Fuel modification based on some metals compounds and their environmental impact

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Abstract
The history of fuel additive use reflects the interplay between chemistry, technology and public health concerns related to environmental effects. Decisions to use specific type of chemical modification during combustion process have been made in the absence of toxicological data on health and environmental effects or exposure. The influence of these important issues has extended globally, and the effects of various compositions impact for decades after the removal of these compounds. Fuel modifications are widely used for petrol, oil and solid fuels. According to market screening and literature review, additives containing some dangerous compounds are still in used today. $Pb(C_2H_5)_4$ was used for long time as fuel additive and is still used as an additive in some grades of aviation gasoline, and in some developing countries. It is obvious that additives containing copper, lead and cerium should be replaced by organic substitutes or inorganic oxidizers during combustion processes.

Key words: metals, combustion, pollution, air, solid, additives, coal, petrol

Introduction

Catalytic additives make fuel burn more efficiently and they are largely linked with liquid and solid fuels in the interest of environmental protection by limiting emission during combustion process. Fuel mixtures have a broader impact of being able to change, alter or enhance specific attributes of a fuel whether liquid, solid or gas. This paper covers fuel additives containing lead and cerium for liquid fuels; and copper and chloride for solid fuels. These additives are environmentally unfriendly and have the potential to pose strong environmental and health implications. Fuel additives have been developed to increase combustion rates, to enable fuels work under extreme temperatures, reduce harmful emissions like particulate matter from Diesel engines, and many more [1]. Over the past decades, various compounds and blends have been engineered to create better fuels for industries and commercial purposes [2-4]. Problems common to antiknock agents are low effectiveness, uneconomical and inconvenience. The organometallic gasoline antiknock agents have high efficiency but they are not environmentally friendly. Tetraethyl lead (TEL) used for many years have been prohibited because of the toxicity of lead [5]; however, cerium oxide, which is also not environmentally friendly is still allowed as fuel additive [1]. Nowadays cerium oxide is widely used as a fuel additive for the elimination of toxic exhaust emission from vehicles. The ceria nanoparticles ($nCeO_2$) contained in fuel can act as an oxidative active component, working as an oxygen store by release of oxygen in the presence of reductive gases, and removal of oxygen by interaction with oxidizing species. The mentioned catalytic activity may occur when ceria is added as an additive to fuel, for example diesel or petrol. However, in order for this effect to be useful the cerium oxide must be of a particle size small enough to remain in a stable dispersion in the fuel. Unfortunately, the toxicological information of ceria to environment remains largely unknown and it may have unique toxic properties [6, 7]. Once in the soil, ceria tends to accumulate in suspensions due to natural organic matter reducing its aggregation capability. Cerium in the soil may be absorbed into vegetation or contaminate water and soil. As a result, Cerium has the potential to enter the food chain.

Fuel additives should promote the combustion of fuel and do not cause environmental pollution by itself and by side reaction. Air pollution is a major problem in the world and, in particular, in those countries that utilize coal as a major fuel source for industrial boilers or furnaces, such as Poland. The emissions of airborne pollutants form a lot of chemical compounds on a large scale are threatening the ecological balance. Coal is the main energy source for generating electricity in Poland and many other countries. If the efficiency of combustion of coal could be improved even by only a small percentage, the savings in quantity of fuel per annum will be significant. Partial or incomplete combustion of fossil fuels results in atmospheric pollutants, such as black carbon, polycyclic aromatic hydrocarbons (PAH), polychlorinated dibenzo-p-dioxin and furans (PCDD/F) [8]. *Copper* is the most effective catalyst for *dioxin* formation, but copper compounds are very popular active components of

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coal additives. The fossil fuels generate black carbon during the combustion process and are released as emissions of particulate matter (PM) [9]. Black carbon emitted from furnaces and vehicles is a short term climate threat, active for a few days in the atmosphere and a few months on snow and ice surface. Over these short times black carbon is an important contributor to global climate warming [10]. The purpose of this paper is to describe the potential negative environmental effects of certain fuel additives and suggest possibilities of replacing lead (which is still used in aviation fuel) cerium and copper occurred in fuels.

The role of fuel additives and their environmental impact

Numerous individual boilers and furnaces need to be considered when attempting to solve a combustion problem by the use of chemical additives. The most crucial are the chemical composition, quantity and mode of application of the fuels additive, the nature of the fuel, and parameters affected by furnace design and operation such as residence times, temperature gradients and the efficiency of air, fuel and additive interactions. The term *nature* of the additive includes particle size, chemical and physical form and thermal history as well as composition. All of these factors affect additive performance and should be considered.

The additives for solid fuels

Combustion of solid materials in small heating units provides a lot of pollutants to the atmosphere (Figure 1). Additives for solid fuels improve combustion of fossil fuels and wood.

Figure 1. Coal combustion and air pollution (fot. M. Chyc)

The use of fuel additives reduce exhaust emissions of particulate matter (PM), polycyclic aromatic hydrocarbons (PAH) and other harmful substances. As concerns for the environment grows worldwide, the safety of various chemical products used as fuel additive concentrates is becoming more of a concern for manufacturers and distributors. It is of paramount importance for the industry to examine the use of functional fuel additive components that are also compatible with environmental issues. In particular, substances that are persistent in the environment [11], bioaccumulative [12], and toxic may be banned from use (e.g. Stockholm Convention), but they are generated during combustion with additives. Hence, there is the need for fuel additive compositions containing such additive compositions that incorporate more environmentally acceptable materials. Fuel additives are useful for all types of ovens and improve their efficiency. There are several solid fuel additives to improve efficiency of coal combustion process in power plant and domestic ovens. Since power plants consume huge quantities of coal, even small efficiency improvement can imply significant savings. It is well recognized that the burning of many fuel types cause troublesome fireside-deposits, such as black carbon, slag, creosote which are difficult to remove. When burned, all but a few fuels have solid residues, commonly called deposit. The management of this ash is a major consideration in the design and operations of all combustion systems. A portion of the ash goes with the smoke to particulate collection equipment (fly ash). Another portion of the ash is discharged through the bottom of the furnace (bottom ash), friable residue (dry bottom furnace), and flowing residue (cyclone furnace and wet-bottom furnace). The remainder of the ash collects on the fireside surfaces as either a loosely adhering material that is easily removed by conventional mechanical ways or a strongly adhering, tenacious deposit that resists the typical cleaning procedures. Wood burning stoves have made a huge rebirth of use in the home. People nowadays use these stoves, which are well-known as "air-tight stoves" to supplement if not heat their buildings totally during the winter time. Coal deposit is a black carbon-like material that builds up on ducts, and if allowed to go unchecked, can cause several problems e.g. chimney fire. First observation, after creosote formation a furnace may not work correctly. Second observation, creosote deposits can eventually build up to the point where they will burn and cause a fire. Progress in the design of these facilities has done little to prevent the formation of deposit within the stove flue ducts. The most effective black carbon removing materials are based upon inorganic chloride salts, especially NaCl and other chlorides, such as NH_4Cl, $CuCl_2$ and $FeCl_3$. When an additional metallic chloride is used in combination with NaCl, the presence of the additional metal chlorides increases the soot removing effect and promotes more effective its removal. However, NaCl and the other metallic chlorides are quite corrosive to the ferrous alloys commonly used in flue ducts of typical air-tight stoves. The inorganic chlorides and sulphates are well-known as progressive corrosion compounds on iron based metal, especially in higher temperatures [13]. Even stainless steel pipe is pitted and severely corroded by use of metallic chlorides and sulphates black carbon and creosote removers. The pit-type corrosion is formed by small electrochemical cells formed on the surface of the metal material. Despite almost all available black carbon removers contain corrosive chemical compounds. Figure 2 shows energy dispersive spectroscopy (EDX) spectrum of popular solid fuel additive, chemical composition of it described in polish patent PL207482.

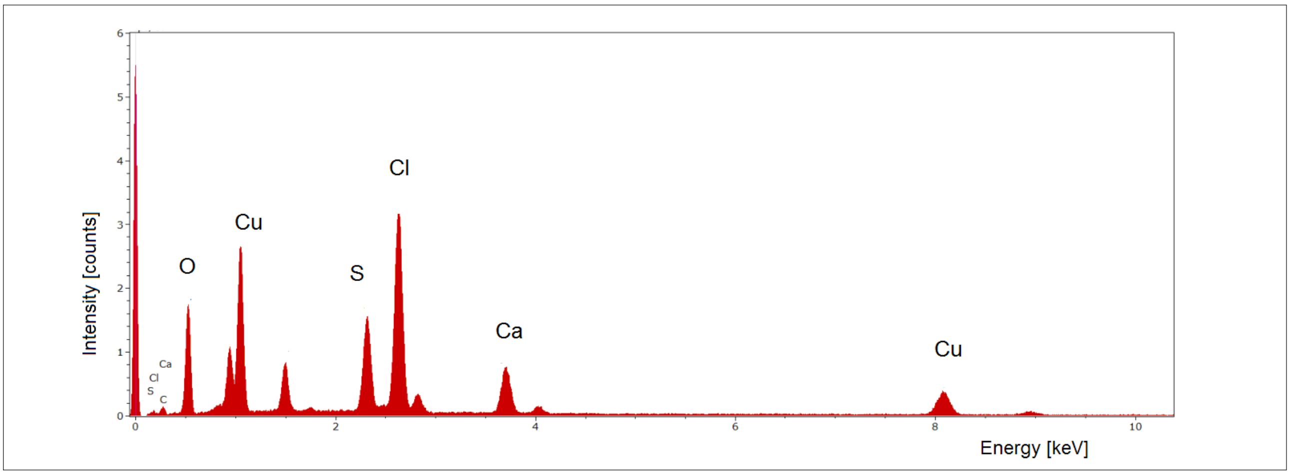

Figure 2. EDX spectrum of popular solid fuel additive

Presented additive (Figure 2) is available on European market. According to patent description the additive contains: $CuSO_4{\cdot}5H_2O$, NH_4Cl, NaCl. These compounds are corrosive and may have negative environmental impact because of copper and chlorine content promote xenobiotic synthesis [14].

The additives for liquid fuels

Liquid fuel is a mixture of hydrocarbons containing a variety of organic compounds and elements including sulphur and carbon. Some of these compounds or elements are undesirable when emitted into the environment. The combustion of fuel is associated with the formation and emission of sulphur compounds (SO_x), nitrogen compounds (NO_x), carbon monoxide (CO), carbon dioxide (CO_2), fly ash, black carbon, and unburnt hydrocarbons. Liquid fuels, by nature, are volatile and produce volatile organic compounds (VOC), as emitted especially by gasoline. These compounds that contaminate the environment are formed in internal combustion engines by the oxidation of hydrocarbon-type fuels. These compounds are known to cause health problems and contribute to global climate change. In addition, a serious problem associated with liquid fuel combustion is the presence of polycyclic aromatic hydrocarbons (PAH). Several PAH are known to be mutagenic and/or potentially carcinogenic toward humans [15].

As a result of the growing concerns of these emissions and their negative human and environmental impacts, fuel additives have been used to reduce emission levels. Liquid fuel additives are chemical compounds added in very small quantities (in the order of mg of additives per kg of fuel) to liquid base fuel to modify or create certain fuel properties. Some examples of these additives include oxygenates, ethers, antioxidants (stabilizers), antiknock agents, fuel dyes, metal deactivators, corrosion inhibitors, detergents, and many more. Fuel additives have been developed to increase combustion rates, as antioxidants, to effect burn rates, to enable fuels to work under extreme temperatures, reduce harmful emissions and overall increase fuel quality. According to Marsch [16], fuel additives suppress particulate emissions in liquid fuels via four methods of action:

1. inhibit particle formation chemistry;
2. promote fuel atomization, which enhances vaporization and premixing;
3. increase ignition delay (flame lift-off distance), which allows more time for vaporization and premixing before the flame is established; and
4. catalytically enhance oxidation of carbon particles.

Despite the positive role of using fuel additives to increase fuel properties and reduce emissions, additives such as lead and cerium based fuel additives could have serious human and environmental effects. For example, lead pollution from engine exhaust is dispersed into the air and easily inhaled. Long term exposure to leads could lead to acute lead poisoning because lead is a toxic metal that accumulates in the body and is associated with neurotoxic effects such as low IQ and antisocial behaviour. It has particularly harmful effects on children [17]. These concerns eventually led to the ban on tetraethyl lead (TEL) in automobile gasoline in many countries.

While the fate of ceria fuel additive, as a nanoparticle is debatable, controlled combustion studies have shown that cerium based fuel additive decreases particulate and NO_x emissions but dramatically increasing ultrafine particles (diameter <100 nm in size), carbon oxide, hydrocarbon content and volatile organic compounds. Despite the high efficiency in trapping PM, a small amount of cerium is emitted in the exhaust in the particulate phase. Therefore, the environmental, ecological and health effects associated with the use of $nCeO_2$ as a fuel additive is not known and need to be further examined [18].

Negative aspect of using fuel additives

Solid fuels

During fossil fuel combustion, pollutants come out of the smoke stack as flue gas. The opacity of the plume of a smoke stacks says a lot about how clean the plant is burning its coal. Many different types of clean coal technology are used to reduce the black smoke coming from smoke stacks polluting air and soil with CO, NO_x and SO_x and even heavy metals. That is way some customers use fuel modifiers. Chemical modifiers used during combustion are inorganic mixtures containing inorganic compounds mainly copper. Some of them promote synthesis of polychlorinated dioxins (PCDD) and polychlorinated furans (PCDF) [14]. Dioxins are highly toxic and can cause reproductive and developmental problems, damage the immune system, interfere with hormones and also cause cancer. Once dioxins enter the body, they last a long time because of their chemical stability and their ability to be absorbed by fatty tissue, where they are then stored in the body. The higher an animal is in the food chain, the higher the concentration of PCDD/F. Reducing dioxin exposure is an important public health goal for disease prevention. The combination of persistence, bioaccumulation and toxicity identifies a substance to be an environmental pollutant. Consequently, further knowledge of emission sources, formation, environmental distribution and biological effects of dioxins and related compounds is required. The environmental significance of these compounds is emphasized by their inclusion in the Stockholm convention of persistent organic pollutants, which is an agreement signed by more than 150 nations to ban the production, use and trade of ten commercially manufactured substances, and to minimize, or if possible eliminate, emissions of unintentionally formed by-products Low temperature catalytic ways proceed by heterogeneous gas/solid reactions subdivided into de novo synthesis [19]. Starting from macro-molecular carbon or from polycyclic aromatic hydrocarbons compounds, precursor pathways starting from organic molecules similar to PCDF and PCDD, including mainly chlorobenzenes (PBz), chlorophenols (PCP), chlorinated naphthalenes, polychlorinated biphenyls (PCB). The synthesis ways proceed at the surface of black carbon and fuel and on ash, catalysed by various transition metal oxides and salts. PCDFs are released directly by catalytic oxidation of suitable structures in the carbon matrix, while PCDDs form by both paths, i.e. the condensation of precursor compounds and direct release from the carbon black and *de novo* pathway (Figure 3). The catalytic capacity of synthesis PCDF and PCDD of various chlorinated aromatic compounds shows (1) [19]:

$$CuCl_2{\cdot}2H_2O > Cu_2(OH)_3Cl > FeCl_3{\cdot}6H_2O > FeCl_2{\cdot}4H_2O > CuO > Fe_2O_3 > PbCl_2 > Blank > ZnCl_2 > PbO > ZnO \qquad (1)$$

Because of dioxin pollution during the combustion of fuel with some fuel additives and the influence of the emissions on the air quality there is a need for emission abatement.

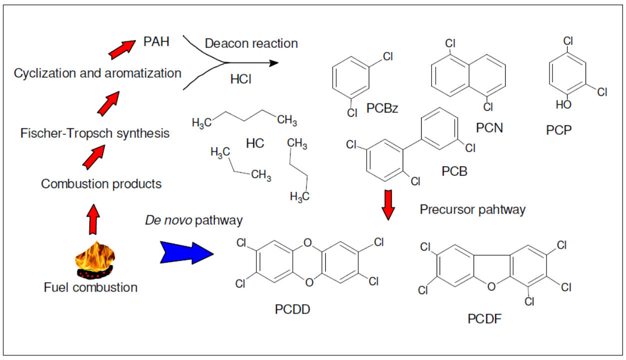

Figure 3. Pathways of dioxin formation

Improvements in combustion process aim to decrease the formation and emissions of chlorinated xenobiotics. However, conventional combustion units with high emission factors are still widely used over the world. Secondary reduction techniques such as filters, electrostatic precipitators, scrubbers and catalytic converters are used to reduce the emissions because of their affinity to particulate matter [20]. However, because of the high price and technical issues, most of these techniques are not suitable for residential combustion units; therefore, the problem is serious on residential areas where coal is basic energy source.

Liquid fuels

Despite the benefits of using fuel additives in reducing emissions and enhancing other fuel characteristics, certain additives have been known to or could pose serious health and environmental problems. While reducing particulate emissions to the environment is an obvious advantage, it is important to understand that the addition of additives to fuel does not alter the intrinsic toxicity of particles emitted in the exhaust. It is already clear that the use of lead as a fuel additive caused serious health and environmental problems, which has caused its ban in most parts of the world. Recent technologies to replace lead based additives includes nanotechnology, which is currently being used to produce $nCeO_2$. Although leaded gasoline is being banned in most parts of the world, it is still being used in many developing countries and also in aviation fuels. Lead, a ubiquitous and versatile metal, has been used since prehistoric times. It has become widely distributed and mobilized in the environment, and human exposure to and uptake of this non-essential element have consequently increased. At high levels of human exposure there is damage to almost all organs and organ systems, most importantly the central nervous system, kidneys and blood, culminating in death at excessive levels [18]. Following the advent of motor vehicles, there was a substantial increase in environmental lead contamination because of the use of lead in petrol. This resulted in an increase in community exposure to lead in many countries. Where lead derived from petrol comprises the major part of atmospheric lead, it is a significant contributor to the body lead burden and is the most widely distributed source of the metal in the environment. It is thus desirable to phase out the use of lead additives in fuels as quickly as possible on the global scale. Atmospheric lead that is deposited in soil and dust may be ingested by children and may substantially raise their blood lead levels. For the population at large, which is not occupationally exposed, food and water are important sources of baseline exposure to lead, in addition to atmospheric lead that is inhaled. According to a report by Seema [21], more than 15 million children in developing countries are suffering permanent neurological damage due to lead poisoning. Lead can be ingested from various sources, including lead paints and house dust contaminated by lead paint, as well as soil, drinking water, and food. The concentration of lead, total amount of lead consumed, and duration of lead exposure influence the severity of health effects. In the soil, lead has no physiological function in organisms.

The environmental fate of $nCeO_2$ is dependent on its applications and uses. As reported by Jessica and Yuji [22], the use of $nCeO_2$ in diesel fuel coupled with a particulate filter drastically decreases PM emissions in automobile exhaust up to 90% by weight. However, the authors also stated that some of the cerium escaping through the exhaust could accumulate in soils. Exhaust PM exists as very fine particles, leading to extremely diffuse pollution through air exposure and contamination of soil and water bodies. Cerium could also be introduced to soils via recycling of sewage sludge, and the aquatic environment via wastewater discharge from ceramic manufacturing plants. The contamination of soil with $nCeO_2$ could lead to the concentration of the nanoparticle in the food chain.

Conclusions

Effective additives may not necessarily reduce the quantity of ash deposited, but may alter its physical characteristics making it more friable and easily removed by black carbon blowers. The operation of heating systems, the boilers, ovens, furnaces and other in which fuel, such a wood, coke, oil, coal or gas is burned, gradually become fouled with solid deposits of black carbon or creosote and scale. The scale usually consists of mineral deposits, such as ashes of the fuel, while the deposit is primarily a sophisticated mixture of large amounts of organic compounds, amorphous carbon and smaller amounts of persistent organic compounds (POP). The organic matter of black carbon is usually good combustible, and particularly in chimneys, presents a definite fire hazard, when once ignited, it burns with great intensity, and such a fire, particularly in case of a defective chimney or line, may lead to accidental fire of the building. Introduction of even a small amount of chemical active compounds in fuel combustion performance of fuel can be changed and this is the intervention of fuel chemical catalytic combustion technology. Private industry has developed several fuel additives designed to decrease the amount of PM in cars exhaust and increase fuel efficiency. To assess the impact of cerium oxide nanoparticle on human health and the environment, especially near roadways where people may face higher levels of exposure on nanoparticles and estimate effect of motor vehicle exhaust and air quality and how they behave near roadways. The modern additives should suppress black carbon emission and improve combustion efficiency. In principle, it is possible by means of facilitation of nucleation of black carbon production that leads to increasing of its dispersion. The rate of black carbon burning in a flame front is proportional to the specific surface of black carbon particles; therefore, increasing of its dispersion leads to increasing of the degree of its burning. The catalysis of fuel burning gives another possibility to the growth of the degree of black carbon burning.

In Africa, where lead additives are still heavily used and where sugarcane production is high, ethanol can be a cheap source fuel of bioadditive. Instead of adding TEL, one way to increase the octane value of a gasoline is to change its composition so that it is less likely to generate the free radicals that lead to knock. Molecules with stronger bonds are less likely to break down; and strongly bonded hydrocarbons include low bond order hydrocarbons such as ethanol.

Oxygenated hydrocarbons should be added instead of metals to liquid fuels to make them burn more cleanly, thereby reducing toxic exhaust pollution, particularly carbon monoxide. Oxygenates are favoured not only for their vehicle emission benefits but also their blending properties in motor gasoline (e.g., octane). Oxygenated fuels are known to reduce PM emissions for motor vehicles and have been evaluated as potential sources of renewable fuel additive. Among the alternative fuels, biodiesel and ethanol are the most widely studied biofuels for diesel engines and have received considerable attention in recent years. Ethanol have been used in several countries for several years as it is produced from renewable sources and produces cleaner emission. Many literatures have showed that NO_x emission decreases with the increase in content of ethanol. He et al. evaluated the emission characteristics of five fuel blends with ethanol on a diesel engine [23]. The blends significantly reduce smoke, NO_x and CO_2 emissions. This indicate the potential of diesel reformation using ethanol for clean combustion. The addition of ethanol to diesel fuel changes the physicochemical properties of the blends that resulted in the decrease in density, cetane number, kinematic viscosity, high heat value and aromatics fractions. However, it is important to note that the impacts of ethanol on emissions would vary with engine operating conditions, ethanol content, additives and ignition improver.

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