

An Alternative Method Based on Toluene/n-Heptane Surrogate Fuels for Rating the Anti-Knock Quality of Practical Gasolines

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ABSTRACT

As SI engines strive for higher efficiency they are more likely to encounter knock and fuel anti-knock quality, which is currently measured by RON and MON, becomes more important. However, the RON and MON scales are based on primary reference fuels (PRF) - mixtures of iso-octane and n-heptane - whose autoignition chemistry is significantly different from that of practical fuels. Hence RON or MON alone can truly characterize a gasoline for its knock behavior only at their respective test conditions. The same gasoline will match different PRF fuels at different operating conditions. The true anti-knock quality of a fuel is given by the octane index, $OI = RON - KS$ where $S = RON - MON$, is the sensitivity. K depends on the pressure and temperature evolution in the unburned gas during the engine cycle and hence is different at different operating conditions and is negative in modern engines.

In this paper we propose that the gasolines are ranked against toluene /n-heptane mixtures (toluene reference fuel, TRF). The gasoline is assigned a Toluene Number (TN), which is the volume percent of toluene in the TRF which matches the gasoline for knock in the CFR RON test. Since TRFs will have comparable sensitivity to gasolines for the same RON, they will have comparable OI at different engine conditions and the TN will describe the knock behavior of the gasoline reasonably well at all different conditions. The paper describes the method and shows supporting experimental data. The other advantage of such a method is that it will enable a better quantitative description of fuels, such as ethanol mixtures, with $RON > 100$ and hence beyond the maximum of the current octane scale.

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1. INTRODUCTION

Knock is an abnormal combustion phenomenon in SI engines that imposes a fundamental limit on the efficiency of SI engines [1, 2, 3]. Knock is caused by the autoignition of the end-gas ahead of the flame front and depends on the pressure and temperature history of the end-gas as well as on the anti-knock quality of the fuel.

Fuel anti-knock quality has to be described by empirical measures because the fundamental autoignition chemistry cannot be adequately quantified for practical fuels because of their complexity. It is traditionally measured by Research and Motor Octane Numbers, RON and MON, of the fuel used. The RON test is run in a single-cylinder CFR (Cooperative Fuels Research) engine at an engine speed of 600 rpm and an intake temperature of 52° C while the MON test is run at 900 rpm and with a higher intake temperature of 149°C. These tests are run in accordance with the procedures set in [4] - ASTM D2699 for RON and ASTM D2770 for MON.

The octane scale is based on two paraffins, n-heptane and iso-octane. Blends of these two primary components are referred to as primary reference fuels (PRF) and define the intermediate points in the RON or MON scale. The RON or MON is the volume percent of iso-octane in the PRF. Thus a blend of 95% of iso-octane and 5% of n-heptane by volume is assigned the octane number of 95 in both RON and MON scales. A gasoline is assigned the RON (or MON) value of the PRF that matches its knock behavior in the RON (or MON) test.

In fact the refining industry has developed over the past eighty years or so on the assumption that PRFs are suitable knock surrogates for practical gasolines. However, the autoignition chemistry of non-paraffinic components in gasoline is different from that of PRF and RON or MON describes the anti-knock behavior of the gasoline only at the RON or MON test condition. The true anti-knock quality of a gasoline is best described by an Octane Index, OI [3, 5, 6, 7] which is defined as

$$OI = (1-K)RON + KMON = RON - KS$$


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where K is an empirical constant which depends on the pressure and temperature history of the unburned mixture in the cylinder. OI is the octane number of the PRF that matches the knock behavior of the gasoline that is of interest, at the conditions of the particular engine test. The temperature of the unburned gas increases as pressure increases in the engine cylinder and pressure and temperature are determined by engine design and operating conditions. For instance, at the RON test condition, the temperature in the unburned gas at a given pressure is lower than in the MON test condition as shown in Figure 1. A practical gasoline will match PRFs of different octane numbers at different operating conditions. For instance a typical European gasoline might have a RON of 95 and MON of 85. Hence its OI will be 95, i.e. it will behave like PRF 95 in the RON test condition, but will match PRF 85 in the MON test condition. The difference between RON and MON is known as the sensitivity, S . Practical fuels have $S > 0$, typically around 10. The sensitivity, S , can be taken as a measure of how different the fuel autoignition chemistry is compared to PRF, which by definition, have zero sensitivity.

The value of K has to be determined empirically by measuring a quantity dependent on fuel autoignition quality, such as the Knock Limited Spark Advance, KLSA, at a fixed operating condition for fuels of different RON and MON [5, 6]. K is a measure of how different the pressure and temperature regime is compared to the RON test condition. If the temperature vs pressure curve falls above the RON test condition in Fig. 1, $K > 0$. Otherwise $K < 0$ - such an example is also shown in Fig. 1.

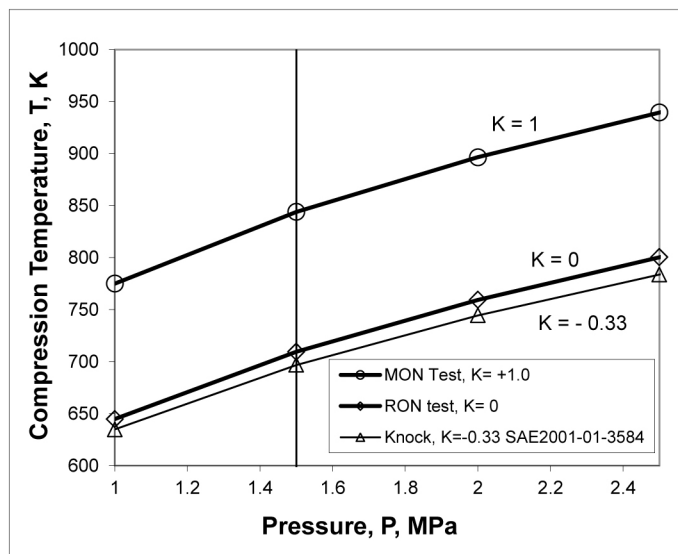


Figure 1. Variation of the temperature of the unburned mixture with pressure. It depends on engine design and operating conditions.

The value of K and hence OI for a given fuel depends on the design and operating condition of the engine. All measures aimed at improving the efficiency of a SI engine, such as increasing the compression ratio and turbocharging (aligned with downsizing) increase the maximum pressure and the temperature of the charge making autoignition and knock more likely. Fuel anti-knock quality becomes more important.

Moreover, with increase in efficiency, for a *given* pressure, the temperature decreases or alternatively, for a *given* temperature, the pressure increases. This happens with turbocharging in down-sized engines and if the compression ratio is increased, because the amount of hot residual charge at the end of the exhaust stroke is reduced. In [5], T_{comp15} , the unburned mixture temperature when the pressure is 15 bar (1.5 MPa) was introduced and helps to place the operating condition of interest in relation to the RON test condition. If T_{comp15} is less than that for the RON test, the condition is said to be "beyond RON" and K in Eq. 1 is negative so that for a given RON, a lower MON fuel has higher OI, i.e. higher anti-knock quality.

Multiple studies indeed show that modern engines and downsized turbocharged engines of the next generation have negative values of K at the conditions where their performance is limited by knock [5, 6, 7, 8, 9, 10, 11, 12, 13]. Studies in HCCI engines have helped greatly in understanding autoignition and the dependence of K on T_{comp15} in pre-mixed mixtures [3].

Thus, as the unburned mixture pressure increases with the temperature held constant (T_{comp15} decreases), the value of K decreases, and non-PRF fuels ($S > 0$) become relatively more resistant to autoignition compared to PRF. This observation is consistent with shock tube measurements of ignition delay for different types of fuels. The ignition delay, τ_i , can be expressed as a function of temperature, T , and pressure, P , in general as

$$\frac{\tau_i}{\tau_{i0}} = f(T) \left(\frac{P}{P_0} \right)^{-n} \quad (2)$$

Here, τ_{i0} is the ignition delay measured at a pressure of P_0 at a given temperature, T , and n is a constant. In IC engines at each point during the cycle the mixture will have a different value of τ_i . It is then hypothesized that auto-ignition occurs when the integral (Livengood-Wu integral) of the reciprocal of τ_i with time, t , reaches unity (Eq. 3).

$$I = \int_0^{te} \frac{dt}{\tau_i(P,T)} = 1 \quad (3)$$

Shock tube experiments show that in the region where ignition delay is low enough to contribute significantly to the Livengood-Wu integral, n is much smaller for non-PRF fuels compared to PRF [3]. In other words, τ_i decreases less for non-PRF fuels compared to PRF fuels as pressure increases for a fixed temperature.

Experimental studies in engines and vehicles also show that, within experimental error, if two fuels have the same RON and sensitivity, i.e. if they have the same OI, they will have the same autoignition phasing at all conditions. The simplest fuel system which can be used to match *both* RON and MON of a gasoline would require three components: iso-octane, n-heptane and toluene [14]. In any case since sensitivity is

zero for PRF fuels (while it is positive, around 10 for practical gasolines) the OI of a PRF will be the same at all values of K while OI will change as the value of K changes for practical fuels. Hence PRF are not appropriate as surrogates for practical gasolines.

A bi-component fuel system on which a linear rating scale can be based and whose autoignition chemistry is comparable to that of practical gasolines is a mixture of toluene and n-heptane [3]. It was suggested in [3] that if the scale in the anti-knock rating system was based on toluene and n-heptane, rather than PRF, a single "Toluene Number", TN might be sufficient to describe anti-knock behavior of real gasolines adequately at different conditions unlike RON or MON. The toluene number is the volume percent of toluene in the toluene/n-heptane mixture. More importantly, such a system would also enable fuels such as ethanol mixtures, with higher anti-knock quality than iso-octane, to be rated quantitatively.

Ricardo suggested using toluene/heptane mixtures for a rating scale to rank fuels even before the introduction of the octane scale. Of course, the ASTM procedure itself recognizes that there are fuels that are more resistant to knock than iso-octane and provides an extension of the scale. This cannot be truly quantitative in that such a fuel, with RON nominally greater than hundred, cannot be directly compared to a PRF since such a PRF does not exist. The extension of the RON scale provided by ASTM is an extrapolation based on using octane enhancers within the scale e.g. by adding a known amount of lead to a lower octane PRF, measuring the increase in RON and assuming that the same increase in RON will be obtained if the same amount of lead is added to a fuel of 100 RON. However we cannot be certain that such an extrapolation is valid quantitatively. For fuels with RON > 100, the actual head position in the CFR engine is a physical measurement but the RON number associated with this is based on an extrapolation. Hence we would argue that the current method to rate fuels that are more resistant to autoignition compared to isooctane is not quantitatively reliable.

This paper demonstrates how a practical gasoline can be rated in terms of TN in the existing RON (ASTM D2699) test in the CFR engine. Tests in a different single-cylinder engine demonstrate that the TRF fuel that matched the gasoline in the RON test describes the knocking behavior of the gasoline better than PRF fuels at different operating conditions.

2. EXPERIMENTAL DETAIL

Two different engines are used in this work.

In the standard RON test (ASTM D2699), the CFR head position is varied to adjust the compression ratio at the fixed operating condition till the test fuel knocks. There is a calibration curve generated by running the engine at the same conditions on different PRF fuels which relates the head position to the octane number. The RON of the test fuel can then be established using the calibration curve. In this work,

the CFR engine is used in the RON mode to generate such a calibration curve relating the head position to the toluene number (TN) for toluene reference fuels.

A second single-cylinder engine, with details as in Table 1, is used to validate the results. It was run with wide open throttle at different stoichiometric operating conditions listed in Table 2. Also listed in Table 2 are T_{comp15} , calculated using the ideal gas law using the volume when the pressure is 15 bar and the estimated number of moles in the cylinder. The results shown are the values using gasoline. Also shown is the K value calculated from knock measurements on different fuels as explained in Section 3.2. At each of these conditions different fuels were compared by running ignition timing sweeps and measuring the knock intensity.

Table 1. Details of single-cylinder engine used for validation

Number of Valves	4
Displacement, cm ³	499
Bore, mm	84
Stroke, mm	90
Compression Ratio, CR	10.5
Fuel Pump/ Range	External motor driven/ 4-15 MPa
Fuel Injector	Outwardly opening piezoelectric
Intake System	Conditioned air with external boost

Table 2. Operating conditions used in the validation experiments All experiments were run at wide open throttle

Cond.	Speed, RPM	Intake Temp. °C	Intake Pr. KPa abs.	T_{comp15} °C *	K value**
1	1500	27	160	600	
2	1500	27	100	660	-0.12
3	1500	75	160	645	
4	1500	75	100	720	0.26
5	3000	27	160	615	-0.65
6	3000	27	100	660	0.1
7	3000	75	100	705	0.26

*Calculated using measured average pressure curve and estimated temperature at the start of compression from $PV = mRT$, where m is the number of moles in the cylinder

**Calculated from knock measurements (where possible) for different fuels as explained in section 3.2

In-cylinder pressure is measured using Kistler pressure sensors and charge amplifiers along with a Leine and Linde 500 series crank angle encoder. The in-cylinder pressure signal was pegged during the intake stroke using a high speed intake pressure sensor located slightly upstream of the intake valve (another Kistler piezo resistive type absolute pressure sensor). We also check and confirm the pressure signal of both the intake and exhaust sensors with respect to the in-cylinder pressure sensor. We clean the surface of in-cylinder pressure sensor to avoid any drift effect. The in-cylinder pressure sensor is also water-cooled to reduce the possible thermal shock during the exhaust stroke. The sampling resolution was adjusted to 0.3° crank angle. Indicated mean effective pressure (IMEP) is