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REFLECTIONS ON COMBUSTION PRINCIPLES AS THEY RELATE TO A MISCELLANY OF PRACTICAL FUELS

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1. Introduction.

I recently visited one of the island communities to the north of Scotland in order to give advice on a project whereby waste cardboard is made into fuel briquettes. I thought it preferable if at all possible to conduct a combustion test on the briquettes during the few days that I was there. We therefore borrowed a pair of thermocouples and a multimeter from the local power station. We took those and four of the cardboard briquettes to the home of one of the local participants in the project, where we placed the four briquettes in a domestic stove and ignited them. We placed one thermocouple in the assembly of briquettes and the other in ice contained in a beaker such as one might serve tea or coffee in. We wrote down the e.m.f. displayed at the multimeter, at first every 30 seconds and later less frequently.

Experimental work does not come any cruder than that, yet we were able to glean a considerable amount of semi-quantitative information from the results, sufficient to answer relevant questions on the part of a potential purchaser of the briquettes. These results will be discussed later in the paper. We also consider the following and examine them according to combustion principles including the now very important one of possible carbon neutrality.

Solid biomass fuels Tyre-derived fuels Fuels for SI engines other than gasoline Fuels for CI engines other than diesel

Also:

Liquid explosives, in relation to the recent terrorist threat at Heathrow Airport.

2. Solid biomass fuels.

2.1. Preamble.

Wood has of course also found fuel use for a very long time: it was not until the 1890s that coal usage exceeded wood usage in the USA. In this paper however we consider wood and other cellulosic fuels in the modern sense, as 'biofuels', 'renewables' and 'carbon neutral' fuels. The last of these terms will be explained in full as it is the 'selling point' of biomass fuels in the modern world.

2.2. Carbon neutrality.

A fuel is said to be carbon neutral if its combustion makes no net contribution to the carbon dioxide content of the atmosphere. The ultimate in carbon neutrality is of course hydrogen fuel: the fuel itself contains no carbon and so obviously cannot produce carbon dioxide. That is why at some scenes of natural gas production, including the Miller Field in the North Sea, natural gas is reformed to hydrogen for subsequent use in power generation. When biomass fuels are considered the term carbon neutrality has to be thought about more carefully. A related calculation follows.

Suppose it is required to produce heat at 10 MW. How much carbon dioxide per hour will this produce:
(a) If the fuel is bituminous coal of carbon content 85% and calorific value 30 MJ kg⁻¹?
(b) If the fuel is wood waste of carbon content 55% and calorific value 17 MJ kg⁻¹?
Solution:
(a) Rate of requirement of fuel = (10⁷ J s⁻¹/30 × 10⁶ J kg⁻¹) × 3600 s hour⁻¹ = 1200 kg hour⁻¹ ⇒ 1020 kg hour⁻¹ of carbon burnt ⇒ 3740 kg CO₂
(b) Rate of requirement of fuel = (10⁷ J s⁻¹/17 × 10⁶ J kg⁻¹) × 3600 s hour⁻¹ = 2117 kg hour⁻¹ ⇒ 1164 kg hour⁻¹ of carbon burnt ⇒ 4270 kg CO₂

It is clear then that the carbon neutral fuel produces significantly more carbon dioxide than the conventional fuel per unit heat produced. The above calculation uses arbitrary though typical values for the quantities involved, and that wood fuel produces more carbon dioxide than the coal other things being equal is in fact a general result. Why then is the wood fuel to be preferred on carbon dioxide emission terms? Simply that unlike the carbon in coal the carbon in wood fuel was in the recent past carbon dioxide in the atmosphere, so when the wood is burnt it is simply being put back where it came from. To burn coal (or a petroleum based fuel) is to introduce carbon dioxide into the atmosphere. To burn wood is to put carbon dioxide back into the atmosphere after its

uptake by the wood in growth. A fully grown tree absorbs about 25 kg per year from the atmosphere which is returned there when the dead wood is burnt.

In order that non-renewables obligations are met, biomass and coal might be co-fired. Imagine that in our above example where the total heat release rate is 10 MW that the coal-derived CO_2 has to be reduced by 5% to comply with local non-renewables requirements. The calculation is in the box below.

Coal-derived CO₂ to be reduced from 3740 kg per hour to 3553 kg per hour. Carbon burnt to be reduced from 1020 to 969 kg per hour. Coal burnt reduced from 1200 to 1140 kg per hour. Heat to be provided by the biomass per hour = 60×30 MJ = 1800 MJ requiring 1800/17 = 106 kg biomass releasing ($106 \times 0.55 \times 44/12$) kg CO₂ = 214 kg CO₂.

Total CO₂ release per hour = (3553 + 214) kg = 3766 kg

There is therefore a slight increase (0.7 of 1%) of total CO_2 due to partial substitution of the biomass for coal but a reduction as required of 5% of the CO_2 from coal which of course is not carbon neutral.

In relation to the above calculation we should remember that the wood will contain a significant amount of oxygen in its organic structure, and the coal will contain some, perhaps 2-3% for the coal in the example. The carbon dioxide produced will therefore not arise solely from combustion: part of it will be from devolatilisation and in such carbon dioxide the oxygen atoms come not from the air oxidant but from the oxygen content of the fuel. The most helpful way to view this is that the fuel is already, before combustion, partly oxidised. This makes no difference to the calculation of carbon dioxide emissions: it does make a difference when the air requirement to burn a fuel is being calculated and the oxygen content of the fuel must be deducted from the oxygen theoretically required to oxidise the carbon content and the hydrogen content of the fuel. This is standard practice in fuel technology, e.g., in boiler operation. Devolatilisation products of solid fuels include methane and other light hydrocarbons, CO, tars and oils which are of course flammable. In a biomass fuel the combustible devolatilisation products, a.k.a. 'volatiles', will account for well over half of the heat value. That not accounted for by the volatiles is of course in the residual char. This is considered more fully in the next section.

2.3. Devolatilisation.

As stated in the previous paragraph, flammable volatiles are tars/oils, light hydrocarbons such as methane, ethylene and ethane, and carbon monoxide. Non-flammable volatiles,

much less abundantly yielded than the flammable ones, are carbon dioxide and water. When a high-volatile fuel is burnt devolatilisation occurs concurrently with combustion; heat from the burning of the flammable volatiles feeds back to the remaining solid to promote further devolatilisation. The extent of devolatilisation as a function of temperature can be represented by suitable Arrhenius parameters. Devolatilisation involves many reactions within the solid fuel structure including methyl group removal, decarboxylation and cleavage of parts of the organic structure. The overall heat effect is the resultant of the individual heat effects of all such reactions occurring and might be net positive (endothermic) or net negative (exothermic). It will however, regardless of its sign in the thermodynamic sense, be small in comparison with the heat of subsequent combustion of volatiles so released. Whether carbon is burnt as a volatile, e.g., a tar fragment, or as part of the residual solid is of course irrelevant to the matter of carbon neutrality. Biomass is always higher than coals in volatile matter; even so, low-rank coals such as the brown coals of the Latrobe Valley in Australia and the similar ones in the deposit which straddles North Dakota USA and Saskatchewan Canada lose 50% or more of their weight by devolatilisation at temperatures in the neighbourhood of 1000° C. The extent of devolatilisation has a moderate dependence on the heating rate as has the distribution of products, e.g., of oxygen in the fuel between CO and CO₂ in the volatiles.

2.4. Control of burning of a high-volatile fuel by ventilation.

We return here to the highly primitive test on the combustion of cardboard briquettes referred to in the introduction. These were burnt in a domestic stove where ideally one would require slow, flameless burning. Flameless burning normally occurs only with fuels already denuded of volatiles such as coke and charcoal. In our tests however the cardboard briquettes did burn flamelessly and the four briquettes, each weighing about 180g, were still burning when temperature measurement ceased after an hour. The reason clearly was under-ventilation resulting in escape of the volatiles with the smoke instead of their combustion in the gas phase. This happened by chance in our test but indicates that a high-volatile fuel which with full ventilation would burn with a vigorous flame can be made to burn like charcoal if ventilation is controlled. Of course, escape of flammable volatiles represents wastage of energy and an enterprise using such fuel on a large scale, e.g., for steam raising (as is proposed for the cardboard fuel which was being examined) would need to avoid this. It is however doubtful whether such 'uneconomic' burning would matter on the scale of a domestic fire, although one might want to consider the release of unburnt hydrocarbons into the atmosphere which such burning would result in.

There were considerable temperature variations in the burning assembly of briquettes. About half way through the test the measuring thermocouple was reading just over 500° C and was moved to a part of the fuel bed glowing red whereupon the reading rose to 800° C. Further deliberations on this point form Appendix 1 of the paper.

2.5. Some examples of biomass fuels.

Examples of these are given briefly below with at least one country of usage in brackets in each case. Biomass fuels include: sugar cane residue (a.k.a. bagasse, Australia and Fiji); citrus peel (Florida USA); coconut waste (The Philippines); coffee waste (El Salvador); waste from maize, rice and mustard collectively called crop waste (India); forest thinnings, obtained from removal of wood from trees to reduce the fire susceptibility of the forest of which they are a part (Colorado USA); Mangrove, a tropical shrub (Malaysia and other far East countries); straw (Ely, UK and also Croatia) and Teak waste (Thailand). The points made above about solid biomass fuels generally, including the very important one of carbon neutrality, apply to all of these.

3. Tyre-derived fuel (TDF).

3.1. Preamble.

There are huge amounts of used tyres requiring disposal at any one time. As a rule of thumb in 'developed' countries the number of new tyres sold each year is roughly equal to the population, and of course one new tyre bought means one old tyre needing disposal. Fuel use of tyres is fairly widely practised and has been the subject of a recent critique which will be discussed below. The calorific value of the rubber component of tyres is much higher than that of biomass, up to about 40 MJ kg⁻¹. This is higher than the calorific values of even higher rank, low-ash coals which very seldom if ever exceed 35 MJ kg⁻¹.

3.2. Summary of a recent critique of fuel use of waste tyres [2]. The salient points are as follows.

(a) Tyre waste fuel can reasonably be viewed as carbon neutral in that rubber is a vegetable material.

(b) The alternative to fuel uses of tyre waste are two: simple incineration and landfill. In the former case there is carbon dioxide release without any return on the heat. In the second decomposition over time will lead to the release of methane, which is a much more powerful greenhouse gas than carbon dioxide. (Each of these points also applies to biomass fuels such as wood which were considered in the previous part of the paper.)

(c) Tyres tend to burn smokily and the combustion therefore needs to be over-ventilated to minimise smoke. This reduces the combustion temperature.

3.3. Selected scenes of significant usage of tyre waste as fuel.

These include two US electricity utilities: Bay Front, Wisconsin and Marmora NJ. Currently the only plant in Europe which uses waste tyres as a fuel for the generation of electricity is Elm Energy and Recycling in the UK. It has the capacity to burn 100000 tonne of tyres per year and significant power for sale to the grid is possible. A 'back-of-an-envelope' calculation of the performance of this facility forms Appendix 2.

4. Fuels for SI engines other than gasoline.

4.1. Introduction.

'Gasoline' can mean mineral gasoline, obtained by refining crude oil or previously retorted shale oil, or manufactured gasoline obtained from coal or other feedstock via synthesis gas or by hydrogenation. The benchmark hydrocarbon against which gasolines are compared in performance terms is of course iso-octane, C_8H_{18} . Natural gas, predominantly methane, is widely used in one of two forms, to be more fully described below, in engines which would otherwise have used gasoline, as is the oxygenated compound methanol.

4.2. Carburetion in SI engines and engine knock.

The working substance in such an engine is of course air, and carburetion takes place in order to provide chemical heat release at the stage in the cycle where it is needed. In a SI engine, ignition is required at a precise stage: ignition ahead of that causes knock, and fuels for SI engines are assessed on the basis of resistance to knock expressed as the octane number. Fuels for SI engines need to be quality controlled at the refining stage for octane rating. Gasoline is the first liquid to come over in refining. If too wide a cut in temperature terms is taken so that the distillate also takes in some of the higher boiling fraction called naphtha it will have too low an octane rating, that is, too low a resistance to knock. This is entirely consistent with the principle that longer hydrocarbon chain lengths signify higher reactivity. Of course, there is no precise upper limit on the fractionation temperature range required to obtain a gasoline of good octane rating or threshold temperature marking the cessation of gasoline production and the commencement of napththa production. Such matters depend on the composition of the crude oil undergoing distillation, and with some crudes a gasoline fraction obtained even across quite a narrow cut will need to be blended with additives, notably aromatics and branched-chain alkanes, to bring the octane number to the required value.

4.3. Methane as a motor fuel.

Currently unleaded gasoline of research octane number (RON) 95 is widely sold on the forecourts of the world, and one pays extra for gasoline of RON 98. Methane has a RON of 120, significantly better than even the more expensive gasoline. The high resistance to knock of methane is due to its low reactivity; being C_1 it is the least reactive alkane and it is this property which makes it suitable for use in vehicles with SI engines. Spark ignition engines which run on methane are well established, there being many makes and models of car designed to run on compressed natural gas. This of course is simply methane gas a long way above its critical temperature stored under pressure in the vehicle for supply to the engine. As an alternative, the gas can be carried as liquefied natural gas (LNG) and evaporated on its way to the engine. The fuel tank in such a vehicle comprises two membranes with an evacuated space between them.

Whenever a new fuel for motor transport is introduced the confidence not only of the consumer but also of the forecourt operators, whose collective influence is considerable,

has to be won. The author's view is that both LPG and methanol as fuels for vehicles were at least initially adversely affected by lack of such confidence. Consequently much R&D went into making refuelling facilities for vehicles powered by LNG not only user-friendly but familiar so that 'filling with LNG' is, to the motorist, no different from 'filling with petrol'.

4.4. Methanol as a motor fuel.

It very often happens that when an oxygenated hydrocarbon is formed from a simple hydrocarbon the former is more reactive than the latter. Accordingly, methanol is more reactive than methane and in engine use has a lower octane rating than methane. The RON of methanol is actually about 108, better than that of regular gasolines. Of course, methanol and gasoline can be blended to give a target octane number.

Natural gas is not of course carbon neutral, any more than petrol from crude oil or from shale is. Whether methanol is carbon neutral depends upon its origin. If it is made from coal or natural gas via synthesis gas it is not carbon neutral. However methanol (one synonym for which is 'wood alcohol') made from the decomposition of wood is carbon neutral if subsequently used as a fuel. Perhaps more importantly, it is also carbon neutral if it is made from synthesis gas produced from biomass (any of the examples given in Section 2.5, and there are many more!) instead of from coal or natural gas. The carbon neutrality of methanol is therefore ambiguous, having a dependence on the method of manufacture.

4.5. Ethanol as motor fuel.

We fasten on just one example: E85, a blend of ethanol (85%) and gasoline (15%). It is routinely available in certain US states including Illinois and has a research octane number of 105. Table 1 below summarises what has been said about fuels for SI engines in terms of octane number and makes some additional comments.

Table 1.	Octane	ratings	of fuels	for	SI	engines.
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Fuel	RON	Comments
Gasoline as sold	95 or 98, the latter	Adjustment of the octane rating by control of
at most retail	more expensive.	the temperature range of the 'cut' and by
outlets.		inclusion of octane enhancers.
		Gasoline is obtainable from shale and tar
		sands as well as from crude oil.

. . . cont.

Natural gas.	120	Higher octane number by reason of lower	
		intrinsic reactivity and therefore lower	
		propensity to knock. Sold in compressed form	
		and (at the present time less widely)	
		in liquefied form.	
Methanol.	108	Ambiguous in carbon neutrality terms.	
E85.	105	A Flexible-fuel vehicle (FFV) can run on any	
		blend of fuel in the composition range from	
		pure gasoline to E85. Most of the	
		manufacturers which supply cars to the US	
		make FFV versions of at least some of their	
		models available.	

5. Fuels for compression ignition (CI) engines other than diesel.

5.1. Introduction.

A fuel for use in such an engine has to be capable of ignition without a spark, the very event which must be avoided in SI engines. The merit of a particular fuel for a CI engine is assessed by its resistance to ignition delay, and the benchmark hydrocarbon is n-cetane $C_{16}H_{34}$ which by definition has a cetane number of 100. In contrast to octane numbers for commercial gasolines, which are close to 100, cetane numbers for routinely available diesel fuels will be in the neighbourhood of 50. Whereas low reactivity favours a good octane rating high reactivity favours a good cetane rating. This point will be enlarged upon when biodiesels are discussed below.

5.2 Biodiesel fuels.

These are plant oils used as substitutes for diesel in compression-ignition engines and obtained from the crushing of seeds. Initial development in this topic was in India in the 1980s where 'Honne oil' was investigated as a diesel substitute with promising results. Currently, widely used plant oils for fuel purposes include Soybean oil and Rapeseed. Being obtained directly from plants, biodiesels are of course carbon neutral.

Equivalence in reactivity terms of biodiesel and mineral diesel requires that the oxygenated organics in the biodiesel, in particular the esters, have a combustion reactivity comparable to that of unsubstituted hydrocarbons up to about the C_{22} in the mineral diesel. The plant oil without any form of processing is sometimes suitable for use as a biodiesel otherwise its properties, not only the chemical reactivity but also certain physical properties including viscosity, can be modified by esterification of the carboxylic acids present in the oil with methanol. The extent of esterification can be matched to a desired cetane number. Alternatively, the plant oils can simply be blended with esters such as methyl oleate, methyl stearate and methyl palmitate (the methyl esters of three of the acids likely to be present, for example, in Soybean oil) to adjust the cetane number whereupon a correlation as simple as:

cetane number = (cetane number of the unprocessed fuel) + Z x (% esters)

where Z is a constant, applies.

Table 2 below identifies some sources of biodiesel and gives brief descriptions of them. It is important to realise that plant oils are not 'fine chemicals' of precise composition and that even oils from the same plant vary in properties. Just as when a distillate in the diesel boiling range is obtained its cetane number might have to be adjusted by incorporation of a cetane enhancer such as isopropyl nitrate, so when a plant-derived oil is obtained its cetane number might have to be adjusted by raising its ester content as previously described. Such processing will also affect the viscosity. Viscosity alone can be adjusted by incorporation of small amounts of animal fat (tallow).

Source.	Description.		
Hemp.	Oil from hemp a possible biodiesel. Because of possible misuse		
	of hemp to make a narcotic drug its cultivation requires a		
	licence in many parts of the world including the EU		
	and the US.		
Jatropha tree.	'Beans' from the Jatropha tree, which occurs in a number of		
	African countries, produce oil at 40% yield which can be made		
	into biodiesel. The Jatropha tree can grow under quite poor		
	conditions of irrigation and nutriment supply.		
Oil palm.	Palm oil currently being widely produced internationally as a		
	component of biodiesel. The world's major producer at present		
	is Malaysia where \approx 9 million tonne of the material are being		
	produced each year. Fluctuations in the demand for palm oil		
	are due largely to the competition from soybean oil.		
Rapeseed.	A major source of biodiesel, 10 million tonnes grown annually		
	in the EU.		
Soybean.	Soybean oil is currently the most expensive of the liquids used		
	as a basis for biodiesel but has some advantages over its		
	competitors one being that Soybean-derived biodiesel is		
	particularly suitable for use in colder regions.		
Straight vegetable oil	Vegetable oil sold for culinary purposes requiring no stringent		
(SVO).	viscosity specification. Vehicles can be adapted to run on this		
	sort of oil in unused or in waste form, though in the latter case		
	filtration is necessary. The adaptations to make a diesel vehicle		
	run reliably on SVO are fairly major.		
	Vehicular use of SVO purchased for kitchen use is illegal in		
	many countries including the UK.		

Table 2. Sources of biodiesel.

5.3. Simple alcohols in CI engines.

The cetane numbers of pure methanol and ethanol are about 3 and 8 respectively and this clearly precludes their use in CI engines. There are however blends of diesel and alcohol which can be used. C_1 and C_2 compounds even when oxygenated would be expected to be less reactive than the C_{16+} ingredients of diesel and therefore to blend a diesel with methanol or ethanol is to lower its cetane number. However, to use ethanol as a blendstock is to make the fuel 'greener' as that component of it, unlike the diesel to which it is added, is carbon neutral. Experience in a number of countries is that ethanol at up to 15% by weight blended with mineral diesel will not cause the cetane number to move outside the range of values suitable for diesel cars and commercials and it will, as noted, have greenhouse benefits.

<u>6. Liquid explosives with special reference to the terrorist scare in August 2006.</u><u>6.1. Introduction.</u>

In August 2006 a plot to blow up an airliner in flight by means of a liquid explosive was foiled. In the days that followed there was much media discussion of 'liquid explosives' to which the writer of this article contributed. He also wrote for one of the professional periodicals the following piece [3]:

The author responded to many media enquiries concerning the 'liquid bomb' threat at Heathrow in August 2006 and broadcast on the topic on UK and Australian radio. He has had some feedback from someone professionally involved in explosives and would like to put his responses to that in the public domain by means of this note. Some commercial explosives are of course liquids, probably the best known being nitroglycerine a.k.a. 'blasting oil'. However, the term liquid explosive can be extended to a pair of liquids which:

(a) will react together exothermically, and(b) in so doing will create an overpressure.

Clearly in such a two-component explosive one liquid will be the fuel and one the oxidiser. The former might for example be mineral turpentine, the latter peroxide or bleach. Development work would establish what size and shape of vessel containing the mixed liquids would lead to an explosion with overpressure.

It is also relevant to note that detonation is not essential for there to be an overpressure. In detonation, of course, speeds are supersonic. However, a deflagration, in which speeds are sub-sonic, can have sufficient overpressure to cause death or injury to persons as well as damage to structures (e.g., an aeroplane fuselage). Hydrocarbon accidents in the refining and processing industries in fact lead to deflagration, not to detonation.

6.2. The inevitability of explosion under adiabatic or close-to-adiabatic conditions. We should note in addition to what is in italics above that any exothermic reaction has the potential to ignite: it is <u>certain</u> to ignite if it takes place under adiabatic conditions.

This brings us to the distinction between 'explosion' and 'ignition', if indeed there be a non-arbitrary distinction between them. For the purposes of this debate we can equate the two, as any ignition of interest will involve gas release with pressure effects. The only ignition known to the author which does not involve product gases is that of certain pyrotechnics, which are said to be 'gasless', and these are such an obscure example that maybe the exception proves the rule!

7. Concluding remarks.

The author is now at a stage where, to coin a euphemism, he seems to be older than the majority of his colleagues. He has had tens of years of teaching, research, writing and consultancy in fuels and combustion and has found that the fundamental principles of thermal sciences have become not less but more helpful and appealing to him. When confronted with a new problem he invokes such principles with more confidence than he might have done at a much earlier career stage, often mentally by-passing more advanced approaches which he has encountered. This paper in a sense invokes a 'return to simplicity' in the approach to particular issues in fuels and combustion including those discussed in the text.

8. References.

- [1] Holman J.P. 'Heat Transfer' McGraw-Hill, NY, any available edition.
- [2] Jones J.C. Fuel Processing Technology <u>87</u> 753 (2006).
- [3] Jones J.C. 'On the use of the term "liquid explosive" International Fire Fighter, expected to appear in the November 2006 issue.

9. Appendices.

Appendix 1.

Semi-quantitative calculations apropos of the red glow observed in the combustion test.

Using Wien's displacement law [1]:

$$(\lambda T)_{max} = 2897.8 \ \mu m \ K$$

 $(\lambda T)_{max}$ being the product of the wavelength and temperature corresponding to the maximum emissive power. Now for combustion at 800°C (1073 K) the calculated wavelength is 2.7 µm which is outside the visible range and at the low wavelength end of the infra-red range. Red light, at the high wavelength end of the visible range, has a wavelength of 0.75 µm. Light of this sort of wavelength is emitted only at a rate of the order of 10 W: this can be reliably inferred from the very dim illumination of the stove interior caused by the glow, less than that which a domestic light bulb of say 40 W would have provided. The glow would have been a very small proportion of the total radiation from the fuel bed, most of which was at wavelengths higher than those of the visible range. Where the bed was at 500°C (773 K) the value of λ from Wien's displacement law is 3.7 µm, also in the infra-red range.

Appendix 2.

Electrical power from the combustion of tyre waste at the Elm Energy and Recycling

 $\frac{\text{plant.}}{100000 \text{ tonne}} = 10^8 \text{ kg}$ Taking the calorific value to be about 35 MJ kg⁻¹ Heat released in a year = 3.5 x 10¹⁵ J x 10⁸ W of heat Taking the generation using a Rankine cycle to be about 30% efficient, electricity is produced at 33 MW