
| F-T diesel         | Good ignition performance, low sulfur content, low aromatic hydrocarbons content, high heat efficiency, good miscibility with diesel | Poor lubricity, high cost, easy precipitation of paraffin hydrocarbons at low temperature |
| Aviation kerosene  | Good spray ability, low sulfur content, low aromatic hydrocarbons content, good miscibility with diesel | Poor lubricity, poor ignition performance          |
| Esters             | Good ignitability, good lubricity, oxygen content, good miscibility with different fuels | Poor spray ability, carbon deposition easily      |
| Alcohols           | Good spray ability, oxygen content, low price | Poor ignition, poor miscibility with diesel, low calorific value |

It can be seen that each kind of alternative fuel has its own advantages and disadvantages. The ideal alternative fuel for reducing the emissions of engine should have the characteristics of higher cetane number, good spray ability, oxygen content in the molecule. However, it’s difficult to satisfy for an alternative fuel alone, it can be achieved only by blended fuels. Therefore, the blended fuel is considered to be a development trend to improve the combustion emissions and heat efficiency of diesel engines [7].

According to the energy structure characteristics of “multi coal and less oil” in China, the coal chemical products F-T diesel and methanol blended fuels are considered as the additives to the diesel for improving the physicochemical properties of the fuel. By the end of October 2016, nine coal-to-oil projects had been put into operation in China, which contained ShenHua Ordos 1.08 million ton direct liquefied coal oil production project, ShenHua ordos 180,000 t coal oil production F-T diesel project, YiTai 160,000t F-T oil project, LuAn 160,000t F-T oil project, YuLin Yankuang 1.1 million ton F-T diesel project and so on, which can produce more than 7.15 million tons of F-T diesel per year, and the production is increasing. Therefore, the production of the F-T diesel isn’t the problem to limit the use of F-T diesel. Herein, this article focuses on the F-T diesel-methanol-diesel-biodiesel blended fuels, and summarizes the current research from the aspects of the physicochemical characteristics of the bended fuels, the study of the spray characteristics, the study of the chemical reaction mechanism, at the last the insufficient development and future research are suggested.

1 Study on the miscibility characteristics and physicochemical properties of F-T diesel-methanol-diesel-biodiesel blended fuel

1.1 Miscibility characteristics of F-T diesel-Methanol-Diesel-Biodiesel blended fuel
The process of dissolving two or more substances mutually is a physical or chemical process or both. F-T diesel and diesel are mainly composed of paraffins, aromatic hydrocarbons and naphthenic hydrocarbon. The molecules mainly contain C-C and C-H covalent bonds, and the positive and negative charge centers overlap, showing non-polar characteristics. However, the C-O bond and H-O bond contained in the methanol molecule are larger than the C-H bond dipole moment and exhibit strong polar characteristics. However, it is difficult for methanol to dissolve in the diesel or F-T diesel, and solvent additive is needed to achieve mutual solubility between methanol and diesel and F-T diesel. Biodiesel contains both hydroxyl groups and hydrocarbon groups which can combine the
methanol molecule and diesel, respectively. Therefore, a small amount of fatty acid methyl ester is needed to promote the fusion between methanol and F-T diesel oil and diesel oil.

The miscibility of the blended fuels was firstly studied in the Gibbs phase formula in 1876, which expressed in the number of phases balance factor, fraction, the degree of freedom and the external factors affecting the properties of the material (such as temperature, pressure, gravity field, electromagnetic field, surface energy, etc.) in the equilibrium coefficient. However, Gibbs theory is only used in the calculation of the balance system and lacks the related calculation of the miscible mechanism. Hidebrand JH et al.[8] proposed the concept of solubility parameter in the middle of the 20th century. It has been widely recognized that the solubility is related to the cohesive nature of the solvent to calculate the miscibility. After that, focus on the Hidebrand J H’s research, various countries started a series study. Hansen CM et al [9] proposed a shaped regarded as the liquid dispersion force, polar force and hydrogen bond force contribution and three molecules on the deficiency of Hidebrand JH’ theory that can not be applied to non-polar liquid mixtures, which extended the theory to the polar system. Bagley EB [10] et al. made a further step, extended the solubility parameters and applied it to the polar components and electronic parameters. On the basis of the thought of Bagley E B. Liu [11] established the calculation of solubility parameters for predicting the non-associative liquid by molecular composition and structure, and perfected the liquid mixing thermodynamic model.

Based on the theory of fuel miscibility, a series of related researches on the miscibility of blended fuels have been conducted at home and abroad. The three liquid phase diagram of Diesel / methanol / oleic acid at normal pressure was measured by Jiang[12], as shown in Figure 1, compared the inter solubility with non cosolvent, rapeseed oil and isobutanol as a cosolvent, such as point 1, 2, 3, they finally found that the isobutanol was the best solubilization. Wang [13] studied the effects of alcohols on the inter solubility of between the methanol and diesel fuel, obtained liquid phase diagrams with methanol, diesel and different solvents, as shown in Figure 2. From the figure, we can come to the conclusion that with the increase of the carbon number of the cosolvent, the solubilization effect increased firstly and then weakened, and the twelve alcohol had better dissolution effect than other cosolvents. Chen[14] focused on the intermixing characteristics of three solutions of ethanol / water / kerosene at different temperatures and atmospheric pressure, as shown in Figure 3. The degree of solution solubility is related to the purity and temperature of the substance. With the increase of temperature, the mutual solution region increases with the increase of temperature, and a MATLAB model has been established to verify the ideal effect of the mutual solution separation curve.

It can be seen that even though the solubility parameter between the hydrocarbon such as diesel, F-T diesel and methanol has great differences, the biodiesel can promote the solubility of the three fuels, and it’s a pretty suitable cosolvent despite the capacity is weaker than high alcohols. The temperature and water content is also impotent for the blended fuel.

![Figure 1. Diesel/methanol/oleic acid three phase diagram.](image-url)
Figure 2. Three phase diagram of diesel/ methanol with different cosolvents

Figure 3. kerosene/ ethanol/ water Detach Curves at different temperature.

1.2. Physicochemical properties of F-T diesel-Methanol-Diesel-Biodiesel blended fuel

The physicochemical properties of fuels largely determine the engine mixture formation, ignition combustion, and load regulation modes, which in turn affect the cycle thermal efficiency, the average excess air ratio in the cylinder, and the charge coefficient, and ultimately determine the power, economy, and emissions.

Recent years, abundant researches focused on the calculation of physicochemical properties of blended fuels. KoreaSeung Hyun Yoon[15] studied the physical and chemical properties of different proportions of biodiesel blended diesel oil at 0-200 °C, and obtained the relationship between density and temperature, finally carried out practical verification. Yasin [16] studied the physicochemical properties of a mixture of methanol and ethanol incorporation into biodiesel-diesel and found that with the addition of alcohol, the density and viscosity gradually decreased, and the flash point decreased first and then increased. The study by Magín Lapuerta [17] showed that with the increase of alcohols, the viscosity of blended fuel contain alcohols, biodiesel and diesel decreases nonlinearly, and the mixed viscosity of ethanol/biodiesel can be calculated with Grunberg-Nissan formula. Han [18] obtained similar physical and chemical characteristics of different ratios of methanol-diesel fuel as Yasin’s study. As the proportion of methanol blended increased, the cetane number, viscosity, and
surface tension of blended fuels gradually decreased, resistance and little change in pour point, low temperature distillation characteristics of the mixed fuel is strong.

The above studies reveal the influencing factors of the miscible mechanism and physicochemical characteristics of the blended fuels in the liquid phase system in different aspects. From the review we can come to the conclusion that the content of F-T diesel is better for decreasing the NOx, while methanol is better for decreasing the PM. The blended fuel has better characteristics of “trade-off” between the PM and NOx than with only F-T diesel. The density decreases linearly with the increase of temperature, while the viscosity has non-linear characteristics, it can be calculated with Grunberg-Nissan formula. However, these studies are based on the Gibbs liquid formula and solubility parameters under the premise of the standard atmospheric pressure. As we all know, the blended fuel from the fuel tank to the injection cylinder needs to experience different environmental pressure changes. With different pressures, the thermodynamic parameter changes, so does the solubility parameter. And with these changes, the error of the physicochemical properties may increase, with the same calculated coefficient. The influence of the atmospheric pressure on the solubility and the physical and chemical properties of the mixed fuel has not been involved at home and abroad.

2. Research on spray characteristics of F-T diesel-methanol-diesel-biodiesel blended fuel

Fuel atomization increases the specific surface area of fuel and accelerates the evaporation and vaporization of fuel, which is beneficial for the mixing of fuel and air. The quality of the atomization directly determines the ignition and combustion process in the cylinder, which is of vital importance to the diesel engine. The research on spray characteristics at home and abroad mainly focuses on two aspects: atomization mechanism and spray characteristics.

2.1. Research on atomization mechanism of F-T diesel-methanol-diesel-biodiesel blended fuel

When fuel is injected into the combustion chamber from the nozzle, the fuel starts to split and atomized, as shown in Figure 4. The spray field can be divided into liquid core area, billowing flow area, dense area, rarefied area and extremely thin area according to the distance fuel state. For the reason of the atomization mechanism, Dejuhasz [19] proposed in 1931 that the atomization process of the jet occurred inside the nozzle, and the turbulence of the liquid itself played an important role. Schweitzer believes that the radial velocity of liquid in the nozzle of turbulent pipe flow will cause disturbance immediately at the outlet of the nozzle, which could result in atomization. Bergwerk considers that the turbulent velocity in the nozzle is not enough to cause atomization. The atomization is attributed to the large amplitude pressure disturbance produced by the cavitation phenomenon in the
nozzle, thus forming the cavitation perturbation theory. Giffen noted that the pressure oscillation produced by the fuel supply system has a certain effect on the atomization. It is suggested that the widespread pressure oscillation in the injection system may play an important role in the atomization. Shkadov[20] studied the change of shear stress at the boundary of the interface, which confirmed the existence of unstable short wave length surface waves. Reitz and Bracco [21] have made a thorough analysis of the atomization mechanism proposed by the predecessors. The aerodynamic interference is the most promising, and it is amended and supplemented. The results show that the aerodynamic interaction between the high speed jet and the surrounding gas leads to the rapid and selective growth of the unstable wave of the liquid surface, which is the basic agent of atomization.

2.2. Effect of spray characteristics

The spray characteristic parameters mainly include spray penetration distance, spray cone angle and spray particle diameter, among which the spray particle size includes average particle diameter, soot mean particle diameter and particle size distribution. The environmental conditions and the fuel's own parameters determine the spray performance of diesel engines by affecting these spray characteristic parameters. Hiroyasu[22] proposed the theoretical calculating formulas of the spray penetration distance of injector, spray cone angle and Sauter mean diameter based on a large number of test results[23]. Siebers [24] studied the effects of nozzles, fuel types, and the thermodynamic environment of the cylinder on the diesel penetration distance and spray cone angle in a constant volume incendiary bomb. A penetration distance scale theory was established, which showed that penetration distance of diesel is influenced by the nozzle aperture, injection pressure, environmental density, ambient temperature and fuel temperature. The evaporation process of the oil beam is influenced by the mixing process of the oil beam and the ambient gas. The evaporation process of oil bundle is affected by the mixing process of oil bundle and environmental gas. According to Jiang[12], two relatively rotating vortex structures appeared on the two edges of the oil bundle after the start of fuel injection in the alcohol fuel, and the tight width of the oil bundle decreased under high back pressure. The spray cone angle of the alcoholic oil spray undergoes a process of increasing first and then decreasing during the spraying process, mainly because the air entrained vortex structure formed after the spraying dispersing the spray, the cone angle becomes large, and the spray is fully mixed with the ambient gas. After the stabilization period, the cone angle becomes smaller. And through the study to obtain alcohol due to its high volatility, increase the injection pressure can effectively promote the atomization and dispersion of oil.

In recent years, with the development of spray technology, the visualization of spray has become the main method for studying the spray characteristics, which mainly includes: schlieren method, sideview shadowgraph method, scattering method, chemiluminescence, laser-induced incandescence, laser induced fluorescence, plane laser induced fluorescence and so on. W.Schindler[25] et al. focused the research on the influence factors of spray penetration distance and spray cone angle by schlieren method, they finally found that the increase of injection pressure and decrease of nozzle diameter enlarger the Reynolds number which causes a larger turbulence downstream of the orifice. The high air density near the wall of the combustion chamber increases the resistance of the air to the droplets, which makes the spray velocity decrease and deflects along the spray shape along the axial direction. With the method of Mie scattering, C.Espey and Jedec et al. [26] studied the influence factors of the fuel liquid penetration. It was found that the initial liquid penetration length increased linearly with temperature and density until it reached the longest distance and then remained stable. As the temperature and density increase further, the liquid penetration distance, the ignition delay period and the premixed combustion stage gradually decrease. With the method of Mie scattering and schlieren, DL Siebers[27] measured the fuel penetration distance and spray angle, respectively, and studied the influence of injection hole diameter, ambient temperature and pressure, fuel volatility and fuel temperature on jetting. They finally found that the volatile components in the blended fuel were first atomized and vaporized, and the non-atomizable components determine the penetration length of the
fuel liquid. As the volatility of the fuel increases, the liquid penetration distance decreases, but when the ambient temperature in the cylinder is high, the effect is no longer significant.

From the above studies, it can be seen that the aerodynamic interference may be the best theory to explain the atomization mechanism, it can be used to analyze the mechanism of the blended fuel, and the Weber and Ohnesorge number play a key role in forming atomized mixture. Because of the density and kinematic viscosity, biodiesel in the blended fuel can extend the spray penetration distance, and the methanol can enlarge the spray cone angle and reduce soot mean particle diameter, it can also increase the air entrained vortex which is benefit for spraying of the blended fuel, for high boiling point and low density and kinematic viscosity.

Furthermore, diesel injection pressure, back pressure, orifice diameter, temperature, density, blended fuel, etc., will affect spray penetration, spray cone angle, and spray particle size, the spray characteristics should be studied in a comprehensive method. However, there are few papers about the coupling effect of blended fuel and environmental parameters on spray characteristics.

3. Research on chemical reaction mechanism of F-T diesel-methanol-diesel-biodiesel blended fuel

The process of burning coupled with a complex system of various physical and chemical processes such as flow, heat transfer, mass transfer, and chemical reaction. Chemical reaction is the core of the system and plays a pivotal role in the key burning phenomenon of fuel on fire, flame propagation, flammable limits, burning stability and pollutant emissions. Through the research of chemical reaction mechanism and set up precise model of combustion reaction power can help to get a clearer understanding of the combustion process and the mechanism of the emissions generation, strengthen the control of burning phenomenon, and provide us with the theoretical guidance of improving the combustion efficiency and the pollutants emissions’ control. The types of chemical reactions mainly include cracking, oxidation reactions, which involve the rates, elementary reactions, total package reactions, molecular collisions, chain reactions, and so on. These reaction processes are accompanied by the formation of a large number of intermediates, and involved the microscopic reactions such as quantum chemistry, electronic transition has unusually complicated research and only can carry out the research to establish a chemical reaction kinetic model by computer. The verification and enrichment of chemical reaction kinetic model is the premise to make sure of the model can apply to different temperature, pressure, equivalence ratio and so on.

The expansion and enrichment of the adaptability of the chemical reaction kinetic model is always the direction that people have been trying hard to study. The initial study of the chemical reaction mechanism should be traced back to the early 19th, the Swedish scientist Arrhenius summed up the empirical correlation of the effect of temperature on the reaction rate from the experiment. Semyonov proposed a chain reaction theory and explained the fuel combustion with elementary reaction layers, at the early 20th. When it came to 1970s, Shell research center launched a self-ignition model consisting of 8 steps on the basis of the degenerate branching reaction mechanism has been successfully applied to the combustion of internal combustion engines. People have been continuously enriching and improving the accuracy and comprehensiveness of chemical reaction kinetics on combustion expression. Cox and Cole[28] added a number of elementary reaction at a low temperature oxidation reaction to expand and improve the ignition process of the Shell model, and finally formed the Cox-Cole model which contained 10 components and 15 elementary reactions. Hu and Keck [29] added a number of reverse reactions on the basis of Cox-Cole model, and amended some constant values, and extend the temperature of the model to 700~1300K to form a Hu-Keck model. Focusing on the problem that the heat release rate calculated with the model of Hu-keck is lower than the experimental results, Li et al[30] added 7 components and 11 reactions with the considered the oxidation of acetaldehyde, olefin and carbonyl, and the formation mechanism of CO. The results showed that the model can simulate the oxidation characteristics of hydrocarbon fuel at low temperature and middle temperature stage, but it still lacks for high temperature combustion. In recent years, with the application of computers in the field of chemical reaction kinetics, the research on the kinetic
mechanism of chemical reactions been explosive growth in the study of the kinetic mechanism of chemical reactions. Only in the study of the n-heptane model, Heidelberg University proposed Nehse[31]model, imperial College London proposed the Lindstedt and Maurice models and the French Nacy University proposed the Buda [32] model. On the basis of this, a series of diesel chemical reaction mechanism models have been carried out. Bui-Pham[33] and Ranz studied the semi detailed mechanism of the diffusion flame of n-heptane and the decomposition and oxidation of n-heptane respectively, but these research were only included in the NTC region in the low temperature oxidation of n-heptane in 1995, LLNL ’s Curran [34] et al studied the ignition delay time and reaction type of n-heptane by means of bellows, fast compressors and flow reactors, and constructed 561 components and 2539 elementary reactions, which covers high and low temperatures. The model can be used for the reaction mechanism of n-heptane with a pressure range of 1 to 42 atm and a stoichiometric ratio of 0.3 to 1.5, and has been widely used in 1998 .Pitz [35]found that the combustion properties of the mixtures of n-heptane, isoctane, methylcyclohexane, and toluene were close to those of diesel fuel. The combustion characteristics of the mixture of n-heptane, iso-octane, and toluene could be approximated by TRF in 2007. In 2008, Andrae et al. [36]proposed a detailed combustion reaction mechanism for a mixture of TRF. Ra and Reitz et al. proposed a simplified mechanism of TRF, including the backbone mechanism of n-heptane and the submechanism of toluene on the basis of the study of Andrae[37]The simplified mechanism showed good adaptability and practicability. In 2009, Chen[38] et al. proposed a simplified mechanism of n-heptane toluene, which contained the PAH sub mechanism including the reaction of the reaction. Tianjin University has also put forward a TRF+CHX model of 80% heptane +10% toluene +10% cyclohexane for the construction of diesel mechanism.

With the development of a simplified mechanism, it has become possible to develop a series of multi-component fuel mixing mechanisms based on the diesel chemical mechanism. Xu[39] established a mechanism for the combustion of diesel/methanol blended fuel combustion cylinders on the basis of the chemical reaction mechanism between diesel and methanol. The results showed that the addition of methanol changes the original n-heptane oxidation of free radicals, the formation and consumption of OH and HO2· radicals in the pool led to inhibition of the spontaneous combustion of n-heptane by methanol, the inhibition increases firstly and then decreases with the temperature increasing. Wang et al. [40] constructed a simplified PAH model of blended fuel containing diesel and n-butanol, there were 76 components and 349 elementary reactions. It was pointed out that n-butanol extended the length of the flame which could enhanced air entrainment near the flame, and the oxygen content in the butanol reduced the equivalence ratio in the rich region. Finally, he optimized the intake pressure, EGR, post spray and n-butanol content in the blended fuel. Yan [41] made his research on the methanol/dimethyl ether and constructed a chemical reaction mechanism which contained 97 components and 508 elementary reactions. It came to conclusion that the low-temperature reaction rate slow down, and the high-temperature oxidation reaction delayed after the fuel is added with methanol.

It can be found that the history of chemical reaction mechanism is a progress from simple to complex, effective simplification of the chemical reaction kinetic models are the direction of the continuous development. Diesel and F-T diesel have similar compositions with n-heptane, iso-octane, cycloalkane, and so on. Therefore, the mechanism which have been developed for years can be used to construct the mechanism of F-T diesel-methanol-diesel-biodiesel blended fuel. The next step what we should do is constructing accurate mechanism of the blended fuel in a wider application range of temperature range and simplify form based on these studies.

4. Prospect
The development of F-T diesel-methanol-diesel-biodiesel blended fuel is an important development direction for optimizing the energy using, increasing the engines’ heat efficiency and improving the emission. However, there are also some deficiencies in the study of F-T diesel-methanol-diesel-biodiesel blended fuels. Future research can be carried out in the following parts:
(1) Combining with the influence of pressure and temperature, the blend ratio’s effect on the mutual solubility characteristics and physicochemical properties could be carried out to get the universal law of blended fuel under different pressure, temperature and blend ratio in the future research.

(2) The research could be constructed to focus on the turbulent kinetic energy, cavitation coefficient and different Reynolds number in different engine working condition with the mechanism of atomization. It can be also concentrate in the study of the F-T diesel-methanol-diesel-biodiesel blended fuels’ effect on the mechanism of atomization.

(3) Constructing the chemical reaction mechanism of F-T diesel-methanol-diesel-biodiesel blended fuel under extreme operating condition such as full load condition or high altitude. Based on the model, constructing the reciprocity between the four fuels and difference with the normal operating condition.

(4) Constructing the research on the combustion and emission characteristics of F-T diesel-methanol-diesel-biodiesel at different blending ratios. Combined with the technique of engine calibration, finding the maximum efficiency point of the blended fuel.

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