

Key Factors and Improvement Measures Changing the Gum Content of Stored Fuels

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Abstract: This paper reviews the research that has been reported in recent years on the stability of liquid fuels. It has been shown that gum is formed as a byproduct of unstable components, and the existent gum content in fuel increases continuously during storage, resulting in a gradual decline in oil quality. The primary purpose of this study is to evaluate the key factors and improvement measures for reducing the gum content of fuels. The study also presents the oxidation mechanism involved in gum formation and indicates the main parameters and specific storage conditions influencing gum accumulation in fuel. We have found that unsaturated olefins, sulfides, nitrides, metals, oxygen, additives and solar radiation have different impacts on gum formation, among them, unsaturated olefins have the most impact on gum formation. In the report, we have also made recommendations for reducing gum accumulation, such as using fiberglass reinforced plastic (FRP) storage tanks, improving the refining process, and integrating nitrogen content into the gasoline quality standard. Moreover, big data and intelligent analysis methods can also be used to model for the change of existent gum content in various fuels. This model can then be used to predict the maximum allowable retention periods of different fuels.

Keywords: Oil Composition, Existent Gum, Fuel Storage, Unsaturated Olefin, FRP Tank

1. Introduce

During the entire lifetime of fuel, from when it is first produced in the refinery, until the moment when it is consumed by the engine, the problem of fuel instability always exists. The instability of liquid fuels has increasingly attracted the attention of those responsible for both commercial and military fuel supply systems [1]. In order to ensure that the properties of the fuel remain stable during production, storage, and transportation, gasoline needs to have oxidation stability, and one of the most important indicators related to it is the gum content [2].

Under specified conditions, evaporation residue of fuels without any further treatment is called the existent gum, which has adhesiveness and is often used to evaluate gasoline or diesel's tendency to form gum in the engine. The existent gum content is one of the most significant indicators of oil quality,

which can help determine how long the fuel can be stored [3]. In most cases, the safety of the intake system can be ensured when the gum content of gasoline is low; otherwise, the possibility of deposits forming in the engine and fuel system increases, and it will cause problems such as engine choking, development of coke, engine power loss, increased fuel consumption, and environmental pollution [1]. In severe cases, abnormal engine noises or start failures will occur. In addition, these deposits can serve as binding agents for water, dirt, rust and other corrosive products that occur in the fuel distribution systems. Therefore, the rate and quantity of gasoline gum formation should be strictly controlled.

Currently, scholars around the world have been studying the complex mechanisms involved in gum formation. Various experimental studies have been carried out to find the main factors influencing the existent gum content, including the composition of fuels and their storage conditions, but there is a lack of comprehensive reviews. Therefore, this paper

reviews the research progress of the structure, formation mechanism and influencing factors of the colloidal molecules in gasoline, and gives some measures to control the stability of the fuel for the reference of the personnel of the fuel supply system.

2. Effect of Fuel Constituents on the Existent Gum

Specific components in fuels can generate various non-volatile high-molecular-mass gums under conditions that accumulate to adverse effects. Studies have shown that the higher the content of unsaturated hydrocarbons and non-hydrocarbon compounds such as sulfur, nitrogen, oxygen, and chlorine in oils, the higher the possibility of gum formation [4].

2.1. Effect of Unsaturated Hydrocarbons

Many scholars have discussed the mechanism of gum formation in fluid catalytic cracking (FCC) gasoline. Zhang [5] et al. showed some conclusions. Conjugated diolefins, fused ring aromatics with alkyl side chains, and olefins with double bonds near the center of the structure are oxidized first, and it has the most significant influence on gum formation,

especially in an environment containing sufficient oxygen or light. Paraffin has stable chemical properties so as not to affect the stability of fuels. The straight-chain paraffin can inhibit the formation of gums up to a certain extent.

Changsoo Kim [6] et al. showed that the precursors of gums produced by oxidation in gasoline are hydrocarbons, and the instability of catalytic oxidation decreases in the order of diolefins, aromatic olefins, mono-olefins, aromatics, isoparaffins, naphthenes, and paraffins. Nagpal [7] and Zanier [8] found similar result.

Nagpal J M [9, 10] observed the primary deposit precursors viz. straight or branched olefins. He proposed that they differ in their contribution to the potential of gum formation. Through aging (ASTM D873, 4 hrs), the various synthetic blends containing straight chain olefins, isoolefins, and a cyclic olefin, he measured their total potential gums. The results are showed in Figure 1 and Figure 2. The studies pointed out that the magnitude of the gum content varied with different types of olefins. The curves showed an increase in the gum-forming tendency with the increase in the carbon number of the same type of straight chain olefins. It also indicated that cyclohexenes have a greater contribution to gum formation compare to straight chain olefins and iso-olefins, but less contribution than cyclic diolefins.

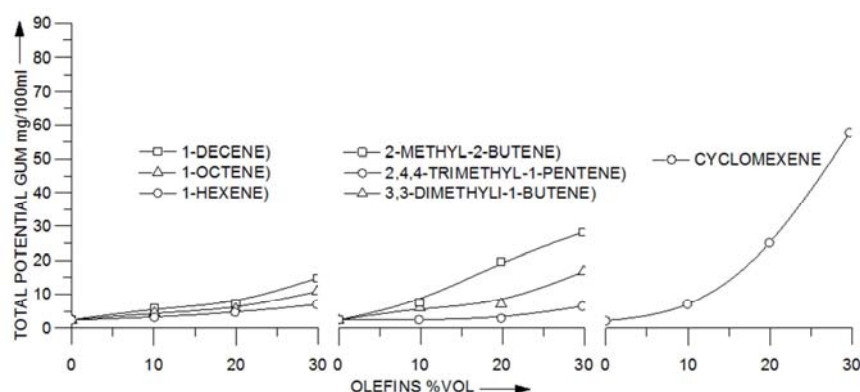


Figure 1. Variation of total potential gum for various concentrations of pure olefins (ASTM D873, 4 hrs. aging) [7].

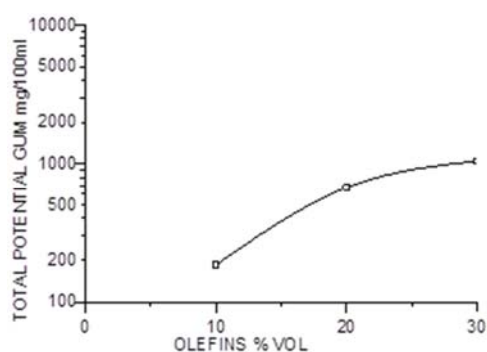


Figure 2. Variation of total potential gum for various concentrations of 4-vinyl-1-cyclohexene (ASTM D873, 4 hrs. aging) [10].

Nagpal J M's work showed that straight chain olefins have the least impact on the potential of gum formation among various olefin types. With the increase in molecular weight,

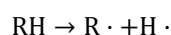
the gum-forming tendencies of olefins with double bonds at the straight chain end are larger than those in the straight chain. The position of branching in the iso-olefins and double cyclic structures play important roles in gum formation.

Relevant research pointed out that olefins have the least stability among hydrocarbons, especially comparing with highly reactive, conjugated diene compounds, branched olefins, and aromatic hydrocarbons double bonds on the side chains [11]. Olefins participate in reactions such as oxidation, polymerization, and condensation with the combined action of light, heat, oxygen, and metal catalysis, which is the main cause for gum formation [10]. Olefins can also act as initiators for the oxidation chain reaction. When the content of heteroatoms or unsaturated hydrocarbons is still significant because of the insufficient hydrogenation of the fuel, the polycondensation reaction can occur even with a small amount of diolefins. In turn causes substantial increase of the potential gum [11]. Therefore, if fuel contains a large amount

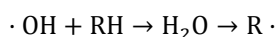
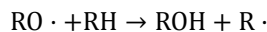
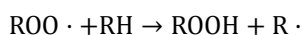
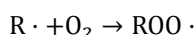
of unsaturated diene, the gum content increases significantly with the increase of unsaturated diene content [10, 12, 13].

Heneghan S P [14] *et al.* and Huang [15] *et al.* suggested that diolefins promote the gum formation by participating in free radical chain oxidation reactions. The C-H bond at the α -position connected to the π bond of the olefin is lower in homolytic fission energy and easier to break. Therefore, hydrogen atoms of olefins can be easily attacked and captured by oxygen molecules, causing a radical chain reaction to generate a hydrocarbon radical $R\cdot$ and a hydrogen peroxide radical $HOO\cdot$. The free radicals react with each other to yield high-molecular-weight compounds ($R\cdot + R\cdot$, $R\cdot + ROO\cdot$, $ROO\cdot + ROO\cdot$) after accumulating sufficient concentration of $R\cdot$ and $ROO\cdot$. If there are still active hydrogen atoms on the α -carbon connected to the π bond, the hydrogen atoms can be seized continuously by the oxygen atoms to undergo a secondary oxidation reaction, which will lead to the generation of polar and high-molecular-mass compounds and forming stable and insoluble deposit [15]. The reaction mechanisms are as follows.

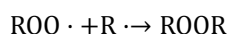
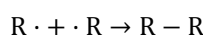
Initiation reaction:



Propagation reaction:



Termination reaction:



For straight chain olefins, if the double bond is at the end of the carbon chain and the π bond has a strong ability to adsorb electrons, the polarity of the C-H bond connected to the unsaturated bond can be enhanced. Therefore, the double bond will be broken more easily by the oxygen molecules, forming free radicals, thus reducing the fuel's stability. If the double bond is inside the carbon chain, the electrons around the π bond are divided equally between the two ends rather than one side, creating a relatively stable double bond.

For the cyclic olefin, the carbon-hydrogen ratio is high. The carbon atom is highly electronegative, which means that it has a robust electron-withdrawing ability. More electrons are adsorbed around carbon atoms on the double bond, which will increase the polarity of the C-H bond and destabilize the double bond to form free radicals.

In general, there is a strong correlation between the formation rate of existent gum and the composition of hydrocarbons in fuels. The position and number of double

bonds can affect the degree of oxidation of olefins with different chemical structures very differently. In terms of the gum formation, cyclic olefins and iso-olefins have the highest formation rate, whereas paraffinic hydrocarbons have the highest inhibition for existent gum formation, and aromatic hydrocarbons do not have a significant impact [16-18].

2.2. Effect of Sulfide

The proportion of active sulfur in sulfide generally increases with the decrease of distillate density. The active sulfur content in fuel is directly proportional to the gum content. Hence, the sulfur content is one of the main factors impacting the gum content and is an excellent indicator of the quality of light fuel.

Based on related research reports, there are four main ways for sulfide to increase the gum content.

(1) Metals are corroded by sulfides to produce metal sulfides and some metal-organic compounds, which will induce the gum formation.

(2) Gum formation can be accelerated by certain sulfides acting as initiators. Mercaptans, thiophenes, sulfoethers, disulfides, and sulfurs, can promote the gum formation and shorten the induction period of fuels. For the oil containing organic nitrides, especially basic nitrides, they can be used as a catalyst with sulfide to accelerate the reaction and generate colored substances, resulting in the darkening and instability of fuels [19].

(3) The sulfur compounds participate in a series of complex chemical reactions during fuel storage. Eventually, they become gums existing in fuels or adhering to the fuel container wall. Acidic substances in fuels can react with hydrocarbons or sulfides to produce high-molecular-weight insoluble substances [20].

(4) Some sulfur compounds are natural gums.

The number of sulfur compounds directly appearing in the form of gum is small. Furthermore, the amount of gums produced by sulfur compounds directly participating in or inducing a chain reaction is limited. In most cases, fuels contain metal ions and a large amount of hydrocarbon. Metals in contact with the fuels can be corroded by sulfurs to form sulfur compounds, which can induce the chain reactions to generate gums. The chain reactions will stop when the active sulfurs are exhausted. Therefore, active sulfur in fuels can directly impact gum formation.

2.3. Effect of Nitrogen Compounds

It is commonly known that the presence of nitrogen-containing compounds will promote the oxidation of hydrocarbons in fuels and generate insoluble gums, leading to instability and darkening of fuels [21].

Zhang [5] studied the effects of a basic nitrogen compound (2-methylquinoline) and a non-basic nitrogen compound (pyrrole) on the induction period of gasoline. The results are showed in Tables 1 and 2. With an increase in 2-methylquinoline and pyrrole content in gasoline, the oxidation induction period gradually decreases. The difference between the maximum and minimum values of the induction

period of gasoline with 2-methylquinoline added is 85.25 minutes, while the pyrrole added is 101.25 minutes, which indicates the effect of pyrrole on the induction period of gasoline is greater than that of 2-methylquinoline. In conclusion, both basic nitrogen compounds and non-basic nitrogen compounds can reduce the stability of fuel, but basic nitrogen compounds have smaller variance in the induction period.

Table 1. Correspondence between 2-methylquinoline content and induction period of gasoline.

2-methylquinoline (μg/g)	10	20	40	100
induction period (min)	407.25	394.75	364.00	322.00

Table 2. Correspondence between pyrrole content and induction period of gasoline.

Pyrrole (μg/g)	5	8	10	30
induction period (min)	452.25	420.75	351.00	264.00

Bauserman et al. studied 21 middle distillate fuels about the relationship between organic nitrogen content and fuel stability. They observed that not one specific organic nitrogen compound (both basic nitrogen compounds and non-basic nitrogen compounds) or a group of compounds is responsible for instability, but their interaction is the leading cause of instability. Stable fuels may contain more non-basic nitrogen compounds than basic organic nitrogen compounds, or vice versa. When the stored fuel is mixed, the concentration of non-basic nitrogen compounds and basic organic nitrogen compounds may be disrupted, causing an equilibrium shift in the reaction, thus resulting in precipitation or gum formation [22].

Zhou [21] et al. selected quinoline and indole as representative model compounds of basic nitrogen and non-basic nitrogen in lube base oil, to study the oxidation performance of nitride on oil. The results indicated that nitrogen compounds accelerated hydrocarbon oxidation through promoting the decomposition of hydroperoxides, while the oxidation products of nitrogen compounds could prevent the hydrocarbons from oxidation. The action of nitrogen compounds and their oxidation products were mutually competitive. Experiments on FCC diesel base oil and its model base oil were carried out by Qi [23] et al. The results show that both non-basic nitrogen compounds and basic nitrogen compounds can affect the stability of the oil product and forming dark-colored polymers which make the oil product darker. Thompson [24] also observed that non-basic nitrogen compounds promote gum formation; the insoluble colloid content caused by pyrrole in the nitrogen-containing compound shows the most substantial impact on gum formation, whereas pyridine has the lowest reactivity.

Overall, the effect of the basic nitrogen compound on the gum formation is higher than that of the non-basic nitrogen compound. Also, the nitrogen compound's reactivity in promoting gum formation depends on its chemical structure, particularly the position of the alkyl chain on the heterocycle [22, 25].

2.4. Effect of Detergents

Different fuels have different manufacturing process requirements. Detergent can effectively disperse the gum

precursor and prevent further oxidation and polycondensation from forming the existent gum. Therefore, if detergent is added to the fuel for long-term storage, it can increase the fuel's stability to a certain extent.

Studies have shown that storage duration, followed by the quality of fresh gasoline and the detergent type, had a decisive effect on the existent gum of gasoline with detergent for the long-term storage. When adding detergent to gasoline with poor stability, the existent gum content increased linearly with the storage period, and the slope of the curve is determined by the gasoline and the detergent [26].

The existent gum content of diesel has similar changes to that of gasoline. During long term storage, the military-grade diesel changes little because of its excellent storage stability, with a small quantity of gum or insoluble matters, while the light diesel oil has a more abundant existent gum content.

3. Effect of Storage Conditions on the Gum Content

3.1. Storage Duration

It is concluded in all studies that gum formation can be accelerated by increasing the storage temperature and duration. Especially for unstable fuel, the gum content has a linear proportional correlation with the storage duration [26].

In 2007, Grishina et al. studied the kinetic of the sediment of gums formed during long-term diesel storage for up to 100 months. As the storage duration of diesel fuel increases, the oxygenated compounds containing at least two functional groups can participate in polymerization or condensation reactions, thus producing high-molecular-mass compounds or gums. In the study, they used a simplified global reaction for the deposit of gums formed in diesel, in which n molecules of low-molecular-mass compounds A react with each other to yield a high-molecular-mass gum particle A_n , defined as the (1) [27]. In addition, a kinetic equation was proposed through the dependence test between diesel storage duration and gum concentration, defined as the (2), which can be used to calculate the order of reaction in diesel.



$$\frac{dc_{\text{gum}}}{dt} = kc_{\text{gum}}^{-n} \quad (2)$$

Where c_{gum} is the concentration of gums at a given time which can be expressed as $c_A - c_x = c_{\text{gum}}$, c_A and c_x represent the initial quantity of olefins and the olefin content converted to gum respectively, k is a constant, and n is the order of the reaction.

Nagpal [10] et al. studied the effects of prolonging the aging period on potential gum formation tendencies and peroxide content of various olefins under accelerated test conditions. The results are depicted in Figure 3.

The test results indicated that gasoline containing some pure olefins has an increase in the gum content and peroxide number with the prolonged aging period under the accelerated

test conditions. For the straight chain and branched chain olefins, the total gum content and peroxide number are almost linearly proportional throughout the two to six hours of aging duration. In the case of 4-vinyl-1-cyclohexene, the total gum

formation tendency increases dramatically with an increased aging duration as compared with 1-octene and 2,4,4-trimethyl-1-pentene. The results indicated that the gum content of olefins increases with aging duration.

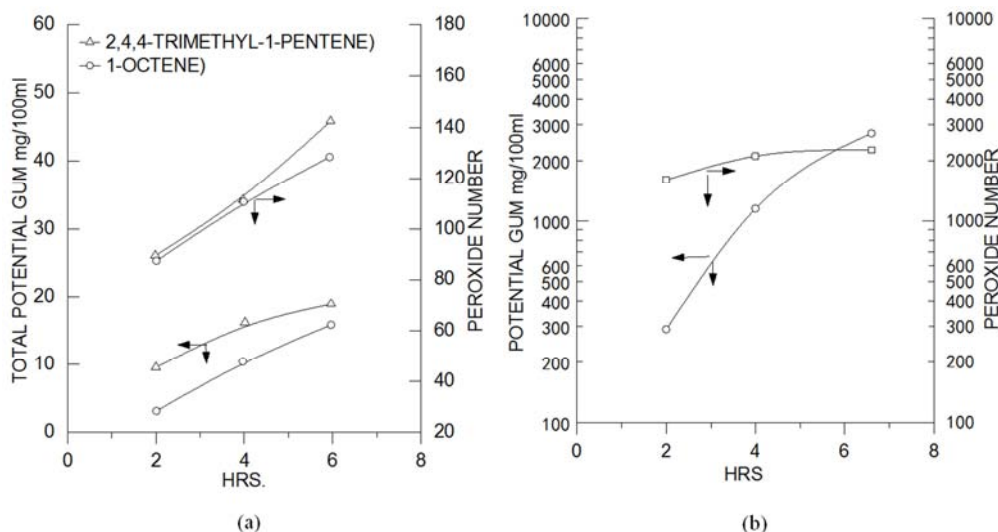


Figure 3. Effect of the aging period on potential gum and peroxide number for a) straight chain and branched olefins b) 4-vinyl-1-cyclohexene.

3.2. Storage Space Conditions

Storage conditions differ in their contributions to the potential of gum formation. The storage conditions that impact the stability of gasoline include the type and content of metals in the tank material, the temperature of the internal gas phase space, the intensity of light, and the dissolved oxygen content.

3.2.1. Metal Contamination

Metals have a significant catalytic acceleration effect on gasoline oxidation, primarily metal ions from transition metals (such as copper, nickel, iron, *etc.*), which can have a greater impact on the oxidation rate of hydrocarbons. Also, different metals contribute differently to the gum formation. Among transition metals, those with one electron transfer are the most

effective [28, 29]. A'reff [30] and Pereira [31] in gasoline and Beaver [32] *et al.* in diesel fuel observed similar behavior for copper and iron.

Teixeira *et al.* [33] investigated the relationship between the tendency for gum formation and the concentration of ions from copper, iron, zinc, lead, and nickel. The metal solutions (1000 $\mu\text{g/ml}$) added to gasohol samples were prepared by accurately weighing the amounts of each metal, followed by dilution with methanol. The washed gum content was measured for samples stored for 7, 14, 21, and 28 days. The results are summarized in Figure 4. The result shows that, in the case of Cu and Fe, there was a notable increase in gum content as the level of contamination increased. Nickel and zinc had a weaker effect on the rate of gum formation, and the influence of lead was almost negligible.

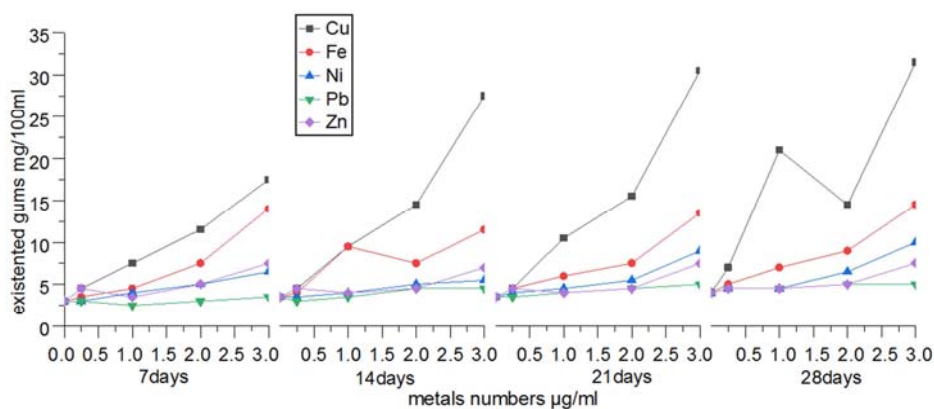


Figure 4. Washed gum content in gasohol samples under different metal contamination levels and storage periods.

Pereira [31] investigated the stability of gasoline samples with different copper contents. The solutions were prepared by

immersing pieces of copper in a container of ethyl alcohol for a month. Copper content was then determined by flame atomic

absorption spectrometry. Anhydrous ethyl fuel with different copper content was added to gasoline to study the effect on gum formation in gasoline. The experimental results are shown in Figure 5.

It could be observed that the gum content in gasoline increased with the copper content, both in the high copper and the low copper regions. In the low-content region, there was

little effect on the copper content below 0.09 mg/kg. When the copper concentration was 0.09 mg/kg, the gum content (38.6 mg/100ml) was almost equal to that of the alcohol-free sample (40.1 mg/100ml). In the high-content region, the gum content increased significantly as the copper content in alcohol increased. Therefore, gasoline must not be in contact with copper-containing metal parts during use and storage.

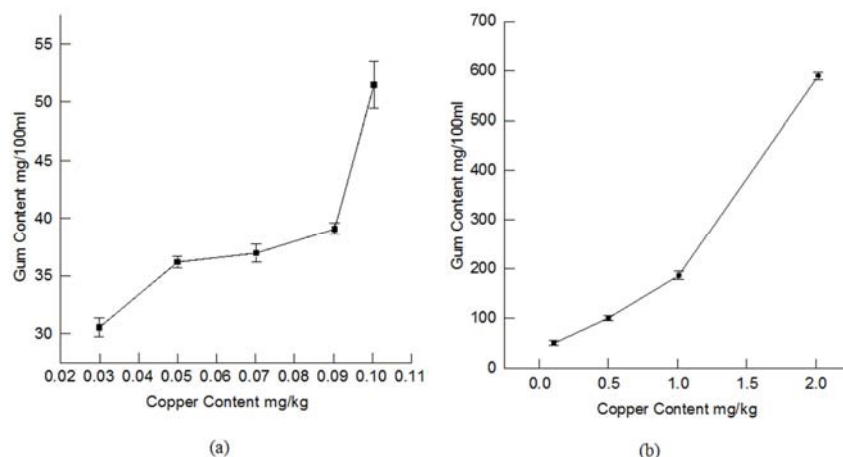


Figure 5. Gum content variation as a function of copper content in anhydrous alcohol with respective standard deviations a) low-content region b) high-content region.

3.2.2. Storage Temperature

The storage temperature has a significant influence on the reaction rate of oxidation of gasoline and decomposition of tetraethyl. Tests showed that the rate of gum formation increased by 2.4 to 2.6 times as the temperature increased every 10°C [34]. Nagpal [10] studied the effect of temperature on the gum formation tendencies in the case of 1-octene, 2,4,4-trimethyl-1-pentene, or 4-vinyl-1-cyclohexene added to pure gasoline. The results showed in Figure 6. The graph shows that the gum formation tendency of the three olefins is consistent with increasing temperature. The tendency of gum formation increases sharply with the increase in temperature, and 4-vinyl-1-cyclohexene shows the least impacted by temperature.

Therefore, lowering the storage temperature of fuels can extend its storage. Research shows that the optimal oil storage temperature for refined oils is 16-20°C.

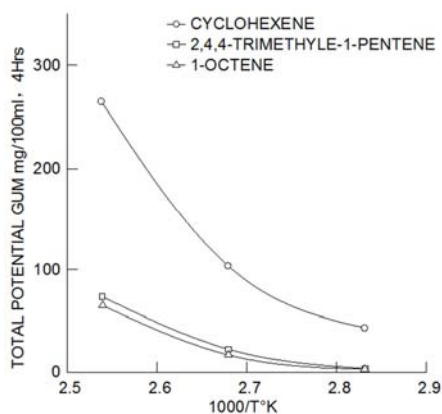


Figure 6. Effect of temperature on gum formation of pure olefins [10].

3.2.3. Light

Light accelerates the oxidation of gasoline. Gasoline absorbs energy under sunlight; then activating its hydrocarbon molecules to initiate a new oxidation chain reaction. Among all the spectrum of sunlight, violet light has the greatest effect [23]. Wang [35] studied the effect of light on the oxidation stability of biodiesel. He exposed jatropha biodiesel samples under light in different wavelengths at 20°C for 48 hours and measured their acid values and induction periods. His results are shown in Figure 7.

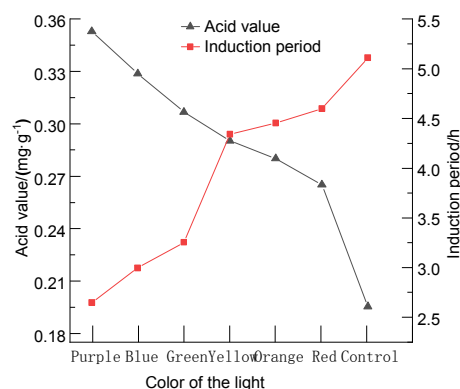


Figure 7. Effects of different wavelengths of light on acid value and induction period of biodiesel [35].

Figure 7 showed that with the increase of the monochromatic wavelength, the oxidation induction period of jatropha biodiesel gradually increased, and the acid value gradually decreased. For example, the induction period of the oil was 5.12 h with no light irradiation, 4.61 h after red light, and 2.65 h after purple light. The experimental results showed that the induction

period shortens after exposure to light. The shorter the wavelength of the irradiated light, the greater the oxidation effect. Therefore, avoiding light or adding light stabilizers (such as ultraviolet absorbers, hydroperoxide decomposers, *etc.*) during fuel storage is beneficial to fuel stability.

3.2.4. Oxygen Concentration

Oxygen flowed intermittently into the storage tank through a breather valve during the storage of fuels. It will act as catalysts to accelerate the gum formation when certain substances in fuels encounter metal ions. Shalotov [36] *et al.* studied the relationship between the amount of absorbed oxygen in automobile gasoline and chemical stability during gasoline storage. It was found that the gum content of fuel during storage is closely related to the oxygen concentration in the storage room. For gasoline, it can be stored for more than three years when the oxygen absorption ratio is less than 25%, and less than one year for 25%-85%. And the fuel should be used immediately when oxygen absorption ratio exceeds 85%.

4. Measures to Reduce Gum Formation

4.1. Reduction of Unsaturated Hydrocarbon Content

Unsaturated olefins are the main cause for gum formation. Peroxides are produced when unsaturated olefins are exposed to oxygen in air, then followed by decomposition. The decomposition products may re-oxidize, decompose, polymerize, and condense to form gums. Reducing the content of unsaturated olefins in the fuel can effectively reduce its existent gum content in the future.

Hydrofining is one of the effective methods to improve the stability of fuels. It does not remove existed gums, but it can reduce the content of the hazardous components such as unsaturated hydrocarbons by hydrogenation. Hydrofining reduces the oxidation reaction of unsaturated hydrocarbons and the gum content to a certain extent [37].

Hydrotreating of oil products does not reduce the existent gum content during storage fuels. For diesels, they are from various petrol origins, processed under different optimal hydrogenation. Furthermore, the adequacy of the hydrogenation has a great impact on the storage stability of the diesels. Wang [38] *et al.* experimented on the quality transition rules of stored light diesels with or without additives from different manufacturers. For light diesel that has not undergone hydrogenation treatment or has insufficient hydrogenation, their color and oxidation stability during storage will be worse, and their total insoluble extent will increase rapidly. Two processes may cause this phenomenon: one is that a natural gum inhibitor in the crude oil, such as phenols, is destroyed in the hydrogenation process, reducing the antioxidant capacity of the fuel [39-42]. The second process is when the hydrogenation of the oil is insufficient. High impurities or nitrogen compounds content will promote gum generation during the storage of diesel oil [43], followed by forming sludge-type substance, the final product, through polymerization of the unstable gums.

Hydrotreating can reduce the existent gum content of the fuel, but at the same time, it dramatically reduces the octane

number in gasoline. According to existing researches, several potential technical solutions have been found to reduce gasoline olefin but not octane numbers.

- (1) Alkylation. Alkylation process is the conversion of isobutane and butene (or a mixture of propylene, butene and pentene) into isooctane (alkylated oil) under the action of acid catalysts such as hydrofluoric acid, sulfuric acid or solid acid. The alkylated oil with high octane number is free of aromatics and contains only traces of olefins and oxygen, which is an ideal blending component for high-octane and low-olefin clean gasoline [44-46].
- (2) Hydrogenation of light gasoline followed by etherification. Gasoline hydrogenation has two stages of desulfurization. During the first stage of hydrosulfurization, the raw gasoline is separated into light and heavy components in the fractionation tower. During this process, large molecular thioether, produced by the reaction of mercaptan with diolefins, enters into the heavier component. At this stage, the sulfur content will reduce to 50 $\mu\text{g/g}$. At the second stage of desulfurization, fixed-bed hydrosulfurization is adopted to reduce the sulfur content to 10 $\mu\text{g/g}$. The light gasoline of desulfurization is selectively hydrogenated so that etherified gasoline with high octane number and low vapor pressure is obtained through the etherification of active olefins of $\text{C}_4\text{-C}_7$ with methanol. After the reaction, the volume fraction of olefins can generally reduce by 10%-20%, and the octane number can increase by 2-3 times [47-49].
- (3) The olefins in FCC gasoline are mostly $\text{C}_5\text{-C}_6$ olefins, which are converted into branched isomers (isomerized oil) through isomerization reaction under the conditions of hydrogen, and metal-acid dual-function catalysts. The isomerized oil has very low sulphur content, free of olefin, benzene, and aromatic hydrocarbon. It is an excellent blending component of clean gasoline [50-52].

4.2. Additive

The oxidation of gasoline depends on its chemical composition, and multiple external conditions can accelerate the reaction. Measures such as reducing storage temperature and fluctuation, reducing the oxygen concentration in the storage room, and keeping the gasoline from light exposure and metals, can slow the deterioration of gasoline but cannot resolve the deposit issue. Adapting a refining process that can remove insoluble deposits can resolve the issue, but it requires significant investment. Therefore, the most economical method is to refine gasoline and add additives to improve its stability.

Adding different additives to gasoline will impact the stability of the fuel for different reactions. According to the composition of additives and their functions performed in the engine, oil additives can be categorized into two types.

The first type is protective fuel additives, such as antioxidants, metal passivators, corrosion inhibitors, light stabilizers, *etc.* Antioxidants can convert active free radicals into inactive ones in gasoline, interrupt the oxidation chain reaction and prevent conjugated diolefins from reacting with free radicals in FCC

gasoline to form gums. Metal ions play a catalytic role in the aging of gasoline. If combined with metal passivators, the metal ions will lose catalytic oxidation capability. During fuel transportation, storage and usage, it will inevitably get in contact with metals. In these situations, the metal passivator added will reduce the catalytic effect of metal ions [53].

The second type is fuel cleaning additives, such as detergents. They will act as a cleaner to scrub off deposits.

4.3. FRP Material

Some sulfur compounds and nitrogen compounds in fuel can easily react with metals on the inner wall of steel storage tanks, resulting in corrosion, tank leakage, and oil quality degradation. Storage tanks made of new material such as fiber reinforced plastics (FRP) material have gradually received attention due to its superior corrosion-resistance and inert to many oxidation reactions [54]. The advantages of FRP material storage tanks and their effects on the gum formation is discussed in detail in Section 4.

5. The Advantages of FRP Material Storage

Before the 1960s, underground storage tanks were all made of steel material. However, traditional steel storage tanks often caused a series of problems due to corrosion and fuel leakage into soil and groundwater. In the 1960s, FRP was introduced as a new material for storage tanks. It has excellent anti-corrosion performance and gradually replaced steel as the primary material for underground fuel storage tanks. The main material for corrosion protection of FRP material products is the resin, which is resistant to ethanol fuels and is more cost-efficient over the long term. FRP materials have been widely used to fabricate fuel storage tanks.

FRP storage tanks will not cause an increase in gum content. The stability of fuel improves due to the beneficial properties of the FRP materials:

(1) Good corrosion resistance. FRP material is corrosion-resistant material to the atmosphere air, water, and general concentrations of acids, alkalis, and metal ions, as well as a variety of oils and solvents. In recent years, due to the superior corrosion resistance of FRP materials, it has become a fundamental solution for new fuels such as biofuel and alcohol-containing gasoline. Alcohol is miscible in water. Therefore, the water solubility of the alcohol fuel will increase, resulting in an increase in fuel conductivity and the proliferation of microorganisms, accelerating the corrosion of storage tanks. Compare with steel, FRP is free from microbial erosion because its material does not oxidize like the metal [51]. Steel storage tanks are also not suitable for new clean fuels such as ultra-low sulfur diesel, because corrosion could occur both in the liquid and gas phase of the storage room. Tank corrosion exists only in steel tanks, for ultra-low sulfur diesel storage, although the corrosion mechanism is not clear, it has never been found in double-layer FRP tanks.

It is inevitable for fuel to contact the storage container

during transportation, storage, and consumption. The storage tank material is made of metals, which will accelerate the oxidation reaction. The concentration of metal ions will increase significantly, especially when corrosive acid or alkali in the fuel coexisting with water and metal ions, thus intensifying the catalytic oxidation of metals [55]. In contrast, the m-phenylene resin in FRP material can resist a variety of acidic, alkaline, or aromatic compounds, solvents, and carbohydrates [56]. Therefore, for fuel stored in the FRP tank, the probability of sulfur compounds forming sulfur salts through corrosion of metals could be reduced. As a result, the probability of gum formation during the chain reaction process induced by sulfur salts can also be reduced.

(2) Low thermal conductivity. The thermal conductivity of FRP material in normal temperature conditions is 1.25-1.67 kJ/(m·h·K), it is only 0.1% -1% of that metallic material. Therefore, the FRP material tank has less heat loss than steel tanks, while containing the same medium and the same temperature fluctuation difference between the inside and the outside [57]. The temperature fluctuation of fuel inside FRP tank is relatively small. Therefore it can help to maintain the stability of the fuel and reduce gum formation.

(3) Smooth surface. The absolute roughness of FRP material is only about 0.01 mm, which is much smaller than the internal surface roughness of traditional steel storage tanks. For FRP tanks, impurities cannot be easily adsorbed and deposited on its surface. Wax formation and gel formation in the tank also reduces. The smooth surface of the tank will be easier to clean as well [57].

6. Discussion

Currently, there are some advanced gasoline quality standards in the world such as California Reformulated Gasoline Phase 3 (CaRFG Phase 3 for short) implemented by the United States, JIS K 2202:2012 implemented by Japan, EN 228:2012 implemented by the EU (EU VI for short), and the Worldwide Fuel Charter VI (WWFC VI). Among them, the EU VI and the WWFC VI are the most influential and have been used as a reference or directly adopted by other countries or regions around the world [58]. China currently implements the National VI Emission Standards For Motor Vehicles (China VI for short), mostly drawing on the experience of the EU VI.

The leading indicators of the current gasoline quality standards in the world are shown in Table 3 [59-61].

Table 3 lists some of the main indicators of the current vehicle gasoline quality standards in various countries. The main indicators include sulfur, olefins, aromatics, benzene, and oxygen content. Moreover, unwashed gums and washed gums are emphasized in the regulations. According to the Table 3, the limit of benzene content in China is stricter than that of EU VI, but the limit of olefins content is not as strict as WWFC VI and CaRFG Phase 3. Most of the olefins in gasoline come from fluid catalytic cracking (FCC) gasoline fractions [62, 63]. Therefore the difference in the regulatory limits for olefins content is inseparable from the composition of the gasoline with blending components and the

composition of refinery plant [64]. Also, WWFC VI stipulates the cleanliness of vehicle gasoline in terms of sediment,

particulate pollutants and sedimentation chambers.

Table 3. Leading indicators of current gasoline quality standards in USA, Japan, the EU and China.

properties	CaRFG Phase 3 (2014)	JIS K 2202-2012 (2012)	EN 228-2012 (2012)	WWFC VI (2019)	GB17930-2019	
					China VI A (2021)	China VI B (2023)
Sulphur content (ppm)	≤10	≤10	≤10	≤10	≤10	≤10
Olefins content (vol%)	≤10	≤10	≤18	≤10	≤18	≤15
Aromatics content (vol%)	≤35	≤42	≤35	≤35	≤35	≤35
Benzene content (vol%)	≤1.1 ¹	≤1.0	≤1.0	≤1.0	≤0.8	≤0.8
Oxygen content (vol%)	≤3.7	-	≤2.7 ≤3.7 ²	≤2.7 ³	≤2.7	≤2.7
Unwashed gums content (mg/100ml)	≤20	-	-	≤30	≤30	≤30
Washed gums content (mg/100ml)	≤5	-	≤5	≤5	≤5	≤5
Oxidation stability (min)	-	-	≥360	480	480	480
Trace metals and other contamination content	Cu≤0.1 mg/kg Visibly free of suspended or precipitated contaminants (clean and bright)	-	-	No trace metal should exceed 1 mg/kg	Pb≤5 mg/L; Mn≤2 mg/L; Fe≤10 mg/L; no methylal, Aniline, halogen and phosphorus and silicon compounds	

1) The flat limit for benzene content in qualifying small refiner is 1.0 vol.%.

2) With the increase in the use of biofuels in Europe, the European VI gasoline standard has been formulated in two versions. The two versions have different regulations on the oxygen content and the amount of ethanol added. For cars produced or used earlier that do not have a high biofuel content, the maximum oxygen content of the sum of other oxides except methanol and ethanol is 2.7 wt%, and the maximum ethanol addition is 5.0 vol.%; the remaining cars use gasoline with a maximum oxygen content of 3.7 wt% and a maximum ethanol addition of 10.0 vol.%

3) An exception to the limit shown in the table, up to 3.7% oxygen by mass (or 10% ethanol content by volume) is allowed if permitted by existing regulation.

It is known that the existent gum content in the fuel will increase with the fuel's storage duration. To ensure that the fuel is still usable after being stored for a long time, it is essential to evaluate the quality of the initially stored fuel to limit the amount of unsaturated hydrocarbons that can induce gum formation. Compounds such as cyclic olefins, sulphurs, nitrides and other unstable components should be analyzed. Although olefins are high-octane components in gasoline, they are also unstable. Therefore, the future of vehicle gasoline standard should be to meet the octane number of gasoline and reduce the content of olefins and aromatics as much as possible. In addition, the presence of basic nitrogen compounds not only causes gums during oil storage but also inhibits the hydrosulfurization reaction [65, 66]. In the current standards, there are no regulatory limits for nitrogen content. Although sulfur and unsaturated hydrocarbon content in vehicle gasoline are the key indicators, to further reduce the existent gum content, we recommend that future standard revisions should consider adding regulatory limits for nitrogen content based on the refining industry. The goal is to meet the environmental requirements and not overburden the refining industry with more capital investment.

Many factors affect the existent gum content in the fuel, and the mechanism of different components producing gum is different, so it is challenging to predict the changing rules of existent gum content accurately. However, we can collect a large amount of data on the changes of the existent gum content for different types of fuels, then combining the gum production rate equation and apply big data and intelligent analysis methods to model for the change of existent gum content over time. These models can be applied to predict maximum allowed retention periods of different fuels and provide a basis for determining their testing interval.

7. Conclusion

The existent gum content of oil products during storage will increase over time due to various factors. However, taking appropriate measures can slow down the fuel deterioration rate and extend the storage period.

- (1) Our findings show that unsaturated hydrocarbons are the major precursors to the gum formation in gasoline. The various olefinic structures present in these unsaturated hydrocarbons contribute differently to gum formation. The position of branching and the number of double bonds play an important role. Cyclic (bicyclic) and isomeric structures have been found to have the most impact on gum formation tendencies, whereas linear alkanes have a certain degree of inhibition.

The stability of fuel is directly proportional to the sulfide content, temperature, irradiation wavelength, storage period and oxygen and metals concentration. Sulfur compounds rarely participate directly in the gum formation reactions. Instead, metals are corroded by active sulfur to form sulfur compounds, then inducing the chain reaction processes to generate gums. In addition, non-basic nitrogen compounds have a greater impact on gum formation compares with other nitrides, and its reactivity depends on the chemical structure, especially the position of the alkyl chain on the heterocycle.

- (2) There are measures that can be taken to improve the oil quality due to gum accumulation, such as improving the refining processes (alkylation, isomerization, or hydrogenation of light gasoline followed by its etherification), and adding nitrogen content as a criteria for oil quality standards. Preventative measures can

also be taken to alleviate oil quality deterioration. For example, adding appropriate additives, reducing the temperature and temperature fluctuation of fuel, and reducing the oxygen content are all actions that can sustain oil quality. In terms of fuel storage, using a FRP tank can reduce the concentration of metal ions in fuel compared with the steel tank. Therefore, reducing the probability of gum formation during the chain reaction process induced by sulfide compounds. The FRP material also has better sealing and thermal insulation thus minimizing the temperature fluctuation that causes gum formation.

The oxidation of fuel to produce deposit is a very complicated process, and the specific oxidation mechanism is not yet clear. In the future, on the one hand, we should further study the existing oxidation mechanism of fuel, and focus on the reaction kinetics and chemistry of gum formation to determine the actual composition and structure of the gum. On the other hand, it is possible to collect the change data of the gum content of different fuels, and data analytics can be used to establish models to predict the gum formation over time for various fuels. This method of prediction provides the basis to evaluate maximum allowable retention period for different fuels.

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