A ranking scheme for biodiesel underpinned by critical physicochemical properties

S.M.A. Rahman[1,]*, I. M. R. Fattah[2,*], S. Maitra[3], T.M.I. Mahlia[2]

[1] *Biofuel Engine Research Facility, Queensland University of Technology, Brisbane, 4000 Australia*

[2] *School of Information, Systems and Modelling, Faculty of Engineering and IT, University of Technology Sydney, NSW 2007, Australia*

[3] *Data & BI, Engineering Department, SureCash, Dhaka, Bangladesh*

**Corresponding Authors:* s2.rahman@qut.edu.au, rizwanul.buet@gmail.com

Abstract

Diminishing oil reserve, escalating energy dependence, and the environmental impact of fossil fuel utilization has led to research on renewable energy resources with a cleaner carbon footprint. Biofuel, especially biodiesel, has become a feasible substitute for petroleum diesel as it can be directly used in existing transport infrastructure without significant alteration. This paper starts by discussing some critical physicochemical properties and their effect on engine performance and emission. The research then proposes a ranking scheme to select the most suitable biodiesel based on six vital physicochemical properties: density, viscosity, heating value, flash point, cetane number and oxidation stability. The solution developed is independent of supervision, contrary to popular learning algorithms and can operate on the only intelligence whether an attribute is favourable by its higher/lower values. The novelty of the work consists in ensuring that the rarer properties pick up the greater weights and in establishing a simple ranker based on descriptive statistics. This scheme first generates transactions against each biodiesel which helps in association rule mining, which is later used to score/rank the biodiesels. The three phases and their subordinate sub-steps have been carried out using the platforms: Python, R and Tableau, respectively. The study endorses *Brassica juncea*, Cardoon (*Cynara carduncul*u), and poppyseed oil as the most desirable biodiesel feedstocks. On the other hand, cedar, castor and hiptage were ranked as least desirable in the list of 71 feedstocks based on the proposed ranking scheme. The proposed ranking scheme will help decision-makers such to analyze and obtain tailored biodiesel feedstock for their purposes.

Keywords: Biodiesel; Physicochemical properties; Biodiesel Ranking; Engine emission; Engine performance.

1. Introduction

Almost all the countries of the world are profoundly dependent on the transport sector for economic growth. It is necessary to reduce the dependence on vulnerable petro-diesel imports to sustain growth. For example, Australia significantly depends on crude oil imports for its transport and energy sector. As a member of the International Energy Agency (IEA)as well as an oil importer, Australia is required to abide by the IEA program treaty which dictates "to hold oil stocks equivalent to at least 90 days of their prior year's daily net oil imports", a benchmark which was set in 1974 [1]. However, since March 2012, Australia has been in non-compliance with the 90-day stockholding obligation [2, 3]. In February 2020, Australia only had 81-days worth of oil reserve [4]. The global pandemic created by COVID-19 brought down international transportation due to restrictions put by various countries all over the world which limited the export loads, and extra port operation delays exposing the country's vulnerability of over-dependence on liquid fuel of other countries.

On the other hand, this pandemic also delineated the importance of a healthy environment with less air pollution for the betterment of human life [5]. Ambient air pollutants are risk factors for respiratory infection as the microorganisms carried by these are highly invasive to humans which affects the body's immune system making people more susceptible to pathogens [6]. A recent study has found a significant connection between the mortality rate of COVID-19 patients and long term air pollution- higher levels of the small particles in the air ($PM_{2.5}$) were associated with higher COVID-19 related death rates [7]. Another study by Mofijur *et al.* [8] reported similar findings that the Air Quality Index (AQI) had a strong influence on the cumulative cases of COVID-19 in Dhaka city.

Vehicle emissions are one of the significant sources of air pollution globally. It is reported that vehicle emissions account for 65% of urban air pollution and compared to road accident fatalities, Australia has higher air pollution death tolls [9]. There are strict emission regulations imposed on a global basis to reduce transport-related to greenhouse gas emissions. Depending on the vehicle technology, emissions include fine particulate matter (PM) which consists of black carbon (BC) and sub-micron primary organic aerosol (POA), and reactive gases. An example of reactive gases is nitrogen oxides (NOx) [10]. NOx emission is associated with ozone (O_3) formation. Fine PM, NOx emitted from combustion processes severely affect human health. Fine PM can damage lung tissues and the brain by penetrating deep into the human body [11-13]. Thus, it is vital to find alternative fuel sources readily available to fuel the transport and energy sector.

Biodiesel is one of the potential alternative liquid fuels, which is readily available, eco-friendly, non-toxic, has technically and economically feasible production process and functional properties similar to those of petro-diesel [14-16]. Biodiesel is defined as mono-alkyl esters of long-chain fatty acids derived from various feedstocks, also known as triacylglycerides (TAGs), or more simply, triglycerides [17, 18]. Biodiesel can be obtained from various resources which can be sub-divided into four types: edible vegetable oil, non-edible vegetable oil, animal fats and other sources (including algal, waste or recycled oil) [19-23]. Edible and non-edible feedstocks are also referred to as first-generation and second-generation biodiesel feedstocks [24, 25]. The disadvantages of first-generation biodiesels are using edible oil as fuel which creates a problem with food supply. Other issues related to the use of edible oils are high cost, limited cultivation area and adaptability issues due to environmental conditions [26]. On the contrary, second-generation biodiesel feedstocks have become popular as they do not contribute to food shortages. Non-edible feedstocks have the following advantages over edible feedstock: less production cost, reduced farmland requirement, and it eradicates food inequality [27, 28]. Third-generation biodiesels are those produced from microalgae [29, 30]. The significant benefits of biodiesels produced from microalgae are reduced greenhouse effect, elevated productivity, small farming land requirement, higher oil percentage and almost zero-impact on food supply. However, the provision of substantial investment, the necessity of sunlight, large scale production issues and difficulties associated with oil extraction are some critical drawbacks of third-generation biodiesels [31, 32]. The conventional process for biodiesel production is transesterification or alcoholysis, in which the triglycerides are reacted with alcohols in the presence of a suitable catalyst (homogeneous/heterogeneous/bio/nano), as a reaction promoter, to produce fatty acid alkyl esters [33]. Table 1 shows some of the feedstocks for biodiesel production.

Table 1. Primary feedstocks for biodiesel production [34-37]

Edible oils	Non-edible oils	Animal fats	Other sources
Soybeans (*Glycine max*)	*Jatropha curcas*	Pork lard	Bacteria
Rapeseed (*Brassica napus L*)	Mahua (*Madhuca indica*)	Beef tallow	Fungi
Rice bran oil (*Oryza sativum*)	Pongame oil tree (*Pongamia pinnata*)	Poultry fat	Algae (Cyanobacteria)
Barley	Camelina (*Camelina Sativa*)	Fish oil	Microalgae
Sesame (*Sesamum indicum L.*)	Cottonseed (*Gossypium hirsutum*)	Chicken fat	Tarpenes
Groundnut	Karanja or honge		Poplar
Sorghum	Neem (*Azadirachta indica*)		Switchgrass
Wheat	Jojoba (*Simmondsia chinensis*)		Miscanthus
Corn	Passion seed (*Passiflora edulis*)		Latexes
Coconut	Moringa (*Moringa oleifera*)		Fungi
Canola	Coffee ground (*Coffea arabica*)		Recycled oil
Peanut	Nagchampa (*Calophyllum inophyllum*)		
Palm and palm kernel *(Elaeis guineensis*)	*Croton megalocarpus*		
Sunflower (*Helianthus annuus*)			

Anwar [38] studied the 16 most popular biodiesel feedstocks for screening the best feedstock for biodiesel production. He focussed on fifteen economic, technical and environmental aspects of the selection process. Four different multi-criteria decision analysis (MCDA) methods, namely Preference Ranking Organization Method for Enrichment Evaluation (PROMETHEE)-Graphical Analysis for Interactive Assistance (GAIA), weighted sum method (WSM), weighted product method (WPM) and Technique for Order Preference by Similarity to Ideal Solution (TOPSIS) with five weighting methods in percentage, namely EQUAL, CRITIC, ENTROPY, Analytical hierarchical process (AHP), and Fuzzy Analytical Hierarchical Process (FAHP) were used to rank the feedstocks. The study found that coconut was ranked the best and soybean was the worst feedstock for biodiesel production. Anwar et al. [39] ranked six non-edible biodiesels using twelve physicochemical properties as criteria for ranking the above biodiesels. Four different MCDA methods, namely PROMETHEE-GAIA, WSM, WPM, and TOPSIS, were used for the analysis. Based on the research, tone fruit kernel oil biodiesel was ranked the best and waste cooking oil biodiesel was the worst performer in the rank. Kamoun et al. [40] used the MCDA approach to select and rank the most suitable growth media and strain for biodiesel production. They applied the PROMETHEE-GAIA analysis algorithm containing a predicted data set of chemical and physical properties of fuel derived fatty acid methyl ester (FAME). Based on the analysis, the nature of lipids produced by *Mucor circinelloides* grown on soapstock of refined soybean oil, soapstock of refined olive pomace oil and glucose were found to be suitable for biodiesel production. Ahmad et al. [41] studied the biodiesel production potential of several oleaginous micororganisms such as microalgae (*C. protothecoides* and *C. zofingiensis*), yeasts

(*Cryptococcus albidus* and *Rhodotorula mucilaginosa*), and fungi (*Aspergillus oryzae* and *Mucor plumbeus*) using MCDA apporach. They used analytic hierarchy process (AHP) and PROMETHEE-GAIA on the basis of different criteria *viz*., oil concentration, content, substrate consumption rate, production rate and yield, fatty acids composition, biomass harvesting and nutrient costs etc. to rank the preferred microorganisms. Based on the analysis they found *A. oryzae* and *M. plumbeus* were the best performers.

In the present study, several critical physicochemical properties were discussed to facilitate in understanding the effect of those properties in engine performance and emission characteristics, discussed in the next section. Those chosen properties were later used to rank biodiesel using association rules mining. A new scheme is proposed for selecting biodiesels that is free from dependence on prior experiences. The method does not rely on refining parameters using any sophisticated learning algorithm but rather, overcomes the limitations of such algorithms' hunger for feeding on prior intelligence. This scheme first generates transaction against each biodiesel which helps in association rules mining, which is later used to score/rank the biodiesels.

2. Critical Physicochemical Properties

The characterization of fuel properties determines the quality of fuel [42, 43]. The key fuel properties of biodiesel fuels are density, viscosity, flash point, heating value, oxidation stability and cetane number.

2.1. Density

Density is one of the critical properties of fuel as it affects engine performance, combustion quality as well as other properties such as cetane number and viscosity [44-48]. ASTM standard D1298 and EN ISO 1676 methods are used generally to measure density [25]. An increase in density generally increases fuel droplet size and affects combustion quality [49]. On the other hand, reduced density expands the efficiency of atomization and air-fuel ratio formation. Furthermore, a higher density of fuel also affects engine emissions [50]. High fuel density increases emissions such as particulate matter (PM) and NOx emission [51, 52]. From Table 2, the density varies for biodiesels, i.e. Jojoba biodiesel has the lowest density (832 kg/m^3) while Castor biodiesel has the highest density (928.5 kg/m^3).

2.2. Kinematic Viscosity

Similar to density, kinematic viscosity is a vital parameter for fuel as it governs fuel atomization, spray characteristics and combustion quality [53, 54]. Biodiesel quality, while stored, can be checked by measuring the kinematic viscosity. Lower kinematic viscosity results in insufficient lubrication and increases wear and tear [55]. On the other hand, high fuel viscosity may form large droplets at the injection, which deteriorates combustion quality and result in higher exhaust emission. In most cases, biodiesel has higher kinematic viscosity compared to that of petro-diesel [56, 57]. According to the ASTM D6751 and EN14214, the kinematic of biodiesel should adhere to 1.9 – 6 mm^2/s and 3.5 – 5 mm^2/s, respectively [58].

2.3. Heating Value

Another critical parameter is the heating value. The amount of energy released when a known volume of the fuel is completely combusted represents fuel energy content [25]. A high heating value is always desired as it favours heat release during combustion and thus improves engine performance [52, 59, 60]. However, compared to diesel fuel, most biodiesel has a significantly lower heating value which can be associated with the fuel bound oxygen content of biodiesel [25]. From Table 2, biodiesel produced from waste fish oil, *Xanthoceras sorbifolium*, camelina, and jojoba oil was reported to have a higher heating value than that of diesel fuel.

2.4. Flash Point

Flash point refers to the flammability of fuel and is a safety indication for storage, handling, and transportation [61]. When the fuel is heated, it is the minimum temperature where the fuel will give off adequate vapours to form a combustible mixture above the fuel surface. Biodiesel has a significantly higher flash point compare to pure diesel and thus is considered safe for storage, handling and transportation [62-64]. The USA and EU standard suggests that the flash point for biodiesel should be at least 93 °C and 120 °C, respectively. The flashpoint of biodiesel is impacted by residual alcohol content [65]. The flashpoint is governed by the chemical compositions of biodiesel such as a number of double bonds, the total number of carbon atoms, etc.

2.5. Cetane Number

Cetane number (CN) governs the ignition quality of fuel and is a measurement of ignition timing inside the combustion chamber [66]. In general, biodiesel has a higher CN compared to that of pure diesel. Higher CN generally indicates shorter ignition delay and earlier combustion, which helps the engine run smoothly [67, 68]. Lower CN results in a delayed ignition and tends to increase HC and PM emissions [69]. In general, the CN is associated with saturation levels of biodiesel, and a higher saturation level will have a higher CN, while higher unsaturation levels result in lower CN [70, 71].

2.6. Oxidation Stability

Oxidation stability represents the biodiesel's ability to maintain the fatty acid composition during extended storage without degradation [72, 73]. Compared to petroleum diesel, biodiesel is less resistant to oxidation [74]. Oxidation degradation generates oxidation products which might compromise fuel properties, worsen fuel quality and thus will deteriorate engine performance [75]. Oxidation stability is an important factor because:

- Sediments and gums may form in equipment during use
- Impacts critical fuel properties, e.g., viscosity and cetane number
- Affects engine exhaust emissions
- Promote corrosion, affects component operation due to deposits, varnishes and sediments accumulation on engine parts
- Oxidation products can attack elastomers, clog fuel filters and infect engine lubricating oil.
- May increase engine wear due to corrosive acids and deposits

Biodiesel oxidation stability is affected by fatty acid composition, specifically by the degree of fatty acid unsaturation. Also, impurities such as metals, FFAs, additives and antioxidants strongly affect stability. Oxidation of biodiesel can only be delayed and not wholly prevented. Table 2 shows the critical physicochemical properties of most reported 71 feedstock based biodiesel.

Table 2: Key physicochemical properties of biodiesels

No.	Ref	Biodiesel	Density at 15 °C kg/m^3	Kinematic viscosity At 40 °C mm^2/s	Heating value MJ/kg	Flash point □C	Cetane Number	Oxidation stability hr
1.	[76]	*Cascabela ovata*	866.8	4.98	-	93	-	-
2.	[77]	Cedar	924	6.1	39.49	58	-	-
3.	[78, 79]	*Crambe abyssinica*	872	6	39.56	136	-	-
4.	[77]	Cypress	912	2.7	39.22	67	-	-
5.	[80]	Dairy Scum	890	5.35	38.94	195	-	-
6.	[81]	Dairy washed milkscum	875	3.8	40.12	151	-	-
7.	[82]	Fish waste	890	5.3	38.1	-	-	-
8.	[83]	Hiptage	837	16.3	33.12	49.33	-	-
9.	[82, 84]	Marine fish oil	860	4.4	41.37	-	-	-
10.	[77]	Red pine	875	1.4	41.42	44	-	-
11.	[78, 85]	Spirulina	860	5.66	41.36	130	-	-
12.	[86]	Waste frying oil	886	4.71	-	169	-	-
13.	[77]	White pine	888	1.6	41.67	48	-	-
14.	[78, 87-89]	Linseed	852	3.95	37.45	241	34.6	1.5
15.	[90, 91]	*Citrus sinensis*	900	3.79	35.47	190	37	-
16.	[92]	*Styrax officinalis L.*	886	3.57	39.023	175	40.47	2.69
17.	[93]	Waste fish oil	875	4.14	51.5	169	41	-
18.	[78, 94, 95]	Bitter almond	884	4.60	41.76	169	45.18	-
19.	[78, 96]	Fish oil	881	4.45	40.54	177	47	-
20.	[97, 98]	*X. sorbifolium*	881.6	4.67	48.56	121.55	47.3	2.53
21.	[99, 100]	Argemone	837	4.38	37.5	193	47.5	1.44
22.	[78, 101-104]	Chicken fat	883	5.14	40.17	175	48	6.46
23.	[78, 105-107]	Cotton Seed	897	4.19	39.75	210	48.1	1.88
24.	[108]	*Tetradesmus obliquus*	-	3.88	39.44	-	48.15	-

25.	[39]	Pappaya Seed	840	3.53	38.49	112	48.29	5.61
26.	[78, 109, 110]	Camelina	885	4.11	45.2	150	48.91	-
27.	[80, 111]	Simarouba Glauca	864	5.48	38.41	175	49.1	-
28.	[78, 107, 112-114]	Sunflower	865.5	4.39	37.5	178	50	0.9
29.	[78, 115]	Michelia champaca	870	5.11	39.51	158	50.28	-
30.	[39]	Stone fruit kernel	855	4.26	39.64	105	50.45	7.15
31.	[78, 107, 116-120]	Soybean	882	4.37	35.74	170	51	1.5
32.	[78, 114, 121-123]	Neem	873	6.1	40	144.75	51.26	2.13
33.	[78, 124, 125]	Tobacco	865	3.56	42.22	165	51.5	-
34.	[78, 114, 126-132]	Castor	928.5	15.55	38.2	188.9	51.9	7
35.	[133]	Waste edible oil	877	4.43	39	-	52	-
36.	[78, 107, 134-136]	Olive	871	4.32	39.96	169.5	52	4.29
37.	[137, 138]	*Chlorella protothecoides*	900	4.22	40.04	124	52	12
38.	[39, 78, 107, 117, 139-141]	Rapeseed	880.5	4.47	37.42	167	52.4	4.5
39.	[39, 78, 82, 142-145]	Waste cooking oil	876	4.13	38.25	185	52.5	0.47
40.	[78, 117, 146, 147]	Rubber	875	5.6	39.17	173.4	53	0.8
41.	[107, 148]	Safflower	885	4.6	38.12	180	53	2.4
42.	[39, 78, 83, 149-152]	Jatropha	872	4.39	38.52	147	53.8	3.01
43.	[153]	Waste eggshell	890	4.98	42.18	180	54	-
44.	[78, 107, 116, 154-156]	Canola oil	878	4.42	39.49	172	54	1.9
45.	[39]	Beauty leaf oil	868.7	5.68	39.38	141.5	54	3.58
46.	[157-159]	Coffee	857.5	5.43	39.43	242	54.55	6.5
47.	[160-163]	Chlorella vulgaris	895	4.1	42.7	163	54.7	5
48.	[78, 164-166]	Mahua	895	4.77	16.9	129.5	55	8.2
49.	[107, 167]	Corn	878	4.42	39.93	172	56	1.3
50.	[78, 157, 168-170]	Mustard (*Brassica Juncea*)	879	5.53	40.4	169	56	16
51.	[171-174]	*Calophyllum inophyllum*	880.7	4.68	38.02	150.8	56.3	3.58
52.	[78, 175-178]	Karanja	889	4.79	36.56	157.4	56.55	2.65
53.	[179, 180]	Date seed oil	878	4.85	41	107	57.1	-

54.	[78, 181, 182]	Terminalia catappa	876	4.3	37.73	90	57.1	32.5
55.	[157]	Cardoon (*Cynara cardunculus*)	880	9.5	40.69	309	57.4	12
56.	[78, 183-185]	Kusum	875	4.34	39.45	152	57.86	6.5
57.	[186]	Tannery waste	870	4.6	-	150	58	-
58.	[187]	Poppy seed oil	896	4.58	39.99	203	55.5	7.81
59.	[78, 188]	Peanut	878	4.69	35.44	176	58.24	5
60.	[78, 116, 189, 190]	Palm	871.9	4.58	36.97	169.7	58.6	10.5
61.	[78, 88, 191, 192]	Sesame	867	4.23	40.25	176.7	58.97	6.25
62.	[107]	Algae	881	4.55	-	140	59	2.3
63.	[78, 193]	Groundnut	920	4.4	39.8	132	59.85	-
64.	[78, 194, 195]	Beef tallow	832	4.89	40.23	152	60.36	1.99
65.	[80, 196, 197]	Yellow Oleander	886	5.81	37.89	160	61.5	6.5
66.	[78, 107, 191, 198]	Hazelnut	896	4.81	39.58	173	63	2.1
67.	[78, 199]	Babassu	872	4.2	31.8	117	63.25	-
68.	[78, 200-202]	Jojoba	866	19.2	44.77	80.5	63.5	-
69.	[78, 203]	Animal fat	875	4.25	36.73	-	63.88	-
70.	[15, 78, 204, 205]	Coconut	867	3.2	35.2	114	64.65	5.12
71.	[78, 206-209]	Rice bran	889	5.15	38.17	161	64.95	1.7

Fig. 1 shows that most of the biodiesel selected for this study adheres to biodiesel standards (ASTM- Kinematic Viscosity, Flash Point, Cetane Number, EN- Density). The standards do not specify a minimum heating value requirement for biodiesels; however, the higher, the better [30].

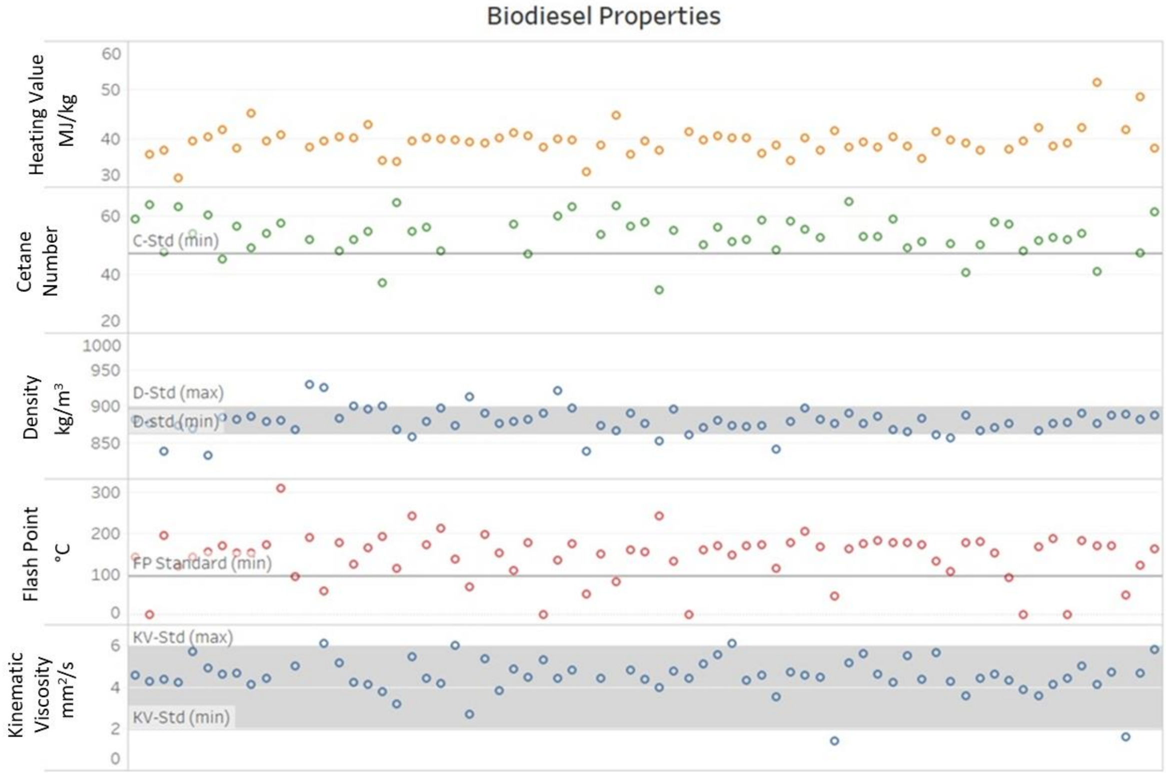

Fig. 1. Selected biodiesel properties. The range of the property is specified in the shaded area. The heating value range is not specified in standards. The only minimum value is specified for flash point and cetane number.

3. Impact of Chosen Properties on Engine Performance and Emission

Several studies have been conducted which explored engine performance and emissions parameters of diesel engine operated with biodiesel. In general, the heating value of biodiesel is lower compared to diesel fuel. As a result, the use of biodiesel and its blends results in a decrease in engine performance. Yoon *et al.* [210] reported that Palm biodiesel blends exhibited higher brake specific fuel consumption (BSFC) (+3.7 to +5.8%) compared to diesel fuel which is attributed to higher viscosity, density and lower heating value. Higher BSFC for Palm biodiesel blends was also reported by Sanjid *et al.* [211]. Patel *et al.* [212] reported a reduction of thermal efficiency for soybean biodiesel blends which can be associated with inferior spray atomization due to higher viscosity. 3.8% increase in BSFC for Soybean biodiesel and its blends. Also, the use of Karanja biodiesel and its blends increases BSFC [213, 214]. Another study reported that waste cooking oil increased BSFC by 14.34% [215]. Poor atomization and lower heating value compared to diesel fuel are the reason behind increased BSFC and decreased brake thermal efficiency (BTE) [216]. Due to having lower heating value biodiesel and its blends releases less heat during combustion and thus to provide the same amount of power needs more fuel to be injected, thus, increases BSFC [61]. Contrary, the study reported that biodiesel decreases BSFC and increases BTE [217]. They attributed the decrased BSFC to the presence of oxygen in the blend and higher cetane number of the fuel, both of which aid in better combustion reducing BSFC.

Biodiesel is a clean-burning alternative fuel which is produced from renewable sources. It has a strong beneficial effect on carbon monoxide (CO), hydrocarbon (HC) and particulate matter (PM); however, it negatively affects NOx emission. Biodiesel contains about 11% oxygen in the chemical structure compared to diesel which allows more carbon molecules to burn and results in complete fuel combustion. Thus, CO and HC emissions are lower when diesel engines burn biodiesel fuel. Also, the cetane number affects HC and CO emission. The higher cetane number of biodiesel compared to diesel reduces combustion delay and thus decreases emission. Also, the lower volatility of biodiesel may be responsible for lower CO and HC emission [218]. A study reported emission reduction of 18.4% and 42.5% of CO and HC, respectively, in the case of soybean biodiesel blends [219]. Jatropha biodiesel blend reduced CO and HC emissions by 14 and 11% respectively [220]. Also, several papers stated that Karanja biodiesel also reduced HC and CO emissions [213, 214]. *Calophyllum* biodiesel blends reduced HC emission by almost 6.84% compared to diesel fuel [221]. However, biodiesel increases NOx emission. A study reported that Jatropha biodiesel and its blends increased emission by 7% compared to diesel fuel [220]. Karanja biodiesel increased emission by 11.2-17.62% compared to diesel fuel [219, 222]. *Calophyllum* biodiesel increased NOx emission by 17.87-22.5% compared to diesel fuel [221]. However, some authors reported an increase of CO emission for biodiesel blends which can be associated with poor combustion quality [223, 224], some authors reported reduced NOx emission due to higher cetane number of biodiesel [225, 226].

4. Biodiesel Ranking

A blend of biodiesel and petro-diesel (without/with additives) may be beneficial for improved fuel property and lubricity, enhanced engine performance, lesser pollutant emissions and greater energy security for the future [227-230]. There being different sources, a legit question may be which biodiesel should be blended with petrodiesel for optimal performance of a diesel engine. As an attempt to aid in making such a choice, this paper puts forth a ranker (**Fig. 2**) designed to take into consideration six attributes: density (in kg/m^3), viscosity (mm^2/s), heating value (MJ/kg), flash point ($^\circ$C), cetane number and oxidation stability.

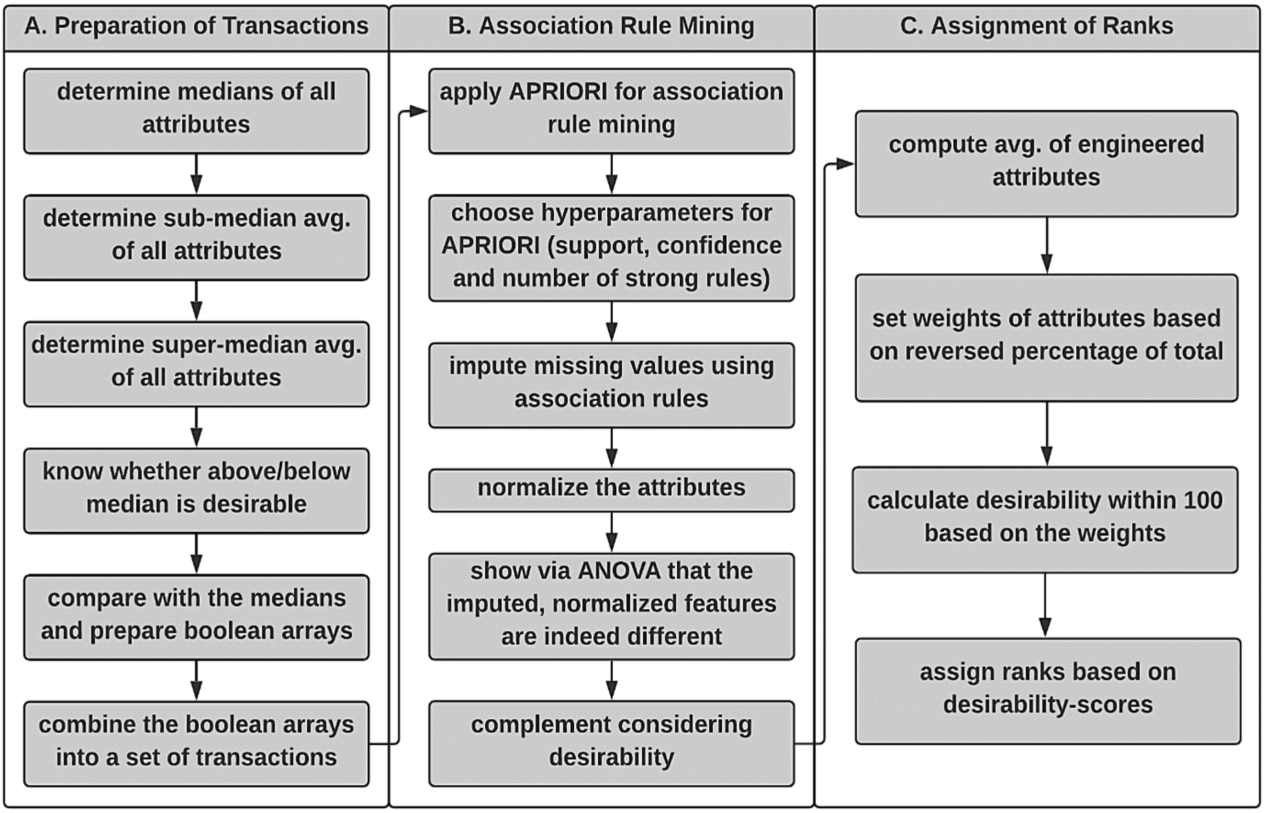

Fig. 2: A flowchart showing the process of ranking biodiesels in three super-steps (A, B, C)[1]

4.1. Preparation of Transactions

This extensive literature review endeavours to extract numerical values of the six separate physicochemical properties of 71 distinct biodiesels. The scrutinized biodiesels have been passed through three phases to assign each of them a rank. The first super-step has the objective of preparing a transaction against each biodiesel. This is for mining association rules to infer values in place of the ones missing. The knowledge that lower values in density

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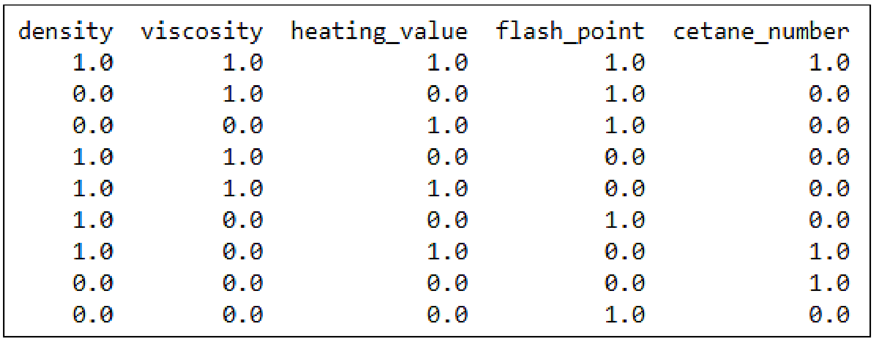
[1] The terms: *sub-median* and *super-median*, are referred to as *lower-median* and *upper-median*, respectively at some places. The two terms refer to the portions less than and greater than the median, respectively.

and viscosity; while upper values in heating value, oxidation stability, cetane number and flash point indicate goodness, helped design the Boolean transactions. This involved two steps:

- Comparison of each attribute with its median put into a boolean array, evaluating to 1 in the case above or equal to the median and 0 in the case below the median (and vice-versa for properties showing goodness with diminishing values). The statistic: median has been appropriate because of its capability to divide any sample into two equal tails.

- Building of transactions against each biofuel, with the highs indicating desirability and the lows, a comparatively less desirable behaviour (**Fig. 3**). From a universal set of 71 transactions against a total of 71 biofuels, only a subset of complete cases are chosen for mining association rules. Since the rules thus generated would be used to fill in missing desirabilities, this exclusion makes considerable sense.



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density  viscosity  heating_value  flash_point  cetane_number
    1.0        1.0            1.0          1.0            1.0
    0.0        1.0            0.0          1.0            0.0
    0.0        0.0            1.0          1.0            0.0
    1.0        1.0            0.0          0.0            0.0
    1.0        1.0            1.0          0.0            0.0
    1.0        0.0            0.0          1.0            0.0
    1.0        0.0            1.0          0.0            1.0
    0.0        0.0            0.0          0.0            1.0
    0.0        0.0            0.0          1.0            0.0
```

Fig. 3: A subset of boolean transactions itemizing 5 of the properties, representative of a single biofuel each

4.2. Association Rule Mining

The second super-step involves mining the association rules using the APRIORI algorithm. The algorithm explores the tendencies of helpful behaviours being found together. Hence if a desirable property is frequently found with another with high confidence, we infer an optimistic value for the missing property. The algorithm (**Fig. 4**) has been tuned to bring out the closest associations covering all the attributes. Lower support (>= 0.1) and higher confidence (>= 0.8) brought to the fore top-25 rules. To infer the missing values, we look at the generated rules (**Fig. 5**) and fill in values using the upper-median and lower-median means calculated in A. For example, the first 3 generated rules in **Fig. 5** show that helpful oxidation stability is frequently coupled with desirable viscosity, heating value and density; a claim supported highly (0.44 to 0.55) with full confidence (1.00). Again, the rules: 7, 10, 12, 17, 19 in **Fig. 5** reveal certain conditions based on which we can infer favourable values for density, i.e., lower_mean_densiy in **Fig. 6**. On the contrary, the absence of these can be indicative of less desirable attributes, i.e., upper_mean_density (**Fig. 6**). After completing the dataset we normalize the values for an ANOVA-test, with an objective of showing that the imputations have indeed maintained the segregation of the distributions of the attributes and the imputation is thus valid (**Fig. 7**).

Under such circumstances, a clarification on the definitions of the terminology used here—with proper justification—may be helpful. Below are the distinctions of the terms with contextual insights:

Properties: Properties are the qualities of biofuels from different perspectives. In the context of this study, the raw data generated by reviewing literature indicates these dimensions, which can be quantified by numerics coupled with certain units.

Features: Properties, when used for any mathematical modelling—such as: applying learning algorithms, applying inferential or descriptive statistics—are called features. Contextually, the six properties can be termed as features for applying ANOVA, calculating scores and ranking.

Attributes: A property, when quantified with numeric values, is said to be an attribute. This concept hails from Database Management Systems (DBMS). In the context of this paper, the six features are furnished with fuel-specific numeric or null attributes.

Treatments: For a statistical ANOVA, multiple groups are formed, and each is served with a different application of any single matter of experimental interest. These are called different treatments. In our context, the different features (X) are causing different attributes (Y) and are playing this role.

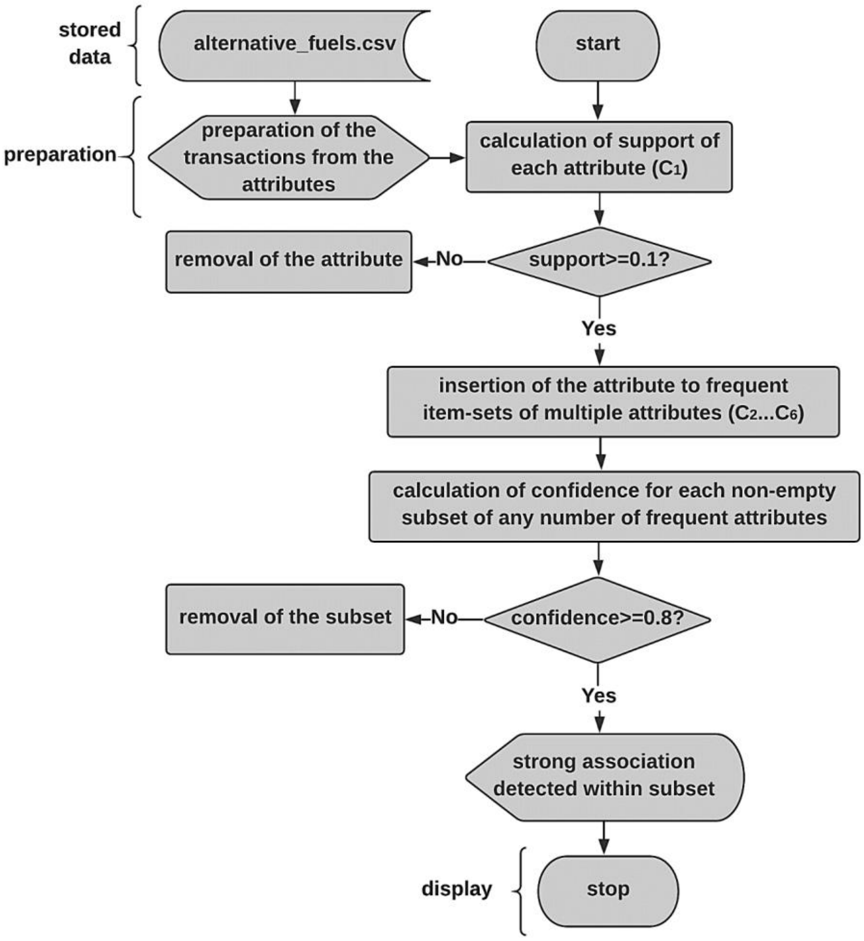
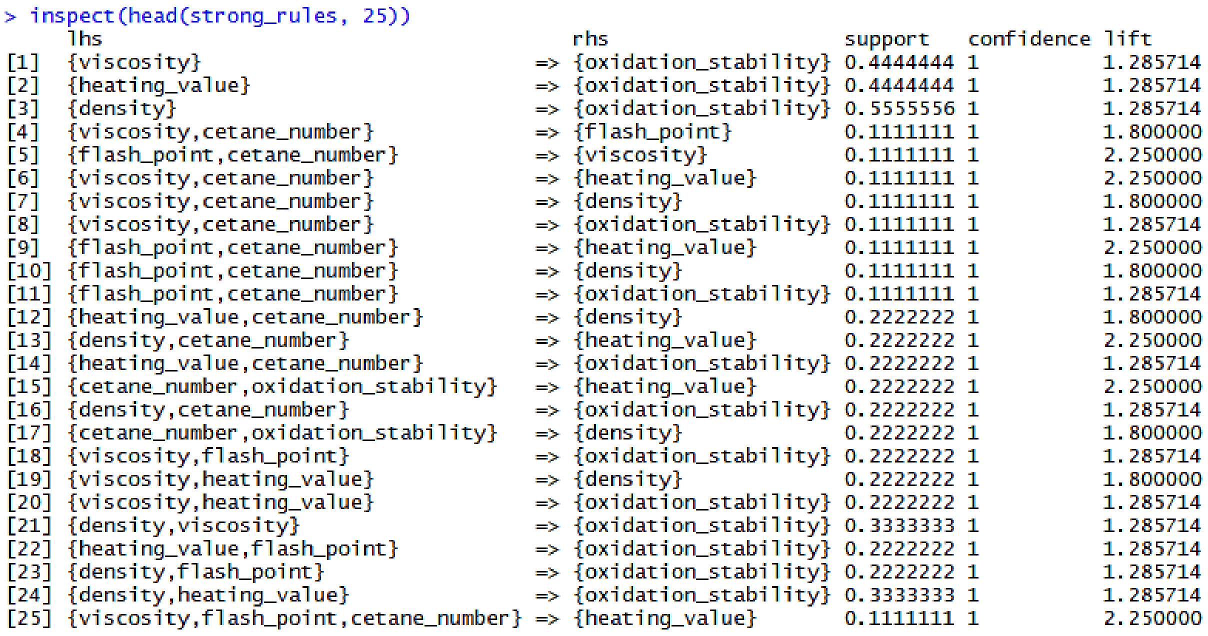
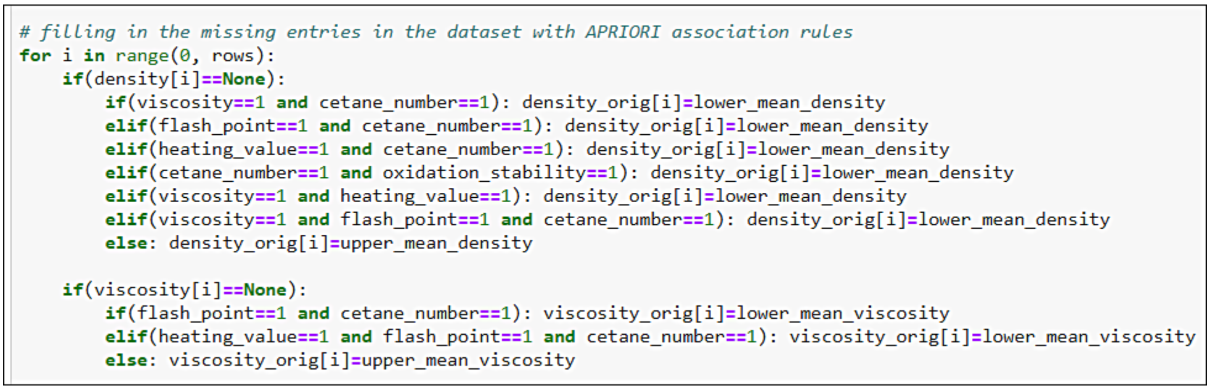


Fig. 4: The APRIORI algorithm, hyper parametrically-tuned for exploring associations



```
> inspect(head(strong_rules, 25))
     lhs                                        rhs                     support   confidence lift
[1]  {viscosity}                             => {oxidation_stability} 0.4444444 1          1.285714
[2]  {heating_value}                         => {oxidation_stability} 0.4444444 1          1.285714
[3]  {density}                               => {oxidation_stability} 0.5555556 1          1.285714
[4]  {viscosity,cetane_number}               => {flash_point}         0.1111111 1          1.800000
[5]  {flash_point,cetane_number}             => {viscosity}           0.1111111 1          2.250000
[6]  {viscosity,cetane_number}               => {heating_value}       0.1111111 1          2.250000
[7]  {viscosity,cetane_number}               => {density}             0.1111111 1          1.800000
[8]  {viscosity,cetane_number}               => {oxidation_stability} 0.1111111 1          1.285714
[9]  {flash_point,cetane_number}             => {heating_value}       0.1111111 1          2.250000
[10] {flash_point,cetane_number}             => {density}             0.1111111 1          1.800000
[11] {flash_point,cetane_number}             => {oxidation_stability} 0.1111111 1          1.285714
[12] {heating_value,cetane_number}           => {density}             0.2222222 1          1.800000
[13] {density,cetane_number}                 => {heating_value}       0.2222222 1          2.250000
[14] {heating_value,cetane_number}           => {oxidation_stability} 0.2222222 1          1.285714
[15] {cetane_number,oxidation_stability}     => {heating_value}       0.2222222 1          2.250000
[16] {density,cetane_number}                 => {oxidation_stability} 0.2222222 1          1.285714
[17] {cetane_number,oxidation_stability}     => {density}             0.2222222 1          1.800000
[18] {viscosity,flash_point}                 => {oxidation_stability} 0.2222222 1          1.285714
[19] {viscosity,heating_value}               => {density}             0.2222222 1          1.800000
[20] {viscosity,heating_value}               => {oxidation_stability} 0.2222222 1          1.285714
[21] {density,viscosity}                     => {oxidation_stability} 0.3333333 1          1.285714
[22] {heating_value,flash_point}             => {oxidation_stability} 0.2222222 1          1.285714
[23] {density,flash_point}                   => {oxidation_stability} 0.2222222 1          1.285714
[24] {density,heating_value}                 => {oxidation_stability} 0.3333333 1          1.285714
[25] {viscosity,flash_point,cetane_number}   => {heating_value}       0.1111111 1          2.250000
```

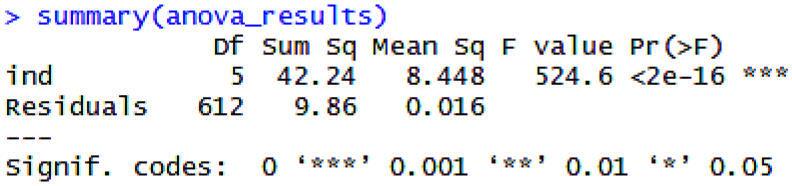
Fig. 5: Strong rules output by ARIORI for imputing blank values



```
# filling in the missing entries in the dataset with APRIORI association rules
for i in range(0, rows):
    if(density[i]==None):
        if(viscosity==1 and cetane_number==1): density_orig[i]=lower_mean_density
        elif(flash_point==1 and cetane_number==1): density_orig[i]=lower_mean_density
        elif(heating_value==1 and cetane_number==1): density_orig[i]=lower_mean_density
        elif(cetane_number==1 and oxidation_stability==1): density_orig[i]=lower_mean_density
        elif(viscosity==1 and heating_value==1): density_orig[i]=lower_mean_density
        elif(viscosity==1 and flash_point==1 and cetane_number==1): density_orig[i]=lower_mean_density
        else: density_orig[i]=upper_mean_density

    if(viscosity[i]==None):
        if(flash_point==1 and cetane_number==1): viscosity_orig[i]=lower_mean_viscosity
        elif(heating_value==1 and flash_point==1 and cetane_number==1): viscosity_orig[i]=lower_mean_viscosity
        else: viscosity_orig[i]=upper_mean_viscosity
```

Fig. 6: Imputation of missing attributes based on ARRIORI-rules (Shown for two properties)



```
> summary(anova_results)
             Df Sum Sq Mean Sq F value Pr(>F)    
ind           5  42.24   8.448   524.6 <2e-16 ***
Residuals   612   9.86   0.016                   
---
Signif. codes:  0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1 ‘ ’ 1
```

Fig. 7: ANOVA-test results verifying the disparate distribution of features after imputation

One-way ANOVA worked on numerical response data (Y)—in our case, different numeric attributes (Y) for individual biodiesels against different features (X). The six quality-metrics created six different groups, and at any point in time, we assume a single feature (treatment) to be different in different biodiesels. ANOVA analyzed if there existed any significant difference among the responses of the biodiesels to a single quality-parameter.

There exists a total of 71 examples for all a = 6—i.e., a_1, a_2, a_3, a_4, a_5, a_6—groups combined. The null hypothesis, H_0 is that all the groups produce the same average numeric response upon a single quality-indicator. Let, n is the count of examples per quality-feature.

- First, determine the mean within each group,

$$\overline{Y} = \frac{1}{n}\sum_j Y_{ij} \qquad (1)$$

where *i* indexes over *a* and *j* indexes over *n*.

- Second, calculate the grand mean, $\overline{Y} = \frac{\sum_i \overline{Y_i}}{a}$ (2)

- Third, determine the between-group sum of squared differences:

$$S_B = \sum_i^n (\overline{Y_i} - \overline{Y})^2 \qquad (3)$$

and divide it by the between-group degrees of freedom, $f_b = (a - 1)$ to find the between-group mean square value, MS_B

- Fourth, determine the within-group sum of squared differences:

$$\sum_{j}\sum_{i}(Y_{ij} - Y_i)^2$$

and divide it by the within-group degrees of freedom, $f_W = a(n-1)$ to find the within-group mean square value, MS_W

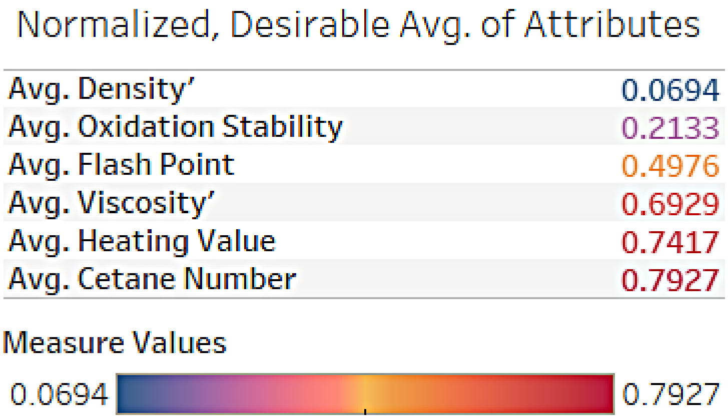
- Finally, compute

$$F = \frac{MS_B}{MS_W} \qquad (4)$$

If the F-ratio surpasses the F-ratio for a threshold p-value = 0.05, then reject the null hypothesis and accept the alternative hypothesis, H_a, which recognizes a significant difference in the distributions of the different features (F-ratio = 524.6, p-value < $2e^{-16}$).

4.3. Assignment of Ranks

The final phase deals with assigning a 'desirability' score to each of the biodiesels, based on which they are finally ranked. To keep the weights of all the attributes positive, we complement the normalized values of particular attributes negatively impacting the overall score, namely: density and viscosity. We then compute the average (**Fig. 8**) of the normalized features and assign weights to them basis this tables reversed percentage of the total, prioritizing the scarcest attribute to assume the highest weight. Next, we transform the sum of products within 100 to generate the desirability and eventual ranks (Eq. 5).



Normalized, Desirable Avg. of Attributes

Avg. Density’	0.0694
Avg. Oxidation Stability	0.2133
Avg. Flash Point	0.4976
Avg. Viscosity’	0.6929
Avg. Heating Value	0.7417
Avg. Cetane Number	0.7927

Fig. 8: Transformed attributes averages after normalization

$$Desirability = (Density * 0.2635 + Oxidation\ Stability * 0.2465 + Flash\ Point * 0.2304 + Viscosity * 0.1654 + Heating\ Value * 0.0709 + Cetane\ Number * 0.023) * 100 \qquad (5)$$

5. Results and Discussions

5.1. Results

This article focusses on ranking the biodiesels based on the critical physicochemical properties chosen. As discussed previously, the properties, e.g. density, kinematic viscosity, heating value and CN etc. directly affect the combustion of these fuels. However, during long term storage of fuels, the physicochemical properties deteriorate which is governed by the composition of biodiesel. Any change in composition determined through change is oxidation stability and flash point. Flash point also determines safe handling temperature for any fuel. As such, flash point and oxidation stability are critical factors for transport and long term storage of biodiesel. A ranking system has been proposed in this study based on those selected properties. The ranking consists of three phases (or, super-steps): A, B, C and their subordinate sub-steps, which have been carried out using the platforms: Python, R and Tableau respectively. Table 3 shows the ranking of the first 25 biodiesel feedstocks. The study endorses *Brassica juncea*, Cardoon (*Cynara carduncculus*), poppyseed oil, coffee oil and rapeseed oil as the Top 5 desirable biodiesels from the ranking policy which have the most desirability scores (ranging from 65.49 to 46.58) (**Table 3**). However, biodiesel ranked between 18 to 25th: Safflower, tobacco, soybean, *citrus sinesis*, beauty leaf oil, dairy scum, fish oil and waste eggshell all have almost similar desirability score. Thus a fast-tracked result has been obtained from a feedstock tally of 71 with six chosen physicochemical properties for each feedstock. A previous study by Anwar [38] used not only physicochemical aspect of biodiesels but also the economic and environmental aspects of those while ranking sixteen most popular biodiesel feedstocks. He used four separate MCDA systems: PROMETHEE-GAIA, WSM, WPM and TOPSIS with five weighting methods, EQUAL, CRITIC, ENTROPY, AHP, and FAHP were used to come to a conclusion. Out of twenty results, fourteen ranked coconut oil as the most favoured feedstock with *moringa* is the second preferred option which signifies a variance based on the weighting method used. Our proposed method is a simplified approach in this regard.

Table 3: Desirability-scores and ranks assigned against each biodiesel

Rank	Avg. Desirability	Biodiesel	Rank	Avg. Desirability	Biodiesel
1	65.49	Brassica	**37**	36.39	*Michelia champaca*
2	63.12	Cardoon	**38**	36.38	Rice bran
3	59.66	Poppy seed oil	**39**	36.38	Canola
4	59.44	Coffee	**40**	36.17	Algae
5	46.58	Rapeseed oil	**41**	36.16	Tannery waste
6	44.43	Stone fruit kernel	**42**	36.16	Waste cooking oil
7	43.36	Linseed	**43**	36.06	Kusum
8	42.74	sPapaya Seed	**44**	35.79	Karanja
9	42.25	Palm	**45**	35.67	*Calophyllum inophyllum*
10	41.27	Jatropha curcas	**46**	35.12	Marine fish oil
11	40.36	Cotton Seed	**47**	35.10	Yellow Oleander
12	40.25	argemone	**48**	35.03	*X. sorbifolium*
13	40.04	Styrax officinalis L.	**49**	34.95	Coconut
14	39.33	Waste fish oil	**50**	34.79	Animal fat
15	39.13	Sesame	**51**	34.62	Chlorella vulgaris
16	38.98	Safflower	**52**	34.47	Tetradesmus obliquus
17	38.96	Tobacco	**53**	34.45	waste edible oil

18	38.59	Soybean	**54**	34.21	Spirulina
19	38.57	citrus sinensis	**55**	34.12	Groundnut
20	38.31	Beauty leaf tree	**56**	34.09	Neem
21	38.19	Dairy Scum	**57**	33.86	*Chlorella protothecoides*
22	38.16	Fish oil	**58**	33.75	*Crambe abyssinica*
23	38.10	Waste eggshell	**59**	33.55	Babassu
24	37.86	Hazelnut	**60**	33.06	Date seed oil
25	37.67	Beef tallow	**61**	32.97	Fish waste
26	37.63	Peanut	**62**	31.93	*Terminalia catappa*
27	37.22	*Simarouba Glauca*	**63**	31.50	Red pine
28	37.10	Olive	**64**	31.28	White pine
29	37.08	Sunflower	**65**	31.27	Cascabela ovata
30	37.07	Camelina	**66**	30.95	Mahua
31	37.01	Chicken fat	**67**	30.64	Jojoba
32	36.98	Dairy washed milkscum	**68**	30.63	Cypress
33	36.97	Rubber	**69**	26.38	Cedar
34	36.73	Bitter almond	**70**	25.38	Castor
35	36.66	Waste frying oil	**71**	17.43	Hiptage
36	36.64	Corn			

5.2. Advantages of Proposed Scheme from Computational and Empirical Viewpoints

The scheme proposed in this paper for choosing general-purpose biodiesels is unique since this is a procedural, intuitive method free from dependence on prior experiences, more popularly known as supervised or labelled datasets in computer science. The method does not rely on refining parameters using any sophisticated learning algorithm but rather, overcomes the limitations of such algorithms' hunger for feeding in prior intelligence. The procedure exhibits more of its intuitive nature in the following ways:

- Usage of a simple statistical median could evenly separate the properties as desirable or unwanted, paving the way to creating a single boolean vector (or, a transaction in the jargon of the Business Intelligence algorithm: APRIORI) against each biofuel. This indicated if the properties were favourable and facilitated the formation of groups, which in turn lent the intelligence, whether certain helpful properties were found together.

- The method could overcome the limitation of missing entries by inferring those from ARIORI-results. The grouped itemsets revealed tendencies from complete tuples, which were eventually mapped to the incomplete ones.

- The method ensures a fair comparison of all the parameters by normalizing them. The comparison is further strengthened by the complementation of properties that are more favourable in their lower numeric values. In short, the act of complementation ensured orientation towards desirability.

- After setting up an ANOVA-tested competitive environment based on favourability for mechanical engineering purposes, the method ensures higher scores of biofuels showing greater values on rarer qualities. The ranker implements this idea by setting weights against each criterion by reversing the percentage of the total of all average—normalized, and in some cases, complemented as well—properties.

- The solution here serves a general-purpose utility, showing the flexibility to be tailored for case-specific purposes. This can be executed by setting up transactions after comparing with specialized desirables, instead of descriptive medians.

6. Conclusions

Biodiesel is considered as one of the most promising alternatives to reduce the dependency on diesel import. The paper describes some critical properties which were used to rank the 71 most reported biodiesel feedstocks in the literature. The biodiesel feedstocks chosen in this study adheres to the biodiesel standards (ASTM D6751 and EN14214). This paper proposes a ranking method to select the most suitable biodiesel based on six vital physicochemical properties which include: density (in kg/m^3), kinematic viscosity (in mm^2/s), heating value (in MJ/kg), flash point (in °C), cetane number and oxidation stability. The three phases and their subordinate sub-steps have been carried out using the platforms: Python, R and Tableau, respectively. The proposed ranking system ranks the biodiesel based on desirability score, and the top 5 are mustard (*Brassica juncea*), cardoon (*Cynara cardunculus*), poppy seed, coffee and rapeseed biodiesel. By no means we claim that the proposed method is the optimal method for ranking biodiesel. There are many other methods discussed in the literature. This work represents the use of more streamlined software to perform the task. We believe that there is a great deal of potential for designing even better techniques for scoring and ranking using association rules mining.

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CRediT author statement

S.M.Ashrafur Rahman: *Conceptualisation, Writing- Original Draft, Software, Validation;* **I.M. Rizwanul Fattah**: *Investigation, Resources, Writing - Review & Editing;* **Shithi Maitra**: *Methodology, Software, Validation, Formal analysis, Data curation*; **TMI Mahlia**: *Writing - Review & Editing, Resources, Supervision.*