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Review

A comprehensive review on the environmental impacts of diesel/biodiesel additives

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ABSTRACT

Biodiesel, in its neat or blended form with petrodiesel, is widely accepted alternative fuel for diesel engines. Although biodiesel is presumably associated with lower CO_2, HC, and PMs emissions, it suffers from its own drawbacks including higher viscosity, lower volatility, lower heating value, and higher NO_x emissions. In order to address these shortcomings and to meet stringent emission norms, diesel/biodiesel additives have attracted more attention recently owing to their ability to improve engine performance and mitigate hazardous emissions. While discrete emissions analysis could provide useful information on environmental impacts associated with various fuel additives, decision-making on such basis would be very difficult or even impossible since different fuel additives may have different positive/negative effects on pollutants generated during combustion process. This issue becomes even more serious in multi-objective optimization studies due to the fact that considering all emission indices for finding a global optimal point would be very complex as a result of conflicting objectives. Moreover, exhaust gas emission analysis does not consider environmental impacts caused in fuel production process. Discrete emissions analysis also lacks weighting in decision-making procedure since the level and degree of harmfulness of pollutants may not be comparable. Life cycle assessment (LCA) has been recognized as a valuable tool to address these challenges through systematical evaluation of potential environmental impacts of fuel additives. Accordingly, this paper was aimed at comprehensively reviewing and mechanistically discussing the effects of various diesel/biodiesel additives including metal-based, oxygenated, antioxidant, cold flow improver, lubricity improver, and cetane number improver additives as well as engine operating parameters like engine load, engine speed, EGR, and injection timing on both regulated and non-regulated emissions. Moreover, the environmental impacts of various diesel/biodiesel additives by incorporating an LCA approach was also critically discussed.

1. Introduction

The continuous growth of global energy demands and the consequent environmental pollution over the last decades have turned into serious concern shared by policy-makers as well as the general public. On the other hand, the conventional resources of energy, such as oil, natural gas, and coal are non-renewable and fear of their depletion in the next century has also been the subject of intense debate, coal might be an exception though. In response to these challenges and to ensure harmonious coexistence of human and environment while sustainable economic growth and development are also achieved, the production and use of renewable energy carriers such as biofuels have been growing in many parts of the world (Figs. S2 and S3) [1]. Among various types of biofuels, liquid biofuels such as bioethanol and biodiesel have been of great prominence as alternatives to their fossil-originated counterparts used in the transportation sector *viz.* gasoline and diesel. On the other hand, it should be noted that the transportation sector, because of fossil fuel consumption, is among the major sources of air

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pollution globally. For instance, this sector accounts for 28% of all US greenhouse gas (GHG) emissions (Fig. S1).

Accordingly, projections indicate that the usage of biofuels will increase from 1.3 million barrels of oil equivalent per day in 2012 to 4.6 million barrels of oil equivalent per day in 2040 [2,3]. Comparatively, the share of petroleum diesel in the transportation sector is expected to increase at various rates in different countries [4] and, therefore, its renewable alternative, i.e., biodiesel is expected to play a major role in the transport fuel demand growth over the next decades.

Among several feasible alternative resources, biodiesel has been at the center of attention in recent years because of its particular advantages [5]. Hence, since the carbon contained in vegetable oil or animal fat are originated mostly from the carbon dioxide (CO_2) present in the air (i.e., biogenic), biodiesel is considered to contribute much less to global warming than its fossil-oriented counterpart (Fig. S4) [6]. This could be of interest in climate change mitigation policies [7]. Unlike petroleum diesel, biodiesel has more oxygen in its chemical formula which is considered as an important property of this alternative fuel [8]. Other technical and environmental benefits attributed to biodiesel compared with petroleum diesel include higher combustion efficiency, lower sulfur and aromatic content, higher cetane number, higher biodegradability, higher flash point, and inherent lubricity [9], while both biodiesel and diesel share similar fuel properties paving the way for smooth replacement of biodiesel in the transportation sector [10]. The important physicochemical properties of biodiesel are tabulated in Table S1.

Biodiesel can be used in neat or blended forms with petroleum diesel. Although biodiesel is perfectly miscible in diesel at any ratios, generally four main volumetric-based blends of biodiesel are available in the fuel market, i.e., B100, B20–B30, B5, and B2 [11–13]. Nevertheless, B5 and B20 are the most common blends as they do not require any engine modifications [14–16]. According to Fig. S5, combustion of biodiesel in compression–ignition engines leads to lower smoke, particulate matter (PM), carbon monoxide (CO), and unburned hydrocarbon (HC) emissions [17,18].

Nevertheless, and in spite of its advantageous features, biodiesel combustion is still associated with some environmental pollutions, to a much smaller extent compared with petroleum diesel though [19,20]. For instance, increased nitrogen oxides (NO_x) emission through biodiesel utilization *vs.* diesel is regarded as a major shortcoming of this alternative fuel [21–27]. On that basis, various techniques have been put forth aiming at mitigating biodiesel-attributed exhaust emissions including engine improvements [28,29], improving fuel performance and emission characteristics by using fuel additives [28], as well as exhaust gas treatment technologies [29]. Among these remedies, the modification of fuel properties to enhance the combustion process would probably be the most feasible one as no engine modifications nor extra equipment would be required. In line with that, different fuel additives including metal-based additives, antioxidants, oxygenated additives, cold flow improvers, etc. have been used in biodiesel and its blends [28–30]. In fact, additives play a valuable and very cost-effective role in reducing potential operational problems and meeting fuel specification requirements, which might otherwise prove challenging or impossible without their use [29]. The choice of additives for biodiesel depends on a number of factors including fuel blending properties, economic feasibility, additive solubility, toxicity, viscosity of the fuel blend, flash point of the fuel blend, solubility of water in the blend, and water partitioning of the additive [31]. Overall, the use of additives could solve some technical problems associated with the use of biodiesel as elaborated earlier [32].

Emission analysis could provide useful information on the environmental impacts associated with various fuel blends containing different additives, however, decision making on the basis of these indices would be very complicated or even in some cases impractical owing to the fact that different fuel additives may impact the generation of different pollutants during the combustion process differently. For example, although biodiesel incorporation into diesel fuel blends could positively reduce CO, HC, and PM emissions, it could also promote NO_x formation. On the other hand, exhaust gas emission analysis does not include the environmental impacts associated with the production process of fuel additives. Life cycle assessment (LCA) approach could be regarded as a promising alternative to overcome the intrinsic shortcomings attributed to the conventional emission analysis. Using this approach, all emission indices of a given fuel blend containing a particular type of additive can be presented on a consolidated basis, i.e., human health, eco system quality, climate change, and resource damage. This could in turn not only assist with the interpretation of data emerging from emission analysis but it could also facilitate multi-objective optimization by consolidating the environmental impacts associated with each fuel blend. It should be noted that LCA has been frequently used to evaluate and compare the environmental, energy, and economic aspects of biodiesel production from different feedstocks around the world. However, to the best of our knowledge, no attempts have been made to review and investigate the environmental impacts of using biodiesel and biodiesel fuel additive by LCA. Therefore, the present review is aimed at comprehensively reviewing the environmental impacts of different fuel additive used in to diesel/biodiesel blends including oxygenated additives such as biodiesel, bioalcohols, metal-based additives, antioxidant additives, cold flow improver additives, lubricity improver additives, and cetane number improver additives, on engine emissions, i.e., PM, NO_x, CO, HC, CO_2, as well as other non-regulated emissions. Moreover, the impacts of these additives from LCA perspective are reviewed and discussed.

2. Diesel/biodiesel additives

2.1. Metal-based additives

Metal-based additives are known as combustion-improving catalyst and have been reported to result in reduced fuel consumption and emissions [33]. The main metal-based additives investigated as combustion improver are platinum (Pt), cerium (Ce), iron (Fe), barium (Ba), cerium–iron (Ce–Fe), calcium, platinum–cerium (Pt–Ce), manganese (Mn) and copper (Cu) [34–42]. The positive impacts of these additives are generally associated with enhancing soot oxidation through two different mechanisms. First, by reacting with water molecules leading to the generation of hydroxyl radicals which in turn oxidize the existing soot and/or second, by acting as catalyst in lowering the temperature required for the oxidation of the carbon atoms present in the soot [43].

2.2. Oxygenated additives

Some oxygenated additives like biodiesel and ethanol are also used in neat form in customized internal combustion engines as fuel. Nevertheless, these are also used in fuel blends as additives [44]. When used in diesel and biodiesel blends, oxygenated additives could affect important properties of the fuel blends, e.g., viscosity, flash point, low temperature properties (such as cloud and pour points), volatility, cetane number, as well as calorific value [45]. Overall, all the favorable attributes of the oxygenated additives such as redaction of smoke emissions are reportedly dependent on their oxygen content as well as on the molecular structure of the fuel blend [46]. Table S2 tabulates the chemical and physical properties of the commonly used oxygenated fuel additives.

Recently, the use of bioalcohols as oxygenated additives has expanded. Bioalcohols have been long used as an oxygenated additive in internal combustion engines (ICEs) due to their potential to reduce GHG and toxic exhaust emissions as well as to enhance energy efficiency [47]. In spite of such advantages, high recovery cost and low production efficiency of bioalcohols during the production stage have been among the economic barriers limiting their use in modern engines in some parts of the world [48–50]. Bioalcohols have also been widely