

Corrosion Potential of Selected Bio-blendstock Fuel Candidates for Boosted Spark Ignited Engines



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Energy & Transportation Science Division
Co-optimization of Fuels and Engines Program

**CORROSION POTENTIAL OF SELECTED BIO-BLENDSTOCK FUEL CANDIDATES
FOR BOOSTED SPARK IGNITED ENGINES**

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ABSTRACT

Eight bio-blendstock fuel candidates for boosted spark ignited engines were evaluated for their susceptibility to corrode infrastructure metals. These candidates included five alcohols (methanol, ethanol, isobutanol, n-propanol, and isopropanol), diisobutylene, mixed furans, and aromatics. Methanol was also added to the list for comparison. These molecules are not corrosive to metals since they cannot dissociate into ions and anions. However, the addition of contaminants and a separate water phase may facilitate corrosion due to phenomena associated with the formation of ions and anions (from key contaminants), differential aeration cell (due to dissolved oxygen), and microbial-induced corrosion. A literature review was conducted to determine the influence of these phenomena on corrosion for the nine fuel candidates. In addition, the oxygen solubility of each fuel type was determined to ascertain their relative influence on forming and enhancing a differential aeration cell. Of the fuel candidates, only the alcohols are water soluble, and therefore have an opportunity to cause corrosion. However, differential aeration cells, by themselves have not been shown to contribute to corrosion. Other factors, such as galvanic coupling and/or crevice regions are necessary to promote corrosion in laboratory conditions. However, differential aeration as a corrosion mechanism has not been established in the field. Likewise, microbial-induced corrosion is another known corrosion pathway that has yet to be positively identified for blends of the fuel candidates with gasoline. In summary, corrosion is not anticipated with any of the bio-blendstock candidates with infrastructure metals.

1. INTRODUCTION

The overarching goal of the U. S. Department of Energy's multi-laboratory Co-optima effort is to co-optimize fuels and engines to achieve improved efficiency and operational performance. The fuel candidates under consideration all have synthesis pathways derived from bio-blendstocks. They include alcohols, esters, ethers, alkenes, alkanes, aromatics, and furans. These fuel molecules were evaluated according to physical properties relevant to combustion performance in a reciprocating engine. In addition to their properties related to engine performance, compatibility with existing fuel systems is also considered a critical performance metric. Compatibility is a generic term relating to harmonization of the fuel chemistry with metals and polymers. For polymers compatibility usually refers to the level of solubility that exists between the fuel and the polymer, while for metals, it usually means susceptibility to corrosion (oxidation leading to mass loss).

Researchers at Oak Ridge National Laboratory, Argonne National Laboratory, Sandia National Laboratory, Idaho National Laboratory, Lawrence Berkeley National Laboratory, Lawrence Livermore National Laboratory, Pacific Northwest National Laboratory, and the National Renewable Energy Laboratory have been identifying and evaluating potential fuel candidate molecules to be evaluated for their combustion, chemistry, and compatibility properties with current and advanced engine concepts. The early studies identified over 35 bio-blendstock single compounds and blend formulations using a Tiered screening process [1-2]. Nearly 400 candidates were initially identified in Tier 1, with about 40 passing to Tier 2. Tier 2 screening reduced the number further to eight Tier 3 fuel candidates (ethanol, isobutanol, n-propanol, isopropanol, diisobutylene, mixed furans, cyclopentanone, and aromatics) were selected for further consideration. The authors also considered methanol even though it is not a fuel candidate. Methanol has similar chemistry to the other alcohols and there exists anecdotal evidence that it contributes to corrosion. As such, its inclusion is useful to provide a more complete understanding of the mechanisms contributing to metal corrosion. The molecular structures of these candidates (plus methanol) are shown in Figure 1. The only candidates having ring structures are the furans and aromatics. The structures of the fuel candidates significantly influence their corrosion potentials.

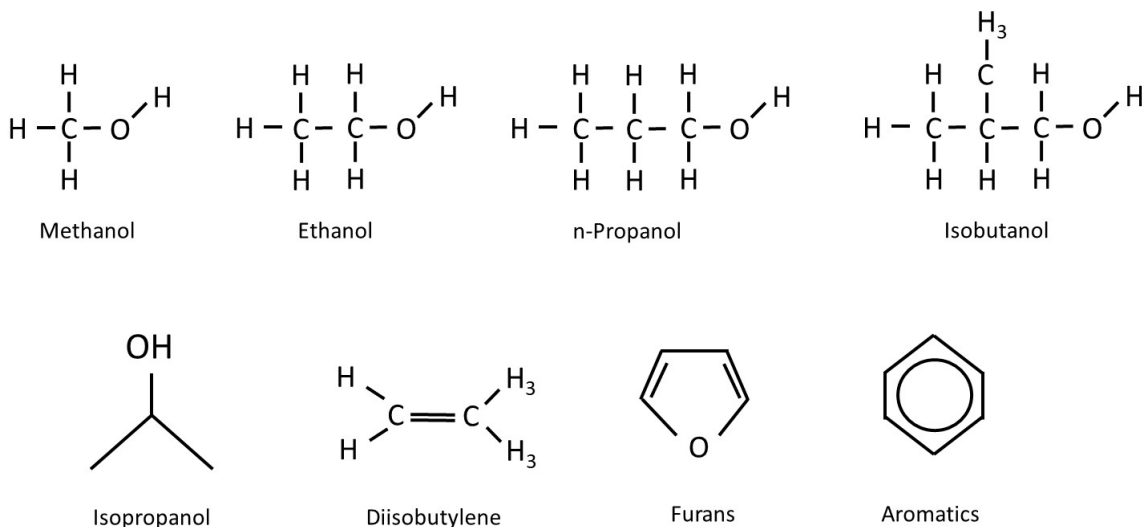


Figure 1. Molecular structures of the Tier 3 fuel candidates.

The compatibility of fuel chemistry with materials used in containment is very important for ensuring human and environmental safety. Incompatibilities are usually manifested as leaks or failures caused by either polymer swelling or corrosion of metallic surfaces. Metal corrosion is a serious concern since

many of the underground storage tanks and associated piping are composed of steel. To a much lesser extent aluminum and copper are also used. Other components containing metal alloys include valves, sensors, meters, riser tubes, and fittings.

When metals or metal alloys contact pure hydrocarbons, such as is the case for petroleum-derived fuels, Fischer-Tropsch fuels, renewable diesel and renewable gasoline, there is no reaction between these fuels and the metals in contact with them. Corrosion due to galvanic coupling is also low due to the low electrical conductivities of these fuels. Since these fuels do not dissociate into anions and cations, they are inherently poor conductors of electricity as seen in Table 1 [3].

Table 1. Electrical conductivity of the fuel candidates and methanol

Candidate	Electrical Conductivity Siemen/cm
Methanol	1.5E-9
Ethanol	1.9E-9
n-propanol	9E-9
Isobutanol	1.6E-8
Isopropanol	6E-8
Diisobutylene	N/A
Furanic mixture (60% dimethylfuran/40% 2-methylfuran)	N/A
Aromatics	N/A
Cyclopentanone	N/A

However, this situation changes if sufficient quantities of contaminants are present. The presence of polar compounds and inorganic impurities in petroleum products may significantly increase electrical conductivity. The primary fuel contaminants of concern are sulfur, potassium, sodium, and chloride. Sodium and chlorine typically originate from sodium chloride present in the environment as road salt, but chlorine contamination can also be derived from contact with chlorinated polymers, such as polyvinyl chloride (PVC) piping. Potassium originates as a carryover from the feedstock material, as does sulfur, which is often derived from the environmental conditions. These and other contaminants may also be introduced during handling and transport.

In addition to electrical conductivity, the chemistry of the fuel itself may be corrosive if it contains acids or olefin branches that are susceptible to acid formation (as is the case for many esters). None of the molecular structures (depicted in Figure 1) have these features, and therefore, they are not considered to be corrosive to infrastructure metals as pure molecules, including methanol. This situation can change, however, if water and, especially water plus microbes are present. Water is almost always present in fuel storage systems, either as water bottoms or as moisture in the headspace (ullage) region. Critical properties affecting aqueous-based corrosion are fuel solubility in water, the solubility of oxygen in the fuel, and whether the fuel molecule can be processed by the microbe into an acid.

Alcohols are poor electrical conductors since they are not electrolytes (i.e. they do not separate into anions and cations). The self-disassociation of ions is low meaning the conductivity is low. However, the dissolution of conductive contaminants and water will greatly affect the conductivity. The electrical conductivity has been measured by Kirk et al. as a function of both dissolved ethanol and water concentration in gasoline as shown in Figure 2 [4]

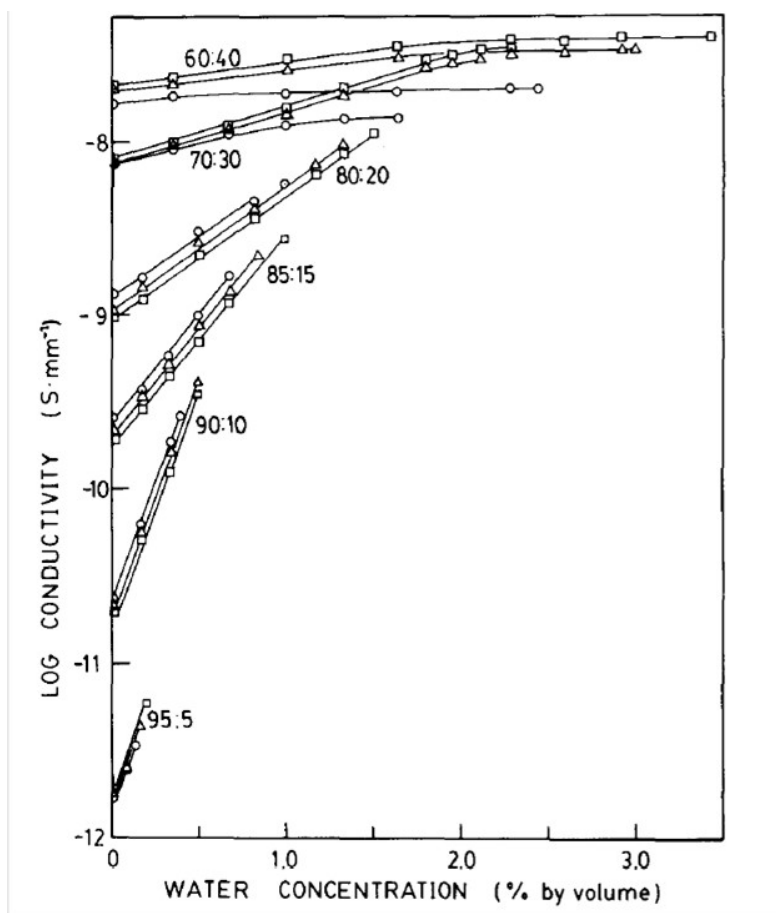


Figure 2. Electrical conductivity of gasoline-ethanol blends containing water up to the maximum solubility point (D. W. Kirk, Fuel, 1983 (62) 12: p. 1512-1513).

In Figure 2, the electrical conductivity is plotted against the water concentration for different fuel blends of gasoline containing different levels of ethanol. The ethanol contents evaluated were 5, 10, 15, 20, 30 and 40% ethanol. As can be seen in the figure, the electrical conductivity increases with ethanol concentration, and with increasing water content. However, as the ethanol content increases the rate of increase in electrical conductivity with increased water content is reduced. It is important to note that even though the electrical conductivity is increased, the values shown in Figure 2 are too low to be considered highly conductive. On the other hand, water will readily dissolve many conductive contaminants such as NaCl. Therefore, the higher the water content, the greater the ability to dissolve higher levels of contaminants.

The two most common transportation fuels are gasoline and diesel. Gasoline is primarily dispensed as E10, a blend of gasoline containing up to 10 percent (by volume) of ethanol. Neither E10 nor diesel are considered corrosive by themselves. However, the presence or formation of a separate aqueous phase (water), can provide a pathway towards metal corrosion. The potential of this water phase to be corrosive depends on several factors, including fuel solubility, toxicity, dissolved oxygen, sulfur, nitrates, and other nutrients) and the presence (or not) of microbes able to degrade the fuel structure. Of these, the two most important are the oxygen concentration and the presence of microbes. Dissolved oxygen leads to the creation of a differential aeration cell, while the presence of microbes may lead to microbial induced corrosion (or MIC).

2. ANALYSIS

2.1 WATER SOLUBILITY AND ELECTRICAL CONDUCTIVITY

A key feature of corrosion potential of the candidate fuel molecules is their susceptibility to dissolve into water. A listing of the known water solubility for each fuel type is shown in Table 2. Of the eight molecules examined in this study, only the alcohols are soluble in water. Methanol, ethanol, n-propanol, and isopropanol are fully miscible in water under ambient conditions, while the maximum concentration of isobutanol that can be dissolved is around 8.7 volume percent. The other fuel candidates are considered insoluble. This solubility is important for two reasons: 1) the dissolution of these molecules in water serve as nutrients for microbe propagation, and 2) their presence in water greatly increases the level of dissolved oxygen which also provides nutrients for microbes as well as enhancing the ability of the water phase to serve as a differential aeration cell (discussed later).

Table 2. Electrical conductivity and water solubility of the Tier 3 fuel candidates.

Candidate	Solubility in Water w/w%
Methanol	Total
Ethanol	Total
n-propanol	Total
Isobutanol	8.7
Isopropanol	Total
Diisobutylene	Insoluble
Furanic mixture (60% dimethylfuran/40% 2-methylfuran)	Insoluble
Aromatics	Slightly soluble
Cyclopentanone	Slightly soluble

2.2 DIFFERENTIAL AERATION CELL

A differential aeration cell may form due to concentration differences of dissolved oxygen in the aqueous phase. The mechanisms responsible for inducing a concentration gradient are usually caused by surface deposits or crevices on the metal surface. As shown in Figure 3, the dissolved oxygen promotes a corrosion reaction by creating and/or maintaining the cathodic and anodic reactions of $O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$, and $2Fe \rightarrow Fe^{++} + 4e^-$, respectively. The follow-on reaction of the Fe^{++} ion with the OH^- radical leads to the formation of $Fe(OH)_2$, which subsequently transforms into $Fe(OH)_3$.

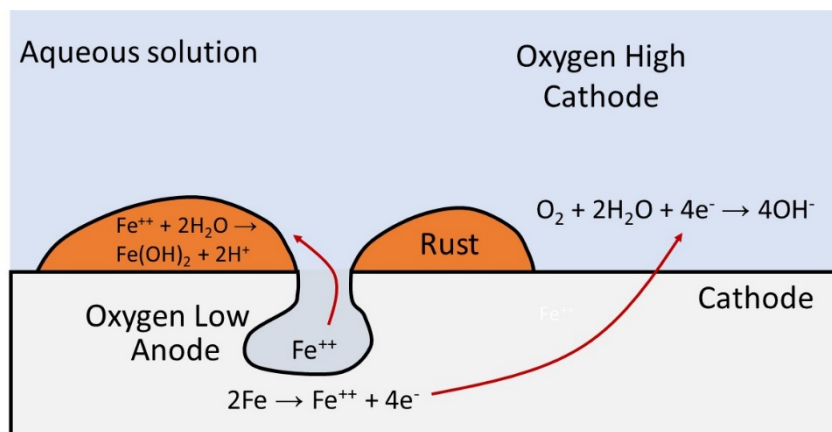


Figure 3. Schematic of the corrosion accompanying a differential aeration cell.

A critical factor controlling the reaction rate is the concentration of dissolved oxygen in the solution. The higher the concentration of dissolved oxygen the more rapid will be the rate of corrosion. Using the method described by Sato et al. [5], the solubility of oxygen for each fuel molecule was determined. The resulting oxygen solubilities are shown in Table 3 along with values provided by Sato [5]. The solubility of oxygen (expressed as the mole fraction, X_G) is significantly greater in the fuel candidates than for water [5-7]. Therefore, the maximum level of dissolved oxygen in the water region is higher for those candidates (alcohols) that exhibit some level of solubility in water. Since the corrosion rate for a differential aeration cell corresponds to the level of dissolved oxygen, then the potential for alcohols ranked from highest to lowest is isobutanol > isopropanol > n-propanol > ethanol > methanol. However, for alcohol concentrations greater than 8.7%, isobutanol would have the lowest potential of the alcohols, since it is only soluble in water at this level, while the other alcohol candidates are fully miscible. It is important to note, however, that other factors (occlusions, crevices, galvanic coupling) significantly contribute to this corrosion process. Studies have shown that, in the absence of these other factors, corrosion will not be significant based on the oxygen concentration gradient alone. It is also worth noting that differential aeration cells do not normally occur in nonaqueous solutions.

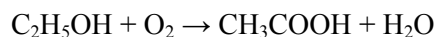
Table 3. Solubility of oxygen in Tier 3 fuel molecule candidates and water. The solubilities of the Tier 3 candidates were derived using the approach described by Sato et al [5], while the water results were obtained from Tokunaga [7].

Alcohol Type	Solubility of oxygen, X_G
Methanol	4.15(E-4)
Ethanol	5.71(E-4)
n-propanol	6.76(E-4)
Isobutanol	10.0(E-4)
Isopropanol	7.78(E-4)
Di-isobutylene	73.6(E-4)
Mixed furans	8.9(E-4)
Cyclopentanone	4.4(E-4)
Aromatics	7.4(E-4)
Water	2.66(E-5)

2.3 MICROBIALY-INDUCED CORROSION (MIC)

Microbially-induced corrosion is the degradation of metals (such as steels, aluminum, and copper alloys) resulting from the microbial production of organic acids, especially acetic acid [8]. It is one of the most pervasive and damaging means of metal degradation in the United States. Hydrocarbon fuels, especially diesel, are essentially sterile when first produced from the fractionation process in the refinery. However, they become contaminated with microbes, which originate from the air, water, and soil, during storage and transport. Although the microbial contamination can survive in these fuels, they normally do not grow (or propagate) unless the water is separate (undissolved) from the fuel. In order to propagate, the microbes need nutrients plus water. These nutrients include hydrocarbons, CO₂, nitrates, oxygen, phosphorous, and sulfates. The fuel chemical structures provide the necessary hydrocarbons, while the other nutrients normally exist as contaminants in the fuel. Microbes preferentially process low molecular weight aliphatic hydrocarbons.

A schematic depicting MIC on steel (or iron) is shown in Figure 4. The microbial film forms on the surface and if ethanol is present in the water phase, it is utilized by the microbes (especially *acetobacter*) to convert ethanol (C₂H₅OH) into acetic acid (CH₃COOH) according to the following reaction [8,9]:



Acetic acid then reacts with Iron to form iron oxide (rust):

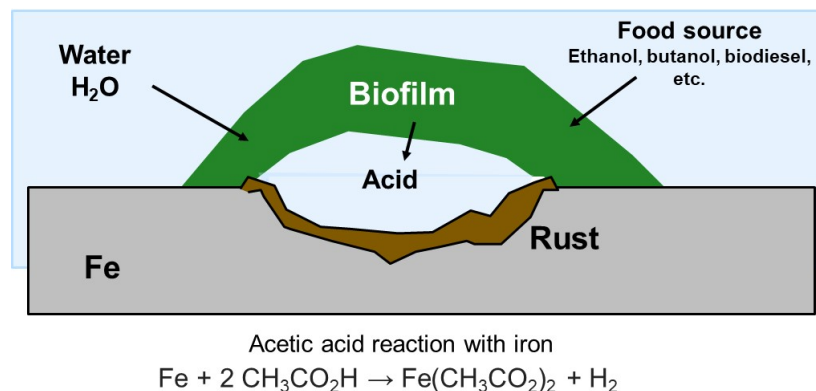
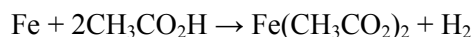


Figure 4. Schematic showing the processes associated with microbial induced corrosion.

There is considerable diversity in the types of microbes that can propagate in hydrocarbon fuels (8-13). Around 30 types of bacteria, 80 types of fungi, and 12 or more types of yeast have been identified as capable of degrading hydrocarbons found in transportation fuels. Microbes preferentially process low molecular weight alkanes (or aliphatic hydrocarbons). In contrast, aromatics are not normally utilized by microbes and are therefore not considered biodegradable. Therefore, the aromatic content of gasolines can serve to inhibit microbial growth. As a result, MIC is not common for neat gasoline. The quantity of water necessary to propagate microbes is extremely small. Microbes can propagate in water layers only a hundred microns deep, which is nearly undetectable. Biocides and rapid turnover of the fuel are the primary means of controlling MIC, as it is extremely difficult (and costly) to remove moisture in fuel systems. The susceptibility of the candidate fuels to promote MIC is based on their chain length, level of aromatics, dissolved oxygen, and solubility in water.

Alcohols, especially ethanol, are known to contribute to MIC (8-13). Microbes have been demonstrated to convert ethanol and isobutanol into acetic and isobutyric acids, respectively [14,15]. The ability of microbes to process methanol, n-propanol, and isopropanol into short chain acids has not been reported extensively in the literature, but it is reasonable to expect that these three alcohol types may also be processed into short-chain acids. The isobutylene molecule has also been shown to be attacked by aerobic bacteria [15], though it has not been demonstrated that this mechanism leads to acid or hydrogen formation. The ring structures of cyclopentanone, furans, and aromatics are not favored for microbial utilization, and therefore their potential to contribute to MIC is considered low [10, 16-18].

Interestingly, the exact mechanisms contributing to microbial growth in gasoline fueling systems are not completely understood. Some studies have indicated that alcohols facilitate microbial activity, while others suggest that they inhibit microbial growth [13]. Microbial growth and MIC are not considered to be common in gasoline storage systems, even in the bottom water present in the fuel tank. A recent study [13] showed that ethanol and isopropanol, when blended into gasoline, reduced microbial activity in the bottom water. This effect seems counterintuitive, but it is likely that the concentration of either alcohol in the bottom was too high to enable microbe propagation. In summary the ability of the dissolved alcohols to promote, or inhibit MIC, is not clear and further studies are needed to identify the conditions favoring MIC.

2.4 IMPACT OF CONTAMINANTS

Contaminants can have a large impact on corrosivity, especially if the contaminants can disassociate into ions and anions, thereby creating an electrolyte solution. One of the most pervasive and corrosive contaminants is chlorine, which occurs as naturally occurring sodium chloride or can be leached from PVC. Contaminants are believed to be responsible for much of the corrosion observed with methanol and its blends with gasoline. Studies by Lash (19) have shown significant corrosion rates for aluminum and steel in methanol gasoline blends. These results also included a separate water phase. However, the concentrations of acid, chlorine and sulfates present in these fuel mixtures were exceptionally high. In fact, they exceeded acceptable levels by an order of magnitude for fuel grade methanol. Other studies have shown methanol may not be corrosive, depending on contaminant levels and test conditions.

3. METHANOL CORROSION

Methanol has been known to accelerate corrosion in oil and gas extraction systems, including piping. These systems contain high levels of sulfur which participate in the corrosion process. In fact, this type of corrosion is termed sulfide stress cracking and requires the presence of sulfur, in addition to methanol to occur (21). Fuel-grade methanol contains only trace sulfur levels, which are too low to promote sulfide stress corrosion. A study by the Steel Tank Institute showed no measurable corrosion occurred for methanol-water solutions with carbon steel (22-23). Accelerated erosion on engine component, including pistons, has been observed with methanol fuels (including blends with gasoline) (24-25). In these studies, erosion was determined by the increasing Fe content in the lubrication oil over time. However, it is not clear whether the iron was removed by either corrosion or wear. It is our recommendation that further investigations be conducted on bench rigs at elevated temperatures and additional tests focused on surface reactions be conducted.

4. CONCLUSIONS

The fuel candidates, by themselves, are not considered to be corrosive to metals since they are not highly reactive nor dissociate into conductive ions and anions. High levels of contaminants, especially chlorides and sulfates can promote corrosion. However, acceptable levels of these compounds in fuels are too low to cause aggressive corrosion. As such, the only viable pathway toward corrosion of metals and alloys occurs with the presence of a water (aqueous) phase, which exists separate from the fuel. Only the alcohol candidates are water soluble. Because the other fuel types are not soluble, they are not expected to contribute to corrosion. Associated with this aqueous solution are three potential pathways that can contribute to corrosion. They are 1) formation of a differential aeration cell, 2) microbially-induced corrosion, and 3) high levels of electrically conducting contaminants. The ability of differential aeration cells to corrode metals is questionable, and studies suggest that metallic coupling and/or occluded regions are necessary to induce corrosion. Of the alcohols, isobutanol and isopropanol exhibit the highest levels of oxygen solubility, and therefore, have the highest potential (albeit unconfirmed) for producing conditions necessary for corrosion to occur via a differential aeration cell. Microbial-induced corrosion is another possibility. However, even though it has been established that MIC can occur with dissolved alcohols, empirical studies have not been able to establish MIC for alcohols blended with gasoline.

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