

# Analytical Assessment of C2–C8 Alcohols as Spark-Ignition Engine Fuels

Thomas Wallner, Andrew Ickes and Kristina Lawyer

**Abstract** The U.S. Renewable Fuel Standard (RFS2) requires a drastic increases in production of advanced biofuels up to 36 billion gallons over the next decade while corn-based ethanol will be capped at 15 billion gallons. Currently ethanol is the predominant alternative fuel and is widely distributed at 10 vol % blends in gasoline (E10). However, certain properties of ethanol make it less desirable as a blending agent in particular at higher blend levels. Therefore the engine- and vehicle-related properties of longer chain alcohols are evaluated in comparison to gasoline to determine their suitability as blending agents for spark-ignition engine fuels. This analytical study aims at providing comprehensive property data for a range of alcohol isomers with a carbon count up to C8. Relevant physical property data is used to determine the general suitability of longer chain alcohol isomers as blending agents based on factors such as melting point and boiling. Based on initial findings the scope of the study was narrowed down to alcohols in the C2–C6 range. It was determined that the engine- and combustion-relevant information is missing from the literature for a wide range of longer chain isomers. Thus fuel testing for engine-relevant properties such as lower heating value, knock resistance (RON, MON) and Reid Vapour Pressure (RVP) for alcohols up to C6 was performed as part of this study. Data suggests that the melting point of alcohols

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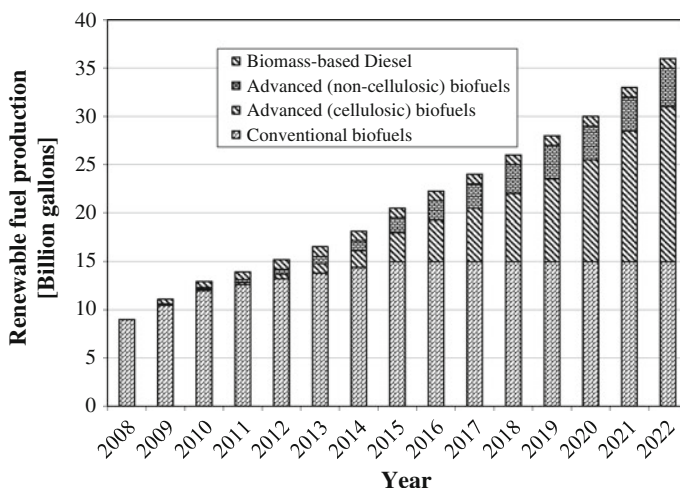
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increases with increasing carbon count and all C7 and C8 isomers exhibit melting points in excess of  $-40\text{ }^{\circ}\text{C}$  making their use as vehicle fuel questionable. Boiling points increase with increasing carbon count and *n*-structures generally have slightly higher boiling points than their respective *iso*-structures. Latent heat of vaporization decreases with carbon count, the mass-specific value for ethanol is triple that of gasoline, the energy specific ratio increases to a factor of 5. Alcohol fuels generally have a significantly lower RVP than gasoline, RVP decreases with increasing carbon count. Stoichiometric air demand and fuel energy content increase with carbon count. Knock resistance expressed as Research Octane Number (RON) and Motor Octane Number (MON) decreases significantly with increasing carbon count, *iso*-structures show increased knock resistance compared to their respective *n*-structures. This study is limited to analytical results as well as fuel property testing according to ASTM standards. Only properties of neat alcohols are evaluated in comparison to gasoline certification fuel, gasoline blend stock for ethanol blending and E10. The analysis of the reported properties is further focused on spark-ignition engine applications only. Future phases of this project will include the assessment of properties of multi-component blends as well as efficiency, performance and emissions testing on a modern direct-injection engine. While data for a limited number of commonly used alcohols such as ethanol and *iso*-butanol is available in the literature, little or no data is available for a majority of other alcohols and their isomers. In addition, engine-related data published in the past occasionally disregards the significant differences between alcohol isomers of the same chain length. This study offers a comprehensive review of physical properties of alcohols and their common isomers in the C2–C8 range as they relate to in-vehicle use and spark-ignition combustion engine application. Data presented in this paper suggests that higher alcohols have certain physical properties that might be desirable for blending with gasoline. Due to their oxygen content all alcohols have an inherent disadvantage in terms of energy content compared to non-oxygenated fuels. While this disadvantage becomes less pronounced with increasing carbon count, other less desirable properties such as a low RVP and reduced knock resistance become more dominant with longer chain length alcohols. In addition to merely evaluating properties, the selection of promising alcohols and blend levels will ultimately depend on the introduction scenario and target properties.

**Keywords** Alcohol fuels • Spark-ignition engine • Physical properties • Combustion • Knock resistance • RON • MON • RVP

## 1 Background

In the United States ethanol is the predominant alternative fuel for spark ignition engine applications and is widely used in blends of 10 vol % in gasoline (E10). Further, the U.S. Environmental Protection Agency (EPA) recently granted a



**Fig. 1** Production targets according to the U.S. renewable fuel standard (RFS2) [7]

waiver for use of blends of 15 vol % of ethanol in vehicles starting from model year 2001 [1]. Although more than 10 Mio vehicles on the road today are Flex Fuel vehicles capable of operating on E85, only a small fraction of these vehicles are actually fueled on the high-level ethanol blend. At the same time, the U.S. Renewable Fuel Standard (RFS2) calls for a dramatic increase in production of alternative fuels until 2022 (see Fig. 1). Other alcohols beyond ethanol may possess properties which make them suitable for vehicle use, providing opportunities for further petroleum displacement.

A research project with the working hypothesis that a certain blend of gasoline with ethanol as well as higher alcohols could provide a fuel with properties superior to those of traditional gasoline-ethanol blends is currently underway at Argonne National Laboratory. In addition to ethanol, which is already widely used in blends with gasoline (E10) in the US market, alcohols with up to eight carbon atoms (C8—octanol) were considered for this study. Three groups of criteria were identified as critical to selecting suitable higher alcohols blends; (1) fuel properties, (2) production process, and (3) Emissions and Performance. The scope of the initial phase of the project, which is summarized here, focuses exclusively on the fuel properties of these higher alcohols. Other aspects which will be covered in later phases include the production process and potential synergies of certain mixed alcohols as well as the impact of alcohol fuels on engine performance and emissions characteristics. The final selection of promising alcohol fuel blends will be based on several sub-criteria within the three main groups with targets for near term, mid-term and long-term scenarios.

**Table 1** Overview of C1–C8 alcohols

Common name	Carbon count	Chemical composition	Oxygen content (mass %)
Methanol	1	CH <sub>4</sub> O	49.9
Ethanol	2	C <sub>2</sub> H <sub>6</sub> O	34.7
Propanol	3	C <sub>3</sub> H <sub>8</sub> O	26.6
Butanol	4	C <sub>4</sub> H <sub>10</sub> O	21.6
Pentanol	5	C <sub>5</sub> H <sub>12</sub> O	18.1
Hexanol	6	C <sub>6</sub> H <sub>14</sub> O	15.7
Heptanol	7	C <sub>7</sub> H <sub>16</sub> O	13.8
Octanol	8	C <sub>8</sub> H <sub>18</sub> O	12.3

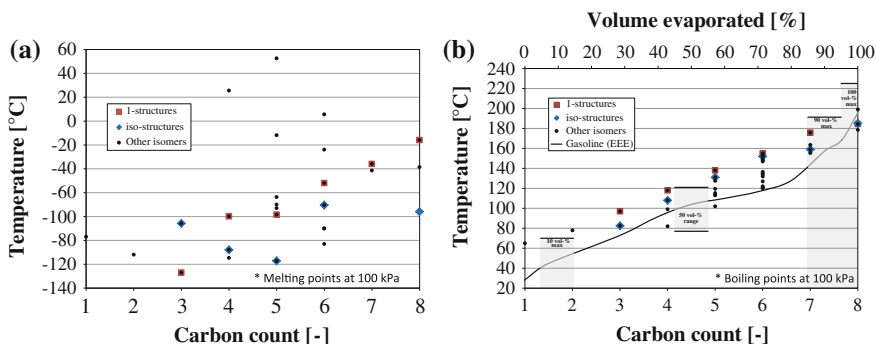
## 2 Overview of Alcohol Fuels

Table 1 shows an overview of the alcohols with carbon counts from one to eight. Alcohols are characterized as organic compounds with a hydroxyl functional group (–OH) bound to a carbon atom. The table shows the common name of alcohols up to eight carbon counts together with their chemical composition. Alcohols contain oxygen as part of the hydroxyl group and the oxygen content is one of the main differentiating factors and will be used alternately with carbon count to distinguish between the different alcohols. For alcohols with a carbon count of 3 or higher, isomers exist that can be differentiated by their structure and the locating of the hydroxyl group. The straight chain isomer with the hydroxyl group connected to the terminal carbon is commonly referred to as *n*-isomer or *l*-isomer (e.g. *n*-butanol or *l*-butanol). The branched isomer with the OH-group at the terminal carbon is called *iso*-structure.

The properties of certain alcohol isomers with the same carbon count differ significantly based on structural differences. Also, there are significant limitations in the availability of property data especially for many of the less common isomers of longer-chain alcohols. Current production levels of the most common alcohol isomers, such as methanol, ethanol, *iso*-propanol, *n*-butanol and *iso*-butanol each exceed 1 billion lbs per year.

## 3 Physical Properties

This section covers the analytical assessment of alcohol fuels based on physical properties relevant to engine and vehicle operation. The melting point of a substance is critical to ensure that the fuel stays liquid regardless of region and season. Figure 2a shows the melting points of a large range of alcohol isomers as a function of carbon count with *l*- and *iso*-structures highlighted were applicable. Although there are several outliers, a general trend of increasing melting temperature with longer carbon chain length can be observed. Using –40 °C as a realistic upper limit shows that several hexanol and octanol isomers are close or

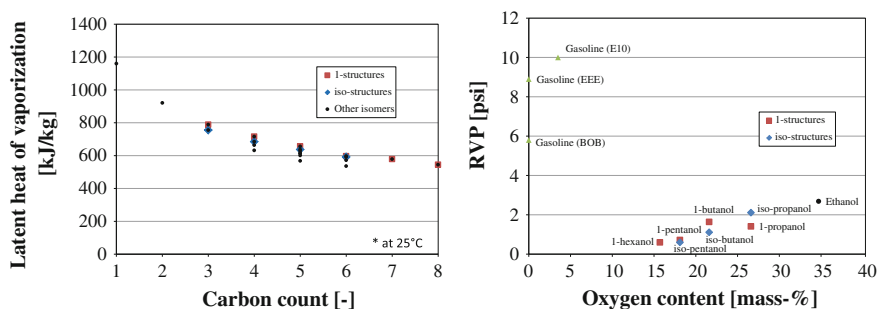


**Fig. 2** a Melting points b boiling points of alcohols and distillation curve of gasoline

even exceed this threshold. It is also worth noting that *tert*-butanol far exceeds the threshold with a melting point of 25.5 °C.

Fuel evaporative behavior is critical to ensure proper engine cold start as well as hot operation. Since alcohols are single components, they have a boiling point rather than an evaporation curve typical for multi-component fuels such as gasoline. ASTM standard D4814 specifies certain points in the evaporative curve of gasoline fuels, specifically the point at which 10 vol % are evaporated at a maximum of 70 °C, 50 vol % between 77 and 121 °C and 90 vol % at a 190 °C maximum. The end point is limited to a 225 °C maximum. Figure 2b shows the boiling point of a range of alcohol fuels as a function of carbon count for *I*- and *iso*-structures as well as other isomers. A clear trend of increasing boiling temperature with increasing carbon chain length can be observed for the alcohols investigated in this study. In addition the *I*-structures show a slightly increased boiling point compared to the respective *iso*-structures. Boiling temperatures for the considered range of C1–C8 alcohols ranges from 65 °C for methanol to almost 200 °C for certain octanol isomers. Figure 2b also includes a distillation curve for certification gasoline (Tier II EEE U.S. Federal Emission Certification Fuel) displaying volume evaporated as a function of temperature (X-axis on the top). The fact that alcohols evaporate at a defined boiling point versus the distillation range typical for gasoline results in an extension of the gasoline curve at the boiling point of the alcohol if blended. Therefore, the boiling point is critical in influencing the evaporative behavior in particular when longer chain alcohols are used or when targeting higher blends levels.

Latent heat of vaporization is another critical factor influencing in-cylinder mixture preparation in engines. Heat of vaporization results in a temperature reduction inside the engine intake system for port fuel injection and in-cylinder for direct-injection engines since energy taken from the intake air is required to evaporate the fuel. Increased heat of vaporization is desirable particularly for direct injection engines because it can significantly reduce in-cylinder process temperatures thus reducing NO<sub>x</sub> emissions formation as well as knock propensity. As shown in Fig. 3a, mass-specific latent heat of vaporization decreases with

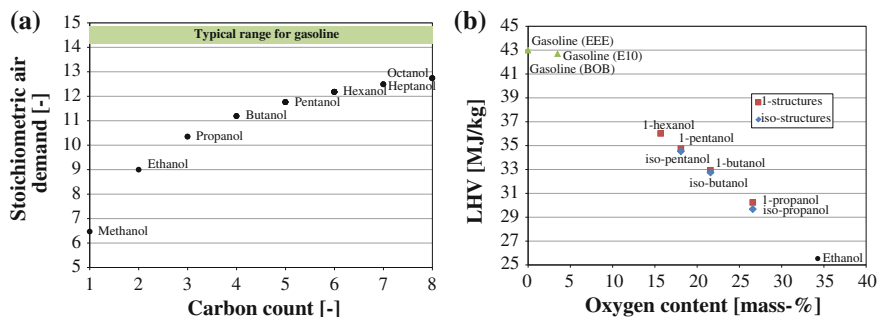


**Fig. 3** **a** Latent heat of vaporization **b** Reid Vapor Pressure (RVP) for various alcohols

increasing carbon count. The differences in heat of vaporization within a group of alcohols are limited compared to the significant differences between different chain length alcohols. The energy needed to evaporate 1 kg of ethanol (921 kJ) is almost triple that of *iso*-octanol (350 kJ). For comparison, the latent heat of vaporization of gasoline is around 300 kJ/kg.

Vapor pressure is used to determine the volatility of gasoline and other spark-ignition engine fuels. The Standard Test Method for Vapor Pressure of Petroleum Products (Mini Method) according to ASTM 5191 was used to compare Reid Vapor Pressure (RVP) for the fuels considered in this study. This test method is suitable for testing samples with boiling points above 0 °C that exert a vapor pressure between 7 and 130 kPa (1.0 and 18.6 psi) at 37.8 °C. Based on comparative tests it was also determined that this method is applicable for gasoline/ethanol blends [1]. The RVP values for a range of gasoline fuels as well as alcohol isomers as a function of oxygen content are shown in Fig. 3b. While all tested fuels meet the boiling point requirements of 0 °C (see Fig. 2b), the two pentanol isomers as well as *l*-hexanol fall outside the specified vapor pressure range. The results show a clear trend of decreasing RVP with decreasing oxygen content for the alcohol fuels, while all tested alcohols display RVP values significantly below those of gasoline. ASTM D4814 specifies the maximum RVP for gasoline fuels with 9.0 psi as the maximum in “volatility attainment areas” and 7.8 psi in “volatility non-attainment areas”. In addition a 1 pound waiver was granted for gasoline/ethanol blends with at least 9 vol % and no more than 10 vol % of ethanol practically increasing the maximum to 10 psi. This is significant since low level ethanol blends (as well as methanol and propanol) show near-azeotropic behavior with an increase in RVP at low blend levels when mixed with gasoline [1]. In general, blending gasoline with higher alcohols (C4 and higher) is expected to decrease Reid Vapor Pressure regardless of blend levels. While lower RVP is desirable from an evaporative emissions point of view, too low an RVP can cause cold start issues and related increase in hydrocarbon emissions.

The stoichiometric air demand describes the mass ratio of air to fuel that is required to create a stoichiometric mixture. Assuming that the amount of air inside an engine is approximately constant for a given load, a decrease in stoichiometric



**Fig. 4** a Stoichiometric air demand b Lower Heating Value (LHV) for alcohol fuels

air demand suggests that an increased amount of fuel is required to create a stoichiometric mixture. Figure 4a shows the stoichiometric air demand for the range of C1–C8 alcohols as a function of carbon count. Since air demand does not depend on molecular structures, there is no difference in air demand among isomers. However, as alcohols are characterized by an OH functional group, the fuel itself already contains oxygen (see Table 1). The impact of the single OH group on air demand decreases with increasing carbon count reflected in increased stoichiometric air demand. For reference the typical range of stoichiometric air demand for gasoline fuels ( $\sim 14.5$ ) is also shown in Fig. 4a.

Mass-specific fuel energy content, characterized by lower heating value (LHV) for several gasoline types and a range of alcohol isomers is shown in Fig. 4b as a function of fuel oxygen content. As can be clearly seen, increasing oxygen content of the fuel almost linearly reduces the lower heating value from approx. 42 to 43 MJ/kg for gasoline to approx. 26 MJ/kg for ethanol. This almost 40 % reduction in mass-specific energy content can be reduced with longer-chain alcohols such as butanol with a LHV penalty compared to gasoline of approx. 23 % or hexanol with a respective penalty of only 16 %. However, due to their oxygen content the energy per unit mass is still significantly reduced compared to the gasoline baseline. Among the alcohol isomers, *1*-structures show slightly higher lower heating values compared to their respective *iso*-structures.

Other relevant factors when evaluating the suitability of alcohol fuels for engine combustion include physical properties such as solubility in water and combustion-related factors such as laminar flame speeds and ignitability limits. Solubility of alcohols in water decreases with increasing carbon count; C1–C3 alcohols are fully miscible, miscibility decreases from C4–C8 alcohols and higher alcohols are immiscible with water. Solubility is particularly relevant because it influences transportability in pipelines [2]. The flammability limits of short-chain alcohols are similar to those of gasoline. At increasing chain length the upper flammability limit decreases while the more relevant lower flammability limits remains constant. Limited data is available to suggest trends in terms of laminar burning velocity of longer-chain alcohol fuels. Data available for C1–C4 alcohols suggests decreasing laminar flame speeds with increasing carbon chain length.

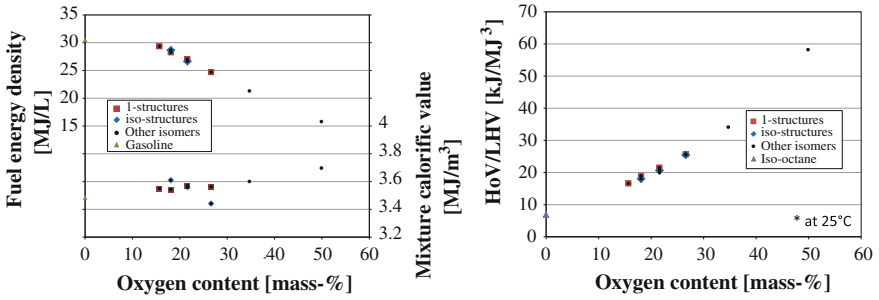


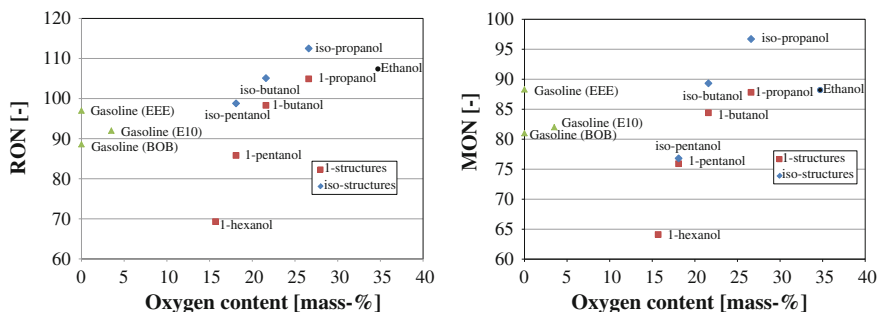
Fig. 5 a Energy content b energy-specific heat of vaporization of various alcohols

## 4 Engine-Related Properties

In addition to relevant physical properties of alcohols compared to gasoline discussed in the previous chapter, there are several critical engine-related properties that need to be considered when selecting alcohol fuels for blending with gasoline. While the lower heating value as a metric for mass-specific energy content is relevant when comparing fuels, the volumetric energy content of a fuel directly affects injection duration when used in an engine. This is particularly relevant since typical engine calibrations utilize injection duration as a metric for the amount of energy introduced to the engine and changes in injection duration influence the mixture formation process in particular for direct injection engines. The top portion of Fig. 5a shows the fuel energy density of gasoline as well as several alcohol isomers as a function of fuel oxygen content. Due to the increased density of alcohol fuels ( $\sim 790\text{--}830\text{ kg/m}^3$ ) compared to gasoline ( $\sim 740\text{ kg/m}^3$ ) the difference in volumetric energy content is not as pronounced as the gravimetric energy density (compare Fig. 4b). Nonetheless the volumetric energy content of ethanol ( $\sim 21.3\text{ MJ/l}$ ) is still more than 30 % below that of gasoline ( $\sim 31.6\text{ MJ/l}$ ) while that of *iso*-pentanol ( $28.3\text{ MJ/l}$ ) is only approx. 10 % below the gasoline benchmark. The lower portion of Fig. 5a shows the mixture calorific value (right axis) for gasoline and alcohols versus oxygen content. The mixture calorific value is a metric for the energy content of a certain volume of stoichiometric air/fuel mixture; it is assumed that the fuel is introduced directly into the combustion chamber in which case the mixture calorific value is calculated based on lower heating value, stoichiometric air demand and air density [1]. The mixture calorific value for gasoline is approx.  $3.5\text{ MJ/m}^3$ , while the values for alcohols are slightly higher with approx.  $3.55\text{ MJ/m}^3$  for propanol, butanol and hexanol,  $3.6\text{ MJ/m}^3$  for ethanol and almost  $3.7\text{ MJ/m}^3$  for methanol. Assuming constant engine efficiencies and constant volumetric efficiency for the different fuels, an advantage in mixture calorific value directly translates into an improvement in engine torque.

Considerations analogous to the gravimetric versus volumetric energy content are the basis of determining the latent heat of vaporization per unit energy. The mass specific latent heat of vaporization shown in Fig. 3a is a useful metric to



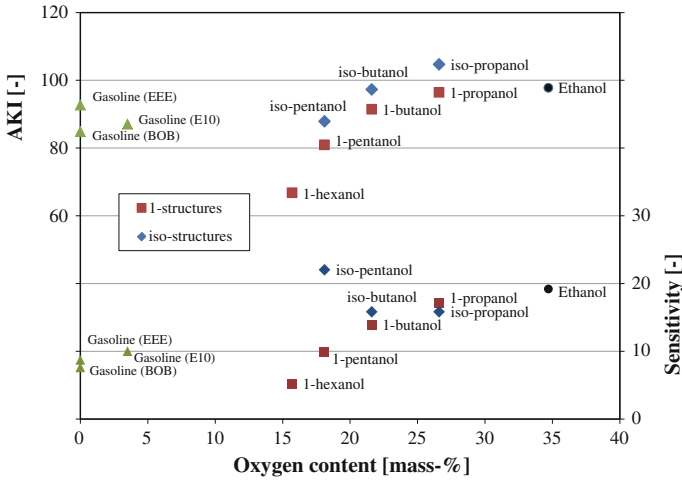


**Fig. 6** **a** Research Octane Number (*RON*) **b** Motor Octane Number (*MON*) for gasoline and various alcohol fuels

compare fuels, the result in the engine is a cooling effect. This charge cooling effect is particularly pronounced for direct injection engines and can significantly improve the knock resistance [3–5]. Therefore, the engine-relevant metric would be the latent heat of vaporization normalized by the amount of fuel energy delivered to the engine. The quantification of this fuel-energy-specific heat of vaporization is shown in Fig. 5b as a function of fuel oxygen content. The latent heat of evaporation value of *iso*-octane (representative for gasoline) is approx. 7 kJ/MJ of fuel energy. For alcohols the value increases linearly with oxygen content and reaches a maximum for methanol. For comparison, the fuel-energy-specific latent heat of vaporization of ethanol is approx. 34 kJ/MJ which is almost a five-fold increase compared to gasoline (the mass-specific latent heat only increased by a factor of three as discussed earlier).

While the heat of vaporization potentially improves the knock behavior, the traditional measure for knock resistance is the octane number. Spark-ignition engine fuels are classified based on Research Octane Number (*RON*) according to ASTM D2699 and Motor Octane Number (*MON*) according to ASTM D2700. Although the applicability of *RON* and *MON* in modern, downsized, turbo-charged engines has been widely discussed [3–6], the values are still a commonly used benchmark for fuel characterization. Both, *RON* and *MON* are determined by comparing a fuel’s knock behavior to that of a mix of primary reference fuels using a single-cylinder CFR engine. Both, engine speed and operating temperatures are higher for *MON* determination compared to *RON*.

Figure 6a shows the measured *RON* values for a range of gasoline fuels as well as several alcohol isomers. Gasoline EEE has a higher *RON* value ( $\sim 98$ ) compared to both, a blend stock for ethanol blending (BOB at *RON*  $\sim 87$ ) as well as a 10 vol % blend of BOB with ethanol (E10 at *RON*  $\sim 92$ ). Alcohol fuels span a wide range of Research Octane Numbers with a general trend of decreasing *RON* with decreasing oxygen content. *Iso*-structures generally have higher *RON* values than the respective *n*-structures. This difference accounts for approx. 7 points for propanol and butanol and increases to 13 points for pentanol. *Iso*-propanol exhibits the highest *RON* of all tested fuels at  $\sim 112.5$ , while *1*-hexanol has a *RON* of only



**Fig. 7** AKI and sensitivity for gasoline and various alcohol fuels

69.3. It is also worth noting that Research Octane Numbers do not scale linearly when blending gasoline and alcohols as shown in [5].

Due to the higher test speed and operational temperatures, Motor Octane Numbers are typically lower than Research Octane Numbers. Figure 6b shows measured MON for the same range of gasoline and alcohol fuels. EEE features a MON of 88, BOB has a MON of 81 and E10 has a MON of 82. A trend in terms of decreasing MON with increasing alcohol chain length similar to RON behavior can be observed. Also, *iso*-structures display increased knock resistance compared to *n*-structures. However, while the RON difference increased with longer chain length, the advantage in MON of *iso*-structures over *n*-structures decreases from approx. 9 points for propanol to 5 for butanol and less than one point for pentanol.

Other widely used fuel characteristics derived from RON to MON values are the Anti-Knock Index ( $AKI = (RON + MON) / 2$ ) and knock sensitivity ( $RON - MON$ ). Figure 7 shows AKI and sensitivity as a function of oxygen content for gasoline and the range of alcohol fuels considered in this study. The AKI for E10 turns out to be 87  $[(82 + 92)/2]$  which is typical for regular pump gasoline (containing up to 10 vol % ethanol). *Iso*-propanol with a value of approx. 105 exhibits the highest AKI of all tested fuels. The previously identified trends of decreasing knock values with increasing chain length as well as improved values of *iso*-structures over *n*-structures remain unchanged. The advantage in AKI of *iso*-structures over *n*-structures remains almost constant with approx. 6–8 points regardless of chain length. This is due to the fact that sensitivity of *n*-structures decreases with increasing chain length while it increases for *iso*-structures as shown in the bottom of Fig. 7. Sensitivity of gasoline fuels is in the range of 8–10 which is considerable lower than most alcohols except for the longer chain *n*-structures of pentanol and hexanol.

## 5 Conclusions

An analytical assessment of the properties of alcohols up to C8 was performed to determine their suitability as fuels for spark ignition engines. While some, mainly shorter chain alcohols that are produced in quantities of more than 1 billion gallons annually are well characterized, little to no relevant property specifications are available for certain longer chain isomers. The following general trends can be derived based on increasing carbon chain length or decreasing oxygen content respectively. Specific differences between common structures, in particular *n*- and *iso*-structures are noted were applicable:

- The melting point of alcohols increases with increasing carbon count and several C7 and C8 isomers exhibit melting points in excess of  $-40^{\circ}\text{C}$  making their use as vehicle fuels questionable
- Boiling points increase with increasing carbon count and *n*-structures generally have slightly higher boiling points than their respective *iso*-structures
- Latent heat of vaporization decreases with carbon count, the mass-specific value for ethanol is triple that of gasoline, the energy specific ratio increases to a factor of 5
- RVP decreases with increasing carbon count and alcohol fuels generally have a significantly lower RVP than gasoline
- Stoichiometric air demand and fuel energy content increase with carbon count
- Knock resistance expressed as Research Octane Number (RON) and Motor Octane Number (MON) decrease significantly with increasing carbon count, *iso*-structure show increased knock resistance compared to their respective *n*-structures.

Overall, the Renewable Fuel Standard (RFS2) requires a significant increase in production of advanced, cellulosic and non-cellulosic biofuels. Longer-chain alcohols might turn out to be an interesting alternative to ethanol due to their properties which more closely resemble gasoline.

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