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## Nanoparticle emissions from spark ignition engines powered by n-butanol blends

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The paper reports on experimental investigation of the effects of introducing n-butanol as a fuel, neat and blended in varying concentrations with gasoline, for a small four-stroke engine typical for garden equipment.

Internal combustion engines are an established, economical, reliable and practical prime mover of most road vehicles as well as many kinds of mobile machinery and equipment. They have also become, in many regions, the dominant source of air pollution in urban areas, and considerable efforts have been and are devoted to reducing exhaust emissions levels. Another challenge facing contemporary engines is their reliance on petroleum based fuels, which are subject to concerns about greenhouse gas emissions, instability in petroleum prices, and petroleum resources depletion. Transition to carbon neutral alternative fuels, preferably derived from renewable resources, is therefore desired. In this work, the potential of n-butanol as a drop-in substitute for gasoline in small engines in hand-held and garden equipment has been investigated.

Historically, for automobile spark ignition engines, various alcohols and other oxygenated fuels have been considered as neat fuels or gasoline extenders. In the existing vehicle fleet, the use of ethanol, the most widely used alcohol, is practically limited to blends in small concentrations with gasoline, due to its hygroscopicity, agressivity, substantially lower heat content, and high latent heat. Butanol is an oxygenated fuel with no aromatic content and relatively low toxicity. It can be produced from biomass, and has higher energy density, lower latent heat, lower hydroscopicity and lower aggressivity than ethanol. Butanol has a research octane number of 94, comparable to ordinary gasoline. Its oxygen content is 21.6%, and viscosity somewhat higher compared to gasoline. Investigation on an experimental single-cylinder research engine have shown that the combustion of n-butanol in terms of ignition delay, combustion variability, knock susceptibility and thermal efficiency, has been found to be similar to that of gasoline [1].

To maintain a stoichiometric air-fuel ratio at a given air flow through the engine, approximately 22% more nbutanol (by volume) is needed relative to gasoline, compared to approximately 48% more for neat ethanol and 39% for E-85. Butanol is available in four isomers, out of which n-butanol and iso-butanol can be practically used as engine fuel. Of these, n-butanol (1-butanol), which can be produced by acetone-butanolethanol (ABE) fermentation from biomass [2], has been examined here in terms of startability, material compatibility, combustion stability and efficiency and exhaust emissions. While garden equipment may not be a large contributor to the overall regional emissions, its exhaust is emitted in the immediate proximity of its operator. Therefore, in addition to the regulated gaseous pollutants, exhaust particle size distributions have been assessed. Previous experiments on an automobile engine have shown that particle number decreases with addition of n-butanol, but is higher for n-butanol than for gasoline [3].

The experiments were performed on a Honda GX-31 single-cylinder, 0.031-liter, 1.1 kW, air-cooled, fourstroke spark ignition engine, with sealed, non-adjustable carburetor, and with non-adjustable ignition timing. The engine was coupled to a hydraulic dynamometer constructed in-house, comprising of a hydraulic motor delivering pressurized hydraulic fluid into a computer-controlled pressure valve, by which the pressure of the hydraulic fluid, and hence the motor torque, is controlled. The engine throttle was controlled manually. The engine was operated on ordinary automotive gasoline, on neat n-butanol, and on mixtures of 15%, 30%, 50%, 85% n-butanol with gasoline by volume.

The exhaust gases were routed into a pipe, from which a sample was taken for gaseous analyzers (heated flame ionization detector for hydrocarbons, non-dispersive infra-red analyzers for carbon monoxide and carbon dioxide, chemiluminescence analyzer for nitrogen oxides), another sample was routed into a partial flow dilution tunnel fabricated for this purpose, and the remaining gases were drawn, together with air surrounding the engine, by the laboratory ventilation duct. The flow in the partial flow dilution tunnel was maintained by an industrial vacuum cleaner operated at constant power. Diluted exhaust was sampled by a particle classifier / spectrometer (Engine Exhaust Particle Sizer, TSI).

The engine was operated at fourteen steady-state points at idle (around 1000 rpm, points 1 and 14) and at three loads – at full load and at 50% and 75% of maximum torque attained on gasoline – at 5000, 6000, 7000 and 8000 rpm. At lower rpm, the engine operation was unstable at higher loads, especially with higher shares of n-butanol. Each operating point was tuned by hand until desired engine rpm and torque was reached and was relatively stable, after which the engine remained at this point for at least one minute. The sequence of the operating points was the same for all fuels, but the length of the entire test has therefore varied among the fuels. Given the very small size of the engine, no additional preconditioning took place.

For selected regimes, the concentrations of hydrocarbons are shown in the upper part, and the concentrations of carbon monoxide in the lower part, of Fig. 1. Left part of the graphs shows concentrations during operation at 6000 rpm and 50%, 75% and 100% load, right part shows operation at 8000 rpm and 100% load, transition into idle and idle. The measured concentrations of notably hydrocarbons suggest that temporary combustion instabilities have occurred at apparently random places throughout the tests, notably at idle. The measured concentrations of carbon monoxide were not constant, but oscillated at a period on the order of ten seconds, suggesting that the operation of the engine was not constant. It appears that the concentrations of both hydrocarbons and carbon monoxide have decreased with increasing share of n-butanol in the fuel, which appears to be, to a great extent, a result of the relative enleanment of the air-fuel mixture; the gasoline operation was almost entirely fuel-rich.



Fig. 1: Concentrations of HC (top) and CO (bottom) for 6000 rpm and all loads (left) and 8000 rpm and 100% load, transition into idle and idle (right)

The particle size distribution are shown in Fig. 2 for the following points: 5000 rpm and 50% load (upper left) and 75% load (upper right), 6000 rpm and 75% load (lower left) and 8000 rpm and full load (lower right). From Fig. 2, a prevailing trend particle concentrations decreasing with increasing share of n-butanol is observed. The data is, however, far from being uniform and linear. At 5000 rpm and 50% load, the concentrations in all size categories decrease slightly from gasoline to 50% butanol (Bu50), after which there is a marked decrease for neat butanol (Bu100). For 85% butanol (Bu85), the concentrations were close to gasoline during one set of tests, and close to Bu100 during another set of tests (not shown). For 5000 rpm and 75% load, the concentrations decline sharply from 0% to 30% of butanol, with effects of higher shares of butanol not being clearly discernable. At 6000 rpm and 75% load, the effects are not consistent between the two tests on Bu85, and a decrease in nucleation mode but a large emergence of accumulation mode became apparent for Bu100. Similar observations happened at several other operating points. Relative increase in accumulation mode is also apparent for 8000 rpm and full load, where, in general, particle concentrations otherwise decrease with increasing share of n-butanol.



Fig. 2: Particle size distribution spectra for selected regimes

It can be argued that the reduction in particle emissions is, to a considerable extent, due to relative enleanment of the air-fuel mixture. In that case, if the engine can tolerate operating at leaner mixture, at least some reduction can be achieved by readjustment (or redesign or a non-reversible modification, as the carburetor does not allow for any adjustment) of the carburetor. If the engine cannot tolerate operating at leaner mixture, then provisions need to be made for increasing the quantity of fuel delivered when operating at least on higher concentrations of butanol in the fuel. In that case, it could be that considerable portion of the particle reduction would be lost due to relative enrichment (more precisely, due to adjustments to maintain a given air excess ratio). This was not, however, the goal of the study, which was directed primarily at a direct substitute of butanol or its blends in existing population of engines.

It should also be pointed out that n-butanol is used as a working fluid of condensation particle counters, therefore, unburned butanol in the exhaust may facilitate transformation of particles or nucleation during operation with high excess fuel.

This work represents the first insight into the effects of using n-butanol and its blends as a drop-in replacement fuel in small hand-held engines, and should be viewed as work in progress. The fraction of volatile particles, the particle chemical composition, and the relative toxicity of the produced particles have not been evaluated. In the case that higher shares of n-butanol would satisfy a myriad of practical considerations such as material compatibility, engine reliability and longevity, or economic aspects of butanol production, such additional studies would be a logical addition to the portfolio of knowledge.

References:

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