

Review

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An Overview of Solid Fuel Processing, Kinetics, and the Advances in Minimizing Carbon Monoxide Emissions

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Review

An overview of solid fuel processing, kinetics, and the advances in minimizing Carbon monoxide emissions

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Abstract: There is a growing notion that biomass are the best resource to replace the declining fossil fuels, yet both share the long lived human threat: toxic combustion emissions. Among the toxic combustion products is carbon monoxide (CO) that not only causes acute but also chronic ailments. This brief review discusses the solid fuel processing technologies from combustion, thermochemical and biochemical processing to kinetics and thermodynamics including the mechanism for release of CO. It further expounds on the burden that CO has caused England and Wales in the last 25 years. The main gist are the systems that have been developed to minimize human exposure to CO including cooking, heating, catalytic and detection systems. Finally, alternative technologies are discussed that work by changing the chemical nature of solid fuels as a way to minimize CO emissions.

Keywords: pyrolysis; combustion emissions; Improved cookstoves; advanced boilers; CO detectors; catalyst impregnation

Highlights

- Solid fuel processing methods are discussed including extensive analysis of kinetics and thermodynamics
- Emission of pollutants from solid fuels is examined
- Mechanism for release of carbon monoxide (CO) is outlined
- Detailed analysis of systems used for minimizing human exposure to CO
- Recommendations for better methods used to process solid fuels especially those used for cooking are discussed

1. Introduction

It is of great importance that the emission of toxic gases from the combustion of solid fuels is minimised while diversifying energy sources to supplement the declining fossil fuels. Wood and coal are regarded as the world's most used solid fuels in homes (Ots et al., 2018), (Char and Graphite, 2017). It is estimated that 90% of solid fuel (coal, crop residue, and woody materials) consumption is by developing countries. This population constitutes more than three billion people (Shen et al., 2014). Biomass is the best foreseeable solid fuel to replace coal due to its ease of processing, friendliness to the environment and abundance.

Based on biological variety, biomass may be classified as wood like barks, conifers, stems, angiosperm, chips, sawdust, softwood, pellets (Sikarwar et al., 2017). These contain very low levels of contaminants (heavy metals and sulfur) and are easily processed by conventional methods like co-firing, combustion, cogeneration, gasification, and torrefaction. Biomass may also be herbaceous and agricultural to include straws like wheat, corn, rice, barley; flowers and grasses including

switchgrass, bamboo and cane and other residues like pulp, husks, grains, bagasse, and shells. Marine life like micro-organisms and plants like algae, water hyacinth, and weed also have a high potential for bioenergy. Industrial and contaminated waste like refuse-derives fuels, sewage sludge, municipal solid waste, plywood, paper-pulp sludge may also be used as bioenergy sources.

In the UK, the main biomass materials include palm, soya and sunflower oils, sugarcane, and palm or coconut husks (Anderson and Fergusson, 2006). Other solid fuels may be formed from food waste which comprises approximately 10 million tonnes. Most of this is discarded annually in the UK of which only 18% was recycled in 2016 (Nyombi et al., 2019). Charcoal derived from pyrolysis of carbonaceous materials is widely used as a fuel for home, industrial and recreational energy generation purposes. Coal is one of the oldest carbon rich solid fuel. There are four types of coal including peat, lignite, bituminous, and anthracite, with anthracite being most desirable due to its high heat content (Lisandy et al., 2017).

High volatile matter content and moisture render biomass a low energy intensive fuel. Processing such solid fuels is necessary to enhance energy output, reduce toxic and ozone depleting emissions and to increase the share of certain products in the outputs (i.e. gasification for increased gaseous products and bio-oil, torrefaction and low temperature pyrolysis for generating high quality solid products (Wang and Howard, 2018), etc.). During the processing of solid fuels, they undergo certain reaction mechanisms depending on the material properties and the controls used in the process. The data produced from controlled processing are used to determine the kinetic parameters for the processing or usage of solid fuels. The energy variations of enthalpies of activation between the reagent and the activated complex are usually in accordance with activation energies. Such properties are used to assess a given solid fuels for bioenergy production (M. S. Ahmad et al., 2017), (Lee et al., 2017), (Muktham et al., 2016).

During processing or usage of solid fuels, the main evolved gases are usually CO , CO_2 , H_2O , CH_4 , and C_2H_4 (Malika et al., 2016), (Özsin and Pütün, 2017). The production of CO , H_2 and CO_2 is enhanced under CO_2 (Borrego et al., 2009), over N_2 , Ar, or air atmospheres. Pyrolysis enhances the carbon mass fraction while H_2 , O_2 , N_2 and S mass fractions are reduced (Sadaka et al., 2015).

The smoke from burning solid fuels especially coal contains fine particles, CO , benzene, polycyclic aromatic hydrocarbons (PAHs) among other pollutants which are associated with reduced intrauterine growth (Adrian, 2011). Carbon monoxide has always posed serious dangers to human life. The lungs of mummified bodies from the Paleolithic era are frequently black (Borsos et al., 2003), a sign of heavy smoke which is indicative of poor combustion and thus CO production. In 1850's Claude Benard showed that CO blocked respiration in erythrocytes (Russell and Jeraci, 1984). Even in ruminants, CO is reported to decrease the digestion of hemicellulose and cellulose by 40 and 27% respectively (Russell and Jeraci, 1984).

In the effort to reduce toxic and greenhouse gas emissions, the EU through its European decarbonisation strategy 2050, has set targets including the 22% of the energy share should come from renewables by 2030 and 42% by 2050 up from 9% in 2010 (Capros et al., 2014). Furthermore, there are available technologies for minimizing CO including sensors (Nandy et al., 2018), improved cookstoves (Lucarelli et al., 2018), better heating systems (Ozil et al., 2009), catalytic systems (Carltonbird et al., 2018), (Hong and Sun, 2016) among others.

The objectives of this work therefore are to give an overview of the major solid fuel processing technologies, kinetics and the major emissions with focus on carbon monoxide (CO) (section 3). Furthermore, technologies for minimizing CO exposure to humans have been listed and explained in detail (section 4). Finally, impregnation of chemical substances into solid fuels (section 5) is discussed as a way to influence certain reactions to produce desired products.

2. Methodology

This work is neither a comprehensive nor a systematic review. It is an attempt to give a general overview of solid fuel processing, toxic combustion emissions, and the major systems used to minimize human exposure to toxic pollutants under a three-pillar approach:

- a. To succinctly delineate the solid fuel processing technologies including combustion, thermochemical and biochemical processing; give an account on the solid fuel combustion kinetics; and elucidate on the burden associated with combustion emissions with focus on carbon monoxide,
- b. To list the major systems that have been used to minimize human exposure to carbon monoxide emissions from solid fuel combustion
- c. To briefly justify the chemical changes imparted by use of chemical impregnation methods on solid fuel combustion

The methodical approach to this review was three-fold:

- i. The solid fuel processing technologies, combustion kinetics and the challenge brought by CO poisoning was delineated. The authors carefully studied the relevant literature in relation to solid fuel combustion (i.e. smoldering, flaming), thermochemical conversion (torrefaction, flash carbonization, pyrolysis, gasification, hydrothermal, and liquefaction), and biochemical conversion technologies. They further collected literature on solid fuel combustion kinetics including the mechanisms for release of emissions, and kinetic parameters. Finally, the authors enumerated the challenges brought by CO poisoning including statistics on death by year in England and Wales.
- ii. The major systems used to minimize human exposure to CO are listed and briefly explained. These included, improved cookstoves, heating systems, catalytic oxidation systems and CO detection systems
- iii. The authors also briefly highlight the importance of using chemical catalysts as additives to solid fuels to be used for various applications.

Collecting the data involved examining scientific literature (Scopus, Google scholar, Science Direct) and “other” literature including industrial work, and commercial literature as empirical articles or theoretical. The key words used either individually or in combinations included; solid fuel; combustion, thermochemical conversion; biochemical conversion; combustion kinetics; combustion emissions; cookstoves, heating systems; CO oxidation; CO sensors; catalyst impregnation.

3. Solid fuel processing, kinetics, and CO emissions

3.1. Solid fuel processing technologies

3.1.1. Combustion

Combustion may be smoldering, flaming or a combination of both. The occurrence of any type depends on the prevailing conditions, usually temperature, airflow, and material properties.

Smoldering

Smoldering is a low-temperature, slow, flameless combustion maintained by heat from the reaction of oxygen with fuel components (Rein, 2016). It involves both pyrolysis of the original fuel and oxidation of the resulting char. Since smoldering usually occurs in limited oxygen supply, the rate of reaction is directly proportional to the oxygen mass flux. At above 35% oxygen supply, smoldering may transition to flaming (Hadden et al., 2013). This transition may also occur at temperatures just above 450 °C in sawdust materials (Nyombi et al., 2019). For sawdust and charcoal, smoldering is affected by temperature, ash content, volatile matter content, particle size oxygen supply, and heat loss. With sawdust, temperature increases while during charcoal smoldering, temperature decreases. Low ash content allows oxygen diffusion leading to increased smoldering front propagation. High volatile content fuels produce much smoke during smoldering compared to low volatile content fuels (He et al., 2014).

If smoldering starts from the top, the fire will usually spread sideways and downwards Figure 1. This creates a void in the form of a pan semi-filled with ash. The downward spread is facilitated by forward smoldering. The sideways/lateral fire spread is enhanced by oxygen availability and is usually faster than downward smoldering due to ash and char layers that prevent quick penetration of oxygen during the downward flow. Under the natural conditions, the flow induced by the plume ensures sufficient oxygen supply to sustain the horizontal spread of fire to the top-most layer. Oxygen then penetrates by diffusion (Rein, 2016).

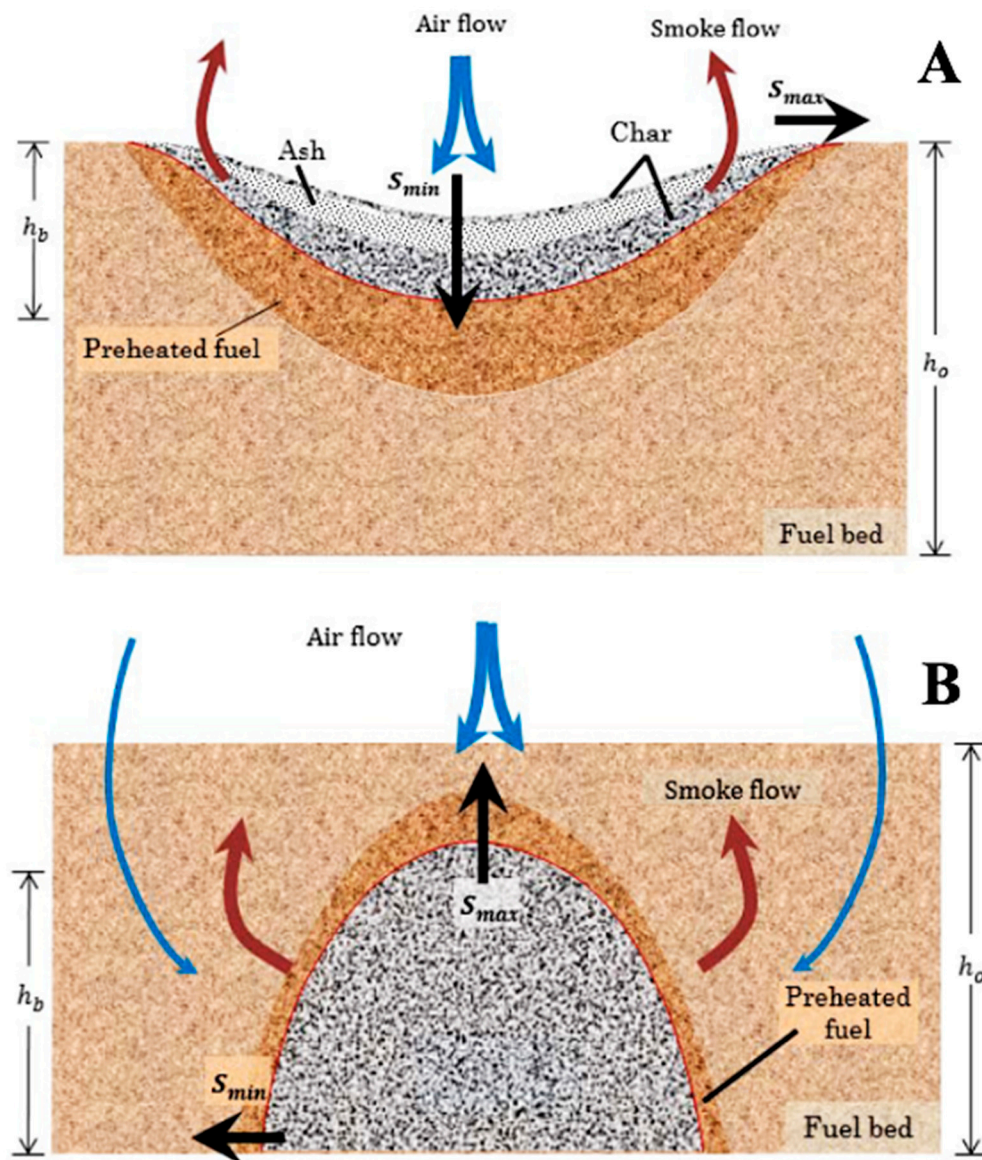


Figure 1. Downward (A) and upward (B) smoldering propagation in a porous solid fuel. Adapted and modified from (Rein, 2016).

Flaming

Flaming combustion is the process occurring when visible flames and plume of a fire are visible. A flame is the visible, luminous body where the oxidation reaction is occurring. In flaming combustion, the fuel is in the gas phase. The reactions and heat release occur in the gas adjacent to the liquid or solid surface. Flaming combustion is controlled by external and internal factors (Moghtaderi and Fletcher, 1998). External factors include the temperature and oxygen

availability, as well as the radiant heat flux. The internal factors include the moisture content and the geometrical properties.

3.1.2. Thermochemical processing

Torrefaction

Torrefaction (Chen et al., 2015) is a low temperature (200–300 °C) process in an inert environment in which moisture, carbon oxides (CO & CO₂), and oxygen are removed from biomass materials including depolymerisation of long-chain polysaccharide forming a solid product with low H/C and O/C ratios. Several studies (Sun et al., 2016) (Weber and Quicker, 2017), (Varjani et al., 2019) have shown that the properties of the final product (weight loss, thermophysical and chemical) are influenced mainly by torrefaction temperature. High temperatures favour biochar while bio-oil decreases. The non-condensable gases remain almost constant (Chen et al., 2015). Torrefaction is aimed at improving the thermochemical properties of final biomass product for application in co-firing with coal, gasification, and combustion.

Flash carbonization

This is a high-pressure process (1-2 Mpa) in which a flash fire ignited on a biomass packed bed results in a solid and gas-phase as the main products. Flash carbonisation is usually operated at mid-level temperatures (300-600 °C) and short residence time in the range of 30-40 minutes. About 40% yield is a solid product. This process is not very much embraced compared to other methods for making biochar (Chen et al., 2015).

Pyrolysis

Pyrolysis is a slow breakdown of organic matter contained in solid fuel at temperatures 300-900 °C under limited oxygen conditions. In biomass solid fuels, the main constituents (cellulose, hemicellulose, and lignin) each undergoes separate reaction routes including depolymerisation, cross-linking and fragmentation at appropriate temperatures. The main products of pyrolysis are solid (char), liquid (bio-oil) and gaseous (syngas - CO, CO₂, H₂, and C₁–C₂ hydrocarbons). The quantity and quality of each product depend on the pyrolysis conditions (heating rate, final holding temperature and residence time) and the properties of the raw materials used (Sun et al., 2016), (Weber and Quicker, 2017), (Brennan and Owende, 2010). Generally, syngas increases while biochar decreases with increasing pyrolysis temperature (Sun et al., 2016).

Gasification

Gasification is a high temperature (800-1000 °C) process in which biomass is partially oxidised using air, steam, oxygen, CO₂ or gas mixture, forming gaseous products mainly CO, H₂, CO₂, N, and CH₄. Other products include biochar which is usually 5-10% of the starting raw material and liquid products (tar & oils). Gasification can produce syngas from a wide variety of raw materials (biomass, coals, and plastics), unlike other similar processes. Much as syngas has a low calorific value (4–6 MJ m³), it can burn directly or be used as a fuel for gas turbines and engines (Sun et al., 2016), (Weber and Quicker, 2017), (Varjani et al., 2019), (Brennan and Owende, 2010). The gasifiers used may be fixed bed reactors; moving bed reactors; fluidized bed reactors; or entrained bed reactors depending on the interaction between the biomass and the gasification agent. Gasification reactions are affected by the gasification agent type, reaction temperature, pressure, and gasification agent–biomass ratio. The reaction temperature is usually the main parameter influencing gasification reactions (Sun et al., 2016).

Hydrothermal

Unlike dry thermal processes (pyrolysis and gasification), hydrothermal processes involve mixing biomass with water and the product is left to stabilise in a reactor. After a certain period, the

temperature of the reactor is raised. To keep the water in the liquid phase, high pressures are used. This process may be called hydrothermal carbonisation if the main intended product is hydrochar; hydrothermal liquefaction for bio-oil; and hydrothermal gasification for syngas. Hydrochar usually has a high carbon content compared to dry processes (pyrolysis and gasification). In addition to heating rate, final holding temperature and residence time used in dry processes, the water-biomass ratio also plays a vital role to the final product yield (Varjani et al., 2019).

Liquefaction

Liquefaction is a high pressure (5-20 MPa), low temperature (300-350 °C) process used to convert high moisture content materials like algal biomass into liquid fuels (bio-oil). The process is aided by catalysts in the presence of hydrogen. Compared to other thermal processes, liquefaction is quite expensive and involves complex reaction systems (Brennan and Owende, 2010).

3.1.3. Biochemical processing

The biological process for energy conversion of biomass into other fuels includes anaerobic digestion, fermentation and photobiological hydrogen production (Brennan and Owende, 2010). Anaerobic digestion is mainly for the production of biogas (composed of CH_4 as the main product, CO_2 and nitrogen); fermentation is for production of bioethanol while photobiological hydrogen production, like the name suggests, is for H_2 production (Anderson and Fergusson, 2006), (Weber and Quicker, 2017).

Figure 2, summaries the different types of biomass processing technologies and the main products.

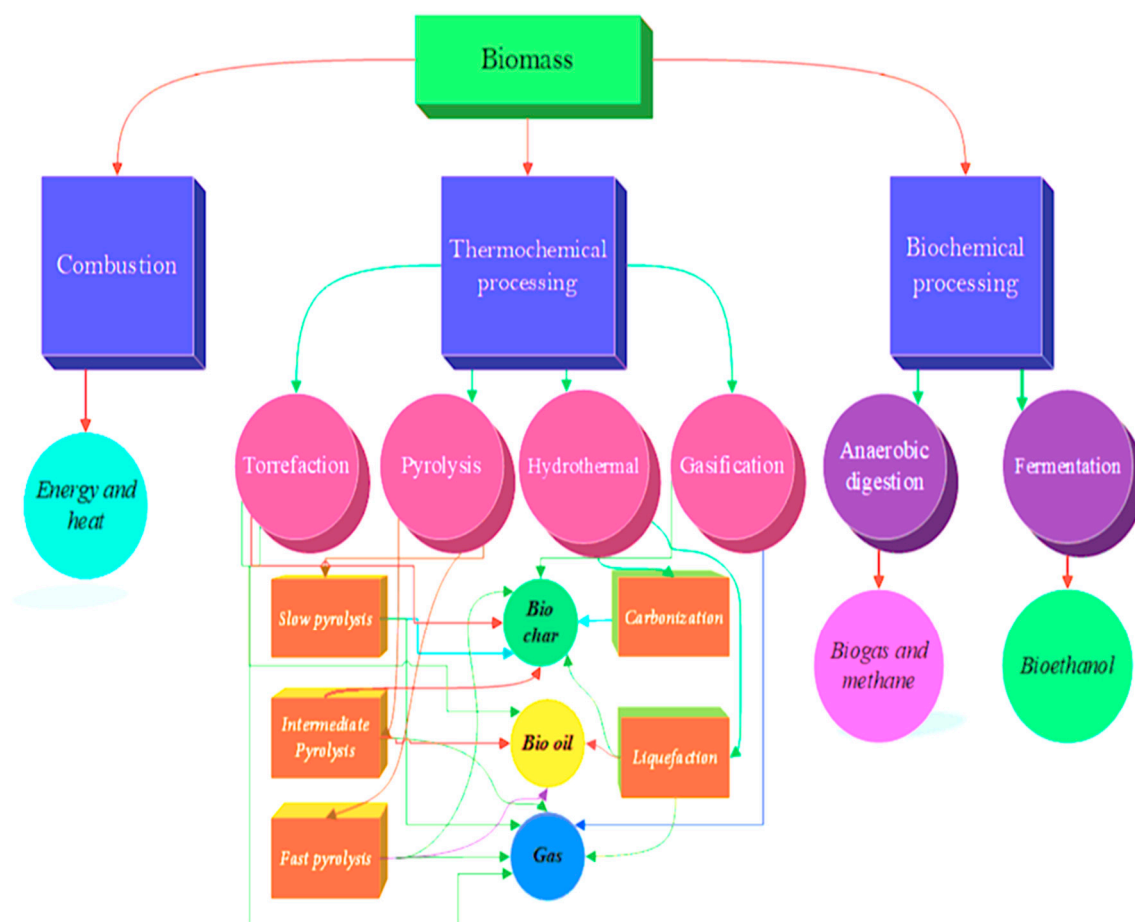
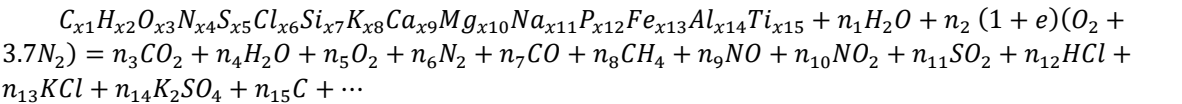


Figure 2. Biomass processing technologies and respective products obtained through each of the technologies (Akhtar et al., 2018).

3.2. Solid fuel combustion kinetics

3.2.1. General mechanism for release of emissions

Solid fuel combustion is a complex process involving simultaneous mass and heat transfer with chemical reactions. The prediction of solid fuel combustion for design, process analysis, and control demands an in-depth understanding of the fuel properties and how these will influence the final desired product (Jenkins et al., 1998). A general global reaction of solid fuel in the air might follow the reaction:



The coefficients x_1 - x_{15} in hybrid poplar and rice straw may take the following values shown in Table 1.

Table 1. Proposed elemental coefficients of a typical biomass solid fuel (Jenkins et al., 1998).

Element	Coefficient	Hybrid poplar	Rice straw	Rice/Poplar
C	x_1	4.1916	3.2072	0.77
H	x_2	6.0322	5.1973	0.86
O	x_3	2.5828	2.8148	1.09
N	x_4	0.0430	0.0625	1.45
S	x_5	0.0006	0.0057	9.50
Cl	x_6	0.0003	0.0165	55.00
Si	x_7	0.0057	0.5000	87.72
K	x_8	0.0067	0.0592	8.84
Ca	x_9	0.0337	0.0141	0.42
Mg	x_{10}	0.0205	0.0135	0.66
Na	x_{11}	0.0002	0.0079	39.50
P	x_{12}	0.0012	0.0086	7.17
Fe	x_{13}	0.0007	0.0029	4.14
Al	x_{14}	0.0008	0.0073	9.13
Ti	x_{15}	0.0002	0.0004	2.00

The 15 elements included in the empirical formula form just part of the complete biomass fuel composition. The metallic constituents are responsible for the ash content of a given solid fuel. The moisture term is responsible for the spontaneity of reactions during combustion. The simplified air term (21% oxygen and 79% nitrogen) has a significant meaning in that the presence of certain gases in the fuel may contribute heavily to the reaction progress in solid fuel combustion. The products are much more complex than what is shown in the equation above and the detailed chemistry varies considerably depending on the system (Jenkins et al., 1998).

3.2.2. Mechanisms for release of CO

The structures of functional groups that are present on the char or found on partially oxidized char surfaces are presented in Figure 3. These structures are responsible for the formation of CO in addition to the free-edge site reactions.

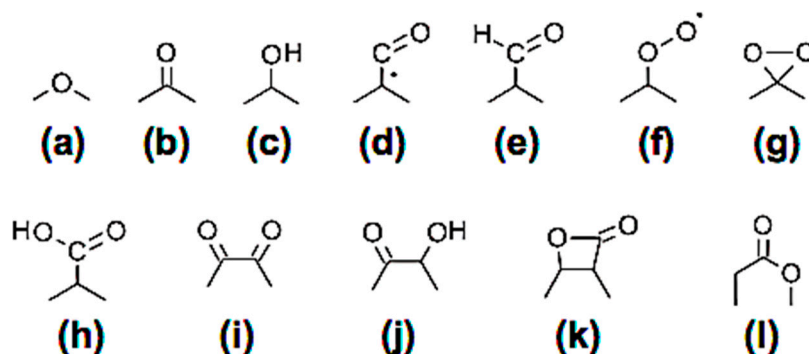


Figure 3. Several oxygenated functional groups present on partially oxidized PAHs or soot. (a) Pyrone (ether). (b) Ketone. (c) Hydroxyl. (d) Dangling CO (carbonyl). (e) Dangling HCO. (f) Peroxide. (g) Dioxyranyl. (h) Carboxylic. (i) Quinone. (j) Phenolic. (k) Lactone. (l) Oxypinyloxy (Raj et al., 2012), (A. Nyombi, M. Williams, 2019).

After the surface oxygenated functional groups are all oxidized, further reactions follow the “free edge site” reactions leading to formation of CO, a reaction similar to the breakdown of soot reported in the literature (Raj et al., 2012). This follows a simplified mechanism, though numerous reactive sites can be present. As seen in Figure 4, free-edge sites and zig-zag sites on a polycyclic aromatic hydrocarbon (PAH) molecule react with oxygen to release CO and the formation of new edge-sites. This process is repeated until all the edge-sites are completed, after which, complete oxidation of the six-membered rings takes place until the entire structure is oxidised (Raj et al., 2012).

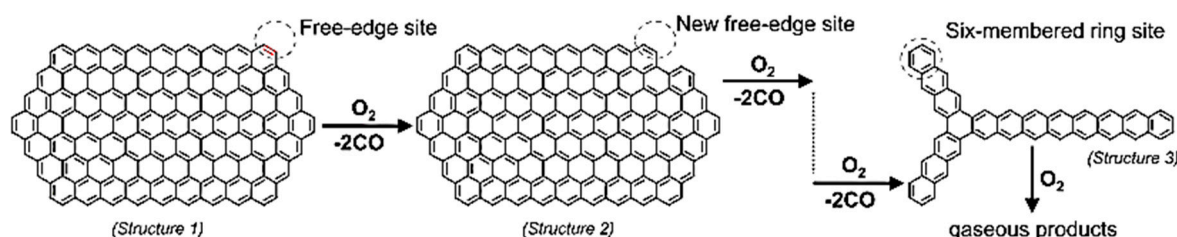


Figure 4. Free-edge oxidation of an example polycyclic aromatic hydrocarbon molecule comprising a soot particle. The oxidation of a free-edge on structure 1 leads to the generation of a new free-edge in this example on structure 2, thus causing chain oxidation through the reaction (Raj et al., 2012).

Further detailed complex reactions involve the breakdown of planar and curved PAHs which involves the formation of oxygenated free radicals that later desorb as CO or CO₂. Other carbon-centered free radicals react with oxygen forming CO as the main product (Raj et al., 2013), (Parker et al., 2015).

3.2.3. Kinetic parameters for pyrolysis or oxidation of solid fuels

The main methods that have been used over the years for the determination of activation energies during pyrolysis or oxidation reactions of solid fuels are here summarized in Table 2. The description and modelling of the reactions/mechanisms involved in solid fuel processing have been a serious challenge. This has been mainly due to multi-step and parallel reactions and the heterogeneous nature of solid fuels during decomposition. Hence researchers have developed model-fitting and model free iso-conversional as well as model methods to solve the challenge. These have been applied to TGA/DSC data and used to predict as accurate as possible the kinetic parameters during solid fuel decomposition. These can sometimes get complex for solid fuel wastes especially Municipal Solid Wastes and Refuse Derived Fuels.

Table 2. Methods for determination of kinetic parameters.

Method	Equation	Reference
Friedman	$\ln \left[\beta \left(\frac{d\alpha}{dt} \right) \right] = - \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right) + \ln A f(\alpha)$	(Starink, 2003), (Fedunik-hofman et al., 2019)
Gupita	$\ln \left[\beta \left(\frac{d\alpha}{dt} \right) \right] = - \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right) + \ln A f(\alpha)$	(Gupta et al., 1988)
Freeman & Car-roll	$Y = - \frac{E}{R} X + n$ where, $Y = \frac{\Delta \ln \left(\frac{d\alpha}{dt} \right)}{\Delta \ln (1-\alpha)}$ and $X = \frac{\Delta \left(\frac{1}{T} \right)}{\Delta \ln (1-\alpha)}$	(Jerez, 1983)
Kissinger-Akahira-Sanose (KAS)	$\ln \left(\frac{\beta}{T^2} \right) = - \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right) + \ln \left(\frac{A_\alpha R}{g_{(\alpha)} E_\alpha} \right)$	(Danvirutai and Noisong, 2015)
Flyn-Wall-Onzawa (FWO)	$\ln \beta = -1.052 \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right) + \ln \left(\frac{A E_a}{R g_{(\alpha)}} \right) - 5.331$	(Ozawa, 1965), (Joseph and Leo, 1966)
Starink	$\ln \left(\frac{\beta}{T^{1.95}} \right) = - \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right) + \ln \left(\frac{A_\alpha R}{g_{(\alpha)} E_\alpha} \right)$ $\ln \left(\frac{\beta}{T^{1.92}} \right) = -1.008 \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right) + \ln \left(\frac{A_\alpha R}{g_{(\alpha)} E_\alpha} \right)$	(Starink, 2003)
Boswel	$\ln \left(\frac{\beta}{T_{max}} \right) = - \left(\frac{E_a}{R} \right) \left(\frac{1}{T_{max}} \right) + constant$	(Starink, 2003), (Boswell, 1980)
Coats and Redfern	$\ln \left(\frac{g_{(\alpha)}}{T^2} \right) = - \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right) + \ln \left(\frac{A R}{\beta E} \right)$	(Sajjad et al., 2017)
ASTM-E698	$\ln \left[\beta \left(\frac{d\alpha}{dt} \right) \right] = - \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right) + \ln [k_o (1 - \alpha)]$	(Osman et al., 2017)
Karaosmanoglu & Cif	$\ln \left[\frac{-1}{\alpha_o - \alpha_f} \frac{d\alpha}{dt} \right] = \ln(A) - \frac{E_a}{RT} + n \ln \left(\frac{\alpha - \alpha_f}{\alpha_o - \alpha_f} \right)$	(Fernandez et al., 2017)
Isothermal method	$- \ln(t) = - \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right) + \ln \left[\frac{A}{g'(\alpha)} \right]$ $\ln \left(\frac{\beta}{H_{(x)}} \right) = \left(\frac{0.0048 A_\alpha E_\alpha}{g_{(\alpha)} R} \right) - \left(\frac{1.0516 E_\alpha}{R} \right) \left(\frac{1}{T} \right)$ $\ln \left(\frac{\beta}{h_{(x)} T^2} \right) = \ln \left(\frac{A_\alpha R}{g_{(\alpha)} E_\alpha} \right) - \left(\frac{E_a}{R} \right) \left(\frac{1}{T} \right)$	(Wang et al., 2014)
FWO and KAS Iterative methods	where $H_{(x)} = \frac{(e^{-x}) h_{(x)}/x^2}{0.0048 \exp(-1.0516x)}$ and $h_{(x)} = \frac{x^4 + 18x^3 + 86x^2 + 96x}{x^4 + 20x^3 + 120x^2 + 240x + 120}$ which is the 4 th degree Senum and Yang approximation that gives an accuracy better than 10 ⁻⁵ % for $x = E/RT \geq 20$.	(Senum and Yang, 1977), (Pérez-Maqueda and Criado, 2000)
Vyazovkin	$\Phi(E_\alpha) = \sum_{i=1}^n \sum_{j \neq i}^n \frac{J[E_\alpha, T_i(t_\alpha)]}{J[E_\alpha, T_j(t_\alpha)]}$ Where the time integral: $J[E_\alpha, T_i(t_\alpha)] = \int_{t_\alpha - \Delta\alpha}^{t_\alpha} \exp \left[\frac{-E_\alpha}{RT_i(t)} \right] dt$ where, T(t) is the actual sample temperature, J is the integral with respect	(Vyazovkin and Wight, A, 1998), (Vyazovkin, 2006)

	to $T(t)$ and $T_i(t)$ is the temperature programs	
Kissinger	$\ln\left(\frac{\beta}{T_{max}^2}\right) = \left(-\frac{E_a}{R}\right)\left(\frac{1}{T_{max}}\right) + \frac{\ln AR}{E_a}$	(Kissinger, 1956), (Blaine and Kissinger, 2012)

In the equations (in table 2 above), α - is the fractional conversion, β - is the heating rate, A - is the pre-exponential factor, R - is the universal gas constant, and E_a is the activation energy. The functions, $g(\alpha)$ - the integral form, and $f(\alpha)$ - the differential form, are used to determine the reaction mechanisms (Vyazovkin, 2006), (Vlaev et al., 2008) as shown in Table 3.

Table 3. Common mechanisms used during pyrolysis and degradation of solids.

No.	Symbol	Name of the Function	$g(\alpha)$	$f(\alpha)$	Rate-determining mechanism
1. Chemical process or mechanism non-invoking equations					
1	F1/3	One-third order	$1-(1-\alpha)^{2/3}$	$(3/2)(1-\alpha)^{1/3}$	Chemical reaction
2	F3/4	Three-quarters order	$1-(1-\alpha)^{1/4}$	$4(1-\alpha)^{3/4}$	Chemical reaction
3	F3/2	one and a half order	$[(1-\alpha)^{-1/2}-1]$	$2(1-\alpha)^{3/2}$	Chemical reaction
4	F2	Second order	$(1-\alpha)^{-1}-1$	$(1-\alpha)^2$	Chemical reaction
5	F3	Third order	$(1-\alpha)^{-2}-1$	$(1/2)(1-\alpha)^3$	Chemical reaction
2. Acceleratory rate equations					
6	P3/2	Mampel power law	$\alpha^{3/2}$	$(2/3)\alpha^{1/2}$	Nucleation
7	P1/2	Mampel power law	$\alpha^{1/2}$	$2\alpha^{1/2}$	Nucleation
8	P1/3	Mampel power law	$\alpha^{1/3}$	$3\alpha^{2/3}$	Nucleation
9	P1/4	Mampel power law	$\alpha^{1/4}$	$4\alpha^{3/4}$	Nucleation
10	E1	Exponential law	$\ln\alpha$	α	Nucleation
3. Sigmoidal rate equations or random nucleation and subsequent growth					
11	A1, F1	Avrami-Erofeev equation	$-\ln(1-\alpha)$	$(1-\alpha)$	Assumed random nucleation and its subsequent growth, $n=1$
12	A3/2	Avrami-Erofeev equation	$[-\ln(1-\alpha)]^{2/3}$	$(3/2)(1-\alpha)[- \ln(1-\alpha)]^{1/3}$	Assumed random nucleation and its subsequent growth, $n=1.5$
13	A2	Avrami-Erofeev equation	$[-\ln(1-\alpha)]^{1/2}$	$2(1-\alpha)[- \ln(1-\alpha)]^{1/2}$	Assumed random nucleation and its subsequent growth, $n=2$
14	A3	Avrami-Erofeev equation	$[-\ln(1-\alpha)]^{1/3}$	$3(1-\alpha)[- \ln(1-\alpha)]^{2/3}$	Assumed random nucleation and its subsequent growth, $n=3$
15	A4	Avrami-Erofeev equation	$[-\ln(1-\alpha)]^{1/4}$	$4(1-\alpha)[- \ln(1-\alpha)]^{3/4}$	Assumed random nucleation and its subsequent growth, $n=4$
16	Au	Prout-Tomkins equation	$\ln[\alpha/(1-\alpha)]$	$\alpha(1-\alpha)$	Branching nuclei
4. Deceleratory rate equations					
4.1 Phase boundary reactions					
17	R1, F0, P1	Power law	α	$(1-\alpha)^0$	Contracting disk
18	R2, F1/2	Power law	$1-(1-\alpha)^{1/2}$	$2(1-\alpha)^{1/2}$	Contracting cylinder (Cylindrical symmetry)
19	R3, F2/3	Power law	$1-(1-\alpha)^{1/3}$	$3(1-\alpha)^{2/3}$	Contracting sphere (spherical symmetry)

4.2 Based on the diffusion mechanism					
20	D1	Parabola low	α^2	$1/2\alpha$	One-dimensional diffusion
21	D2	Valensi equation	$\alpha+(1-\alpha)\ln(1-\alpha)$	$[-\ln(1-\alpha)]^{-1}$	Two-dimension diffusion
22	D3	Jander equation	$[1-(1-\alpha)^{1/3}]^2$	$(3/2)(1-\alpha)^{2/3}[1-(1-\alpha)^{1/3}]^{-1}$	Three-dimensional diffusion, Spherical symmetry
23	D4	Ginstling-Brounstein equation Zhuravlev,	$[1-2\alpha/3-(1-\alpha)^{2/3}]$	$(3/2)[(1-\alpha)^{-1/3}-1]^{-1}$	Three-dimensional diffusion, Cylindrical symmetry
44	D5	Lesokin, Tempelman equation	$[(1-\alpha)^{-1/3}-1]^2$	$(3/2)(1-\alpha)^{4/3}[(1-\alpha)^{-1/3}-1]^{-1}$	Three-dimensional diffusion
25	D6	Anti-Jander equation	$[(1+\alpha)^{1/3}-1]^2$	$(3/2)(1+\alpha)^{2/3}[(1+\alpha)^{1/3}-1]^{-1}$	Three-dimensional diffusion
26	D7	Anti-Ginstling-Brounstein equation Anti-Zhuravlev,	$1+2\alpha/3-(1+\alpha)^{2/3}$	$(3/2)[(1+\alpha)^{-1/3}-1]^{-1}$	Three-dimensional diffusion
27	D8	Lesokin, Tempelman equation	$[(1+\alpha)^{-1/3}-1]^2$	$(3/2)(1+\alpha)^{4/3}[(1+\alpha)^{-1/3}-1]^{-1}$	Three-dimensional diffusion
5. Another Kinetic equation with unjustified mechanism					
28	G1		$1-(1-\alpha)^2$	$1/2(1-\alpha)$	
29	G2		$1-(1-\alpha)^3$	$1/3(1-\alpha)^2$	
30	G3		$1-(1-\alpha)^4$	$1/4(1-\alpha)^3$	
31	G4		$[-\ln(1-\alpha)]^2$	$(1/2)(1-\alpha)[1\ln(1-\alpha)]^{-1}$	
32	G5		$[-\ln(1-\alpha)]^3$	$(1/3)(1-\alpha)[1\ln(1-\alpha)]^{-2}$	
33	G6		$[-\ln(1-\alpha)]^4$	$(1/4)(1-\alpha)[1\ln(1-\alpha)]^{-3}$	
34	G7		$[1-(1-\alpha)^{1/2}]^{1/2}$	$4\{(1-\alpha)[1-(1-\alpha)^{1/2}]\}^{1/2}$	
35	G8		$[1-(1-\alpha)^{1/3}]^{1/2}$	$6\{(1-\alpha)^{2/3}[1-(1-\alpha)^{1/3}]\}^{1/2}$	

The thermodynamic parameters like entropy ΔS , pre-exponential factor (A), Gibbs free energy ΔG , and enthalpy ΔH (Sajjad et al., 2017) are determined as follows:

$$A = \beta \cdot E_{\alpha} \exp\left(\frac{E_{\alpha}}{RT_m}\right) / R \cdot T_m^2$$

$$\Delta H = E_{\alpha} - RT$$

$$\Delta G = E_{\alpha} + R \cdot T_m \cdot \ln\left(\frac{K_b \cdot T_m}{h \cdot A}\right)$$

$$\Delta S = \frac{\Delta H - \Delta G}{T_m}$$

where K_b is the Stefan Boltzman constant ($1.38 \cdot 10^{-23}$ J/K), h is the Planks constant ($6.626 \cdot 10^{-34}$ Js), and T_m is the DTG peak temperature.

3.3. Carbon monoxide from solid fuels: a persistent challenge

Solid fuel emissions are dangerous to human health especially carbon monoxide (CO). It was observed that 5.2% of suicide deaths in England and Wales were due to CO poisoning in 2001-2011. These numbers declined to a total of 53 deaths by 2015 with England accounting for 91% of the total deaths (Office-for-National-Statistics-UK, 2016). Gas suicides cases reduced by 53% for the year between 2001 to 2011. However, there was a 17-fold increase fatal cases due to helium for the two-year period 2001-2002 to 2010-2011. Barbecue charcoal gas fatalities increased from one (1) to eleven (11) in the same period (Office-for-National-Statistics-UK, 2016), (Gunnell et al., 2015) Table 4.

Table 4. Number of deaths from accidental CO poisoning in England and Wales, deaths registered in 2011-2015^{1,2,3}. Adapted from the Office of National Statistics (UK) (Office-for-National-Statistics-UK, 2016).

ICD 10 Code	Underlying cause	England					Wales				
		2011	2012	2013	2014	2015	2011	2012	2013	2014	2015
V00-X59	All accidental carbon monoxide poisonings	75	58	57	52	48	4	7	3	3	5
X47	Accidental poisoning by other gases and vapours	33	23	22	25	24	1	2	2	1	1
X47.0	Occurrence at home	28	17	14	18	23	1	1	2	0	1
X47.1	Occurrence in residential institution	0	0	0	0	0	0	0	0	0	0
X47.2	Occurrence at school other institution/public admin area	0	0	0	0	0	0	0	0	0	0
X47.3	Occurrence at sports/athletics area	0	0	0	0	0	0	0	0	0	0
X47.4	Occurrence on street/highway	1	1	0	0	0	0	0	0	0	0
X47.5	Occurrence at trade/service area	1	0	0	1	0	0	0	0	0	0
X47.6	Occurrence at industrial/construction area	0	0	1	0	0	0	0	0	0	0
X47.7	Occurrence on farm	0	0	0	0	0	0	0	0	0	0
X47.8	Occurrence at other specified place	3	4	6	4	1	0	1	0	1	0
X47.9	Occurrence at unspecified place	0	1	1	2	0	0	0	0	0	0
V01-V99	Transport accident	0	0	0	0	1	0	0	0	0	0
X00-X09	Accidental exposure to smoke, fire and flames	42	35	34	27	23	3	5	1	2	4

¹Cause of death was defined using the International Classification of Diseases, Tenth Revision (ICD 10). Deaths were selected where the underlying cause of death was accidental (ICD 10 codes V01-X59), and where the secondary cause of death was the toxic effect of carbon monoxide (ICD 10 code T58). The original underlying cause of death has been used. ²Figures for England and Wales exclude deaths of non-residents based on boundaries as of November 2016. ³Figures are for deaths registered, rather than deaths occurring in each calendar. Due to the length of time it takes to complete a coroner’s inquest, it can take months or even years for a carbon monoxide poisoning death to be registered. More details can be found on the ONS website: www.ons.gov.uk/ons/guide-method/user-guidance/health-and-life-events/impact-of-registration-delays-on-mortality-statistics/index.html.

The Gas Safety Trust (GST) of the UK has also been compiling statistics on CO related death by fuel type (CO-Gas-Safety, 2015a) as well as other accidental death and injuries (Gas Safety Trust - UK, 2017), (CO-Gas-Safety, 2015b) especially from 1995 to date (Gas Safety Trust - UK, 2019), (Gas Safety Trust - UK, 2018).

4. Systems for minimising CO released from solid fuels

There are various approaches used to minimize the CO emissions from solid fuel combustion, each of which is designed depending on the specific application. The main approaches include cooking systems, heating systems, catalytic systems, and sensors/detectors Figure 5. Some applications employ a combination of two or more depending on the complexity and desired outcomes.

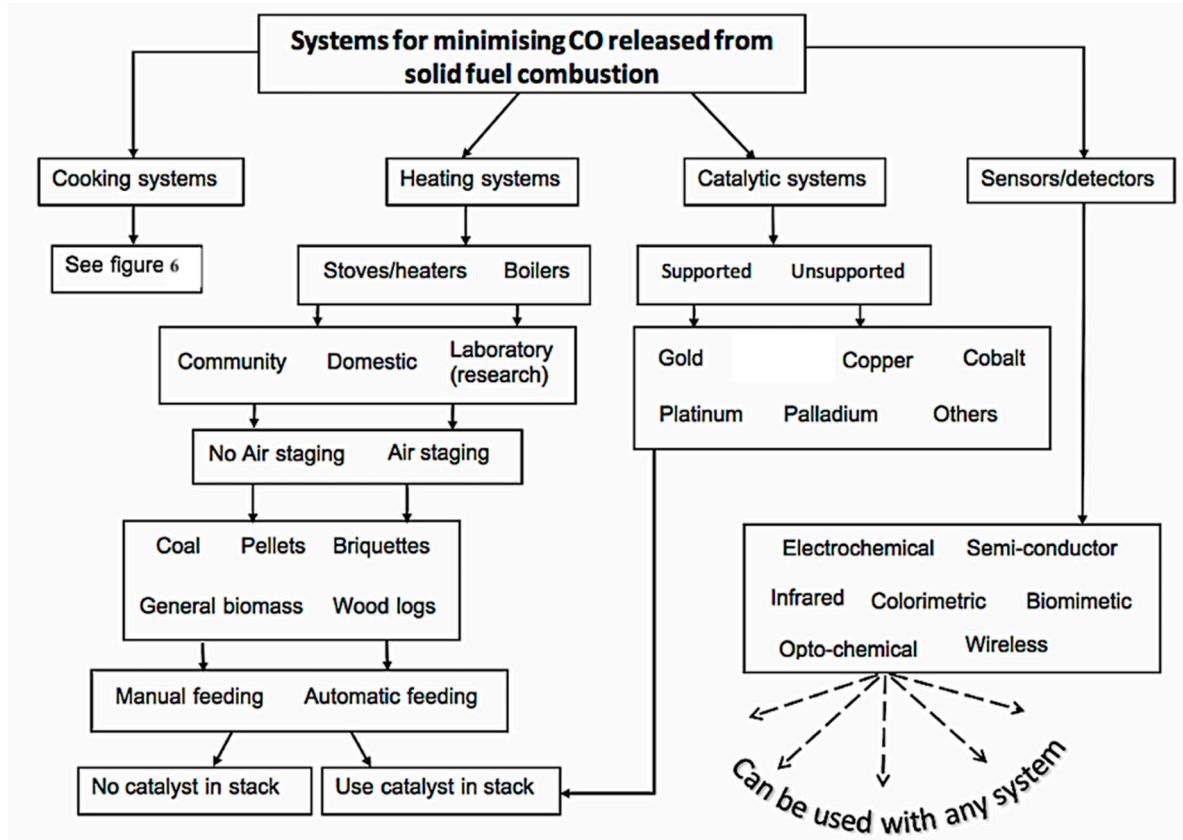


Figure 5. Systems for minimising CO from solid fuel combustion.

4.1. Improved cookstoves

Cooking on traditional cookstoves or three stone open fires has been reported as being inefficient in terms of energy as well as emission reduction of combustion pollutants. Technologically advanced/improved cookstoves (Jetter et al., 2012) (including Phillips cookstoves (Coffey et al., 2017), small scale gasifier cooking stove (MacCarty et al., 2010), (Njenga et al., 2016), (Njenga et al., 2017), stoves with electrical fans and chimneys (Still et al., 2015), rocket-type stoves, pot skirt stoves (MacCarty et al., 2010) among others) emitted less CO and other combustion pollutants and were more fuel-efficient than traditional stoves. However, the amounts of particulate matter are not much reduced (for Phillips cookstoves) (Coffey et al., 2017). Charcoal burning stoves (Still et al., 2015) were better than batch fed stove by up to 40% thermal efficiency. The daily carbon emissions from charcoal stoves in Kenya (Bailis et al., 2003) were lower than both traditional open fire and improved ceramic woodstoves. However, when each pollutant was weighted using a 20-yr global warming potential, charcoal stoves emitted larger amounts of greenhouse gases than a woodstove, open fire and ceramic woodstoves. Even the non-CO₂ emissions from charcoal stoves were higher for the 20-yr equivalent units compared to the three-stone fires and improved ceramic stoves. The combustion transition from wood to charcoal reduces PM emissions by 87% during the burning period and by 92% during smoldering when using ceramic wood-burning stoves compared to traditional cookstoves (Ezzati et al., 2000). Using accessories for pre-ignition may also save energy and minimise emissions. Using a lighting cone (Lask and Gadgil, 2017) as an accessory to shallow-bed charcoal stoves, ignition time, fuel (charcoal) consumption, and CO emissions were reduced by up to 50%, 40%, and 50% respectively. Improved cookstoves are durable and usually manufactured with locally available raw materials (Mehetre et al., 2017). The various classifications of traditional and improved cookstoves are shown in Figure 6.

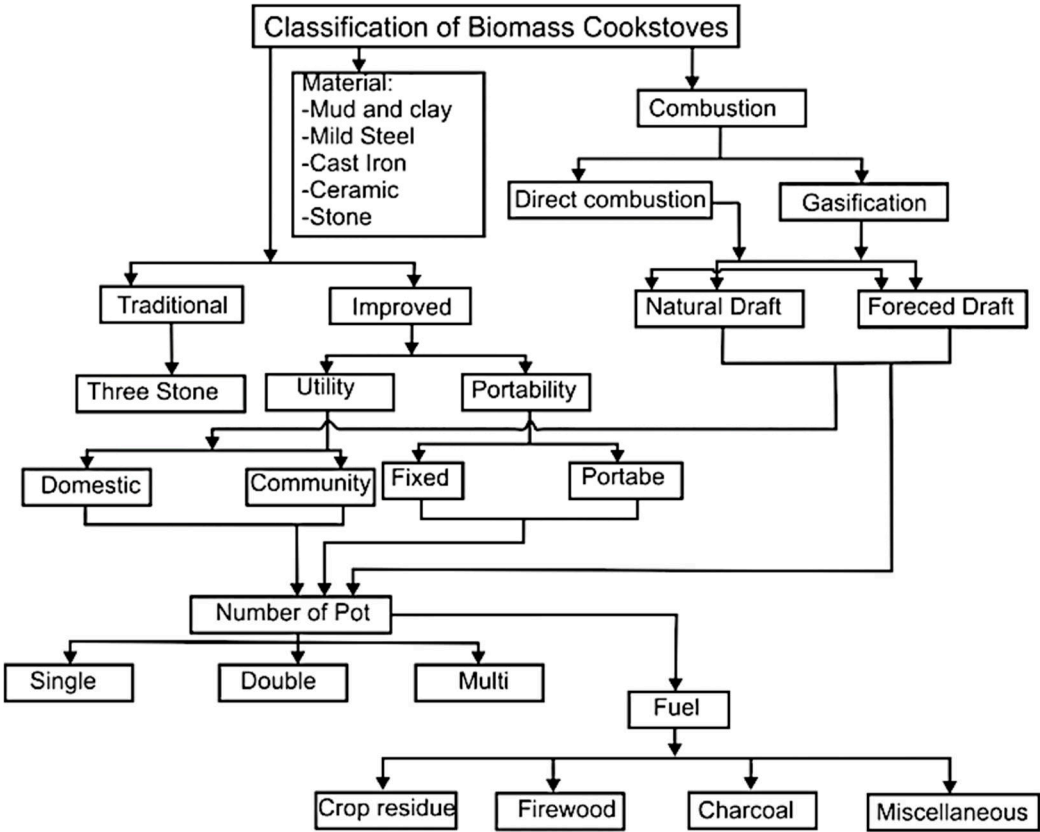


Figure 6. Classification of biomass cookstoves (Mehetre et al., 2017).

Some stoves are constructed with a layer of catalyst to enhance CO oxidation. However, the addition of $K_2Ti_2O_5$ to the monolith reduced CO emissions compared to the blank monolith but was unable to reduce CO emissions to the level of a stove without a catalyst *Figure 7*. The thermal efficiency of all the tested stove configurations were equally the same at about 25% for both low and high power operation (Paulsen et al., 2018). The different relevant research published on improved cookstoves is shown in Table 5.

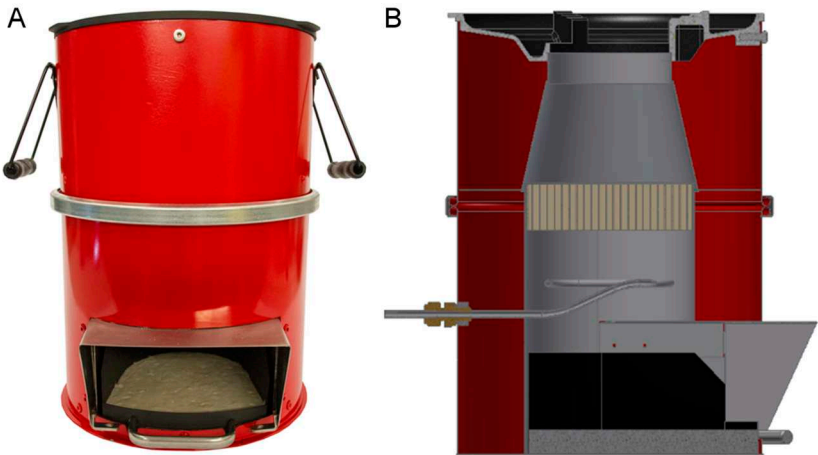


Figure 7. Rocket stove used for catalyst testing. (A) Picture of the rocket stove used during testing and (B) diagram of the rocket stove showing the catalytic monolith placement (Paulsen et al., 2018).

Table 5. Descriptions of the various improvements made on traditional cook stoves.

Description of traditional stoves	Description of improved stoves	CO from traditional stove	CO from improved stove	% CO reduction	Reference
Three-brick stove, a cooking pot placed over the bricks uses fire wood	Made of clay and husk, two cooking pots, and a stack	10 ppm	2.5 ppm	75	(Jamali et al., 2017)
	stoves with chimneys or smoke hoods	11.9 ppm ^a	5.1 ppm ^b	57.1	(Pope et al., 2017)
	stoves without chimneys or smoke hoods	10.8 ppm ^a	6.6 ppm ^b	38.7	(Pope et al., 2017)
	Charcoal stoves	27.4 ppm ^a	9.6 ppm ^b	64.9	(Pope et al., 2017)
	Advanced combustion stoves	11.3 ppm ^a	5.7 ppm ^b	49.7	(Pope et al., 2017)
The Hifadhi stove has an air entrance and a combustion chamber.	Galvanised Gasifier cook stove	42 ppm	20 ppm	52.0	(Njenga et al., 2016)
Three stone cook stove - burning prunings, maize cobs, and coconut shells	Galvanised Gasifier cook stove	36.5 ppm	20 ppm	45	(Njenga et al., 2016)
Three-stone open fires for burning wood	Metal braziers for burning charcoal	19.4 ppm	7.6 ppm	60.8	(Tagle et al., 2018)
Open in the home, or in an annexed kitchen or outside the home, or under a ledge along outer house wall	Stove with chimney, in good condition; little to no visible damage	5.0%	4.6 %	8.0	(Lucarelli et al., 2018)
Open fire or poorly designed combustion chambers- burning wood	Improved stove: with a chimney and a better combustion system – burning wood	14.3 ppm	1.8 ppm	87.4	(Clark et al., 2009)
The traditional stationary hearth and the portable hearth. All burn biomass	Philips advanced biomass combustion stoves, with two-stage combustion & forced air	30 ppm	7.4 ppm	75.3	(Mukhopadhyay et al., 2012)
Traditional three-stone open fires for burning wood	Envirofit B1200-Natural Draft (rocket stove)	9.6 ppm	6.4 ppm	33.3	(Sambandam et al., 2015)
	Envirofit G3300 Natural Draft rocket stove	10.2 ppm	7.5 ppm	26.5	(Sambandam et al., 2015)
	Prakti Leo-Natural Draft (rocket stove)	11.6 ppm	4.7 ppm	59.5	(Sambandam et al., 2015)
	Philips-Natural Draft (micro gasifier)	3.6 ppm	3.2 ppm	11.1	(Sambandam et al., 2015)
	Philips-Forced Draft (micro gasifier)	29 ppm	9.6 ppm	66.9	(Sambandam et al., 2015)

Traditional 3 stone	Oorja forced draft micro gasifier using pellets	4.3 ppm	2.7 ppm	37.2	(Sambandam et al., 2015)
	Eco Chula - Electric fan-assisted gasifier	6.5 ppm	5.4 ppm	16.9	(Yip et al., 2017)
	EcoZoom - Improved rocket	6.5 ppm	6.7 ppm	-3.1	(Yip et al., 2017)
	Envirofit - Improved rocket	6.5 ppm	4.9 ppm	24.6	(Yip et al., 2017)
	Philips - Electric fan-assisted gasifier	6.5 ppm	3.8 ppm	40.0	(Yip et al., 2017)
	Prakti - Double pot rocket with chimney	6.5 ppm	4.5 ppm	30.8	(Yip et al., 2017)
	Built-in rocket stove	6.5 ppm	4.4 ppm	32.3	(Yip et al., 2017)

^aBefore intervention. ^bAfter intervention. Interventions to reduce exposure to household air pollution can be classified broadly as (i) those acting to change the primary household fuel, (ii) those promoting cleaner-burning and more efficient solid fuel stoves, (iii) those improving the living environment and (iv) those modifying user behaviour.

In India, (Ministry of New and Renewable Energy - India, 2015) approving stove designs was based on their performance as per BIS 13152(part 1) 2013 specification. These include domestic size stoves with natural draft operation having a thermal efficiency in the range 25 – 33.57 % and CO emissions in the range 2.50 – 4.64 g/MJ. Similarly, domestic size cookstoves with forced draft systems had a thermal efficiency range of 35.3 – 40.90 % and CO emission range of 1.12 – 3.2 g/MJ. Community size stoves with natural draft were also approved having a thermal efficiency of 28.1 – 30.28 % and CO emission range of 1.15 – 1.73 g/MJ. Forced draft community stoves, on the other hand, had a thermal efficiency of 35.11 – 42.8 % and CO emission range of 0.83 – 1.97 g/MJ.

4.2. Heating systems

4.2.1. Air staging/two-stage combustion

Air staging or two-stage combustion is the introduction of overfire air into the furnace. During the furnace overfire air technology, combustion air is injected into the system that separates it into primary and secondary flow sections. The aim is to convert all the CO to CO₂. About 70-90% of the primary air is introduced into the fuel to produce an oxygen-rich, low temperature, fuel-rich zone which minimizes the amount of CO formed. On the other hand, 10-30% of secondary air is injected through nozzles above the combustion area in a special wind-box. This increased flow volume completes the combustion process. Figure 8, shows a representation of a modern and a traditional masonry heater employing air staging.

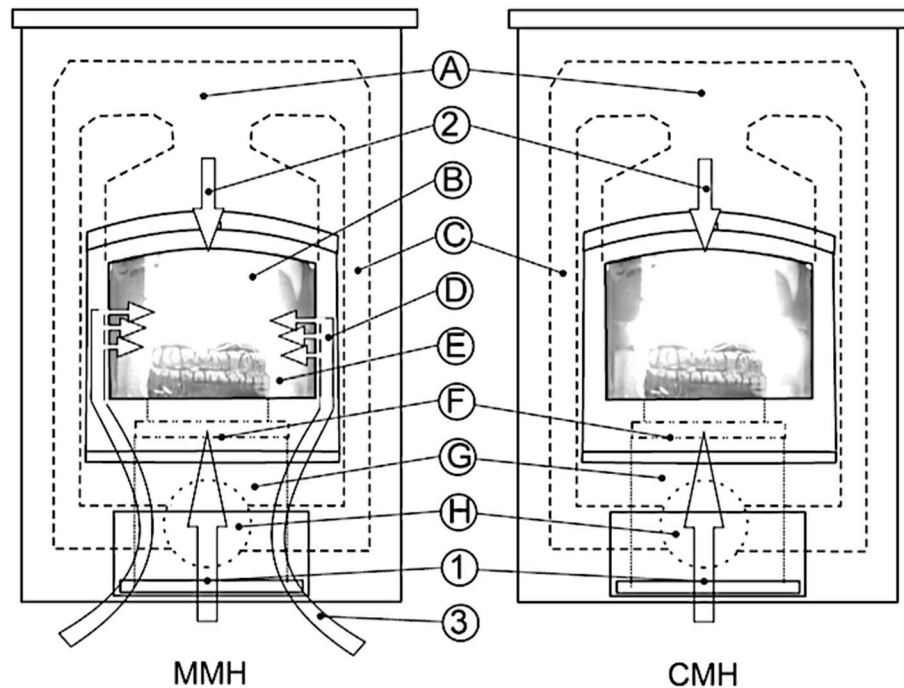


Figure 8. Representation of a Modern (MMH) and Conventional Masonry Heater (CMH). 1, Primary air supply; 2, Window flushing air supply; 3, Secondary air supply; A, Upper combustion chamber; B, Secondary combustion zone; C, Flue gas ducts; D, Secondary air flow between ceramic plates; E, Primary combustion zone; F, Rift grate; G, Ash box; H, Flue gas exhaust to chimney (behind the heater) (Nuutinen et al., 2014).

With air staging, the samples for analysis of emissions are collected from the flue gases arising from the combustion system/boiler outlet/stack of the boiler. In 2002, a 30 kW boiler (Ross et al., 2002) with a ceramic chimney-stack fitted with temperature and pressure sensors to burn Polish bituminous coal, Lump wood pine, Pine sawdust briquettes, and Coal + sawdust briquettes. They used on-line flue gas monitoring using IR analysis for CO analysis. They observed that lump wood produced a higher total organic content, methane and CO emission than biomass briquettes. Laboratory wood pellet boiler (25-kW) have also been studied (Lamberg et al., 2011), measuring emissions with ABB Cemas gas-analyzing rack system. Their system produced CO emissions as low as 3.55 mg/MJ and as high as 335 mg/MJ. A similar laboratory masonry heater burning birch wood and CO detection with ABB Cemas Gas Analyzing Rack & FTIR were used (Nuutinen et al., 2014) in 2014. A CO reduction in the range of 26 to 81% was achieved.

A slightly stronger 35 kW ETA Hack35 tilting grate biomass boiler was used a year after (Carroll et al., 2015) burning wood, willow, and cocksfoot. They used a portable gas analyser with NDIR for CO analysis and achieved low CO emissions of 18 mg/Nm³ at a primary split ratio of 1.6. In 2016, a 5–12 kW low-scale combustor (Combustor, 2016) with air staging was analysed while burning biomass and measuring CO with a servomex gas analyser. They observed that CO emissions were in the range of 0.2 – 0.6 % with 6% oxygen and air excess of 1.4 – 2.0. A year later, laboratory-scale reactor (Khodaei et al., 2017), burning standard cylindrical wood pellets were used to simulate CO reduction strategies. Emissions were analysed with Servopro 4900. They observed that CO was in the range of 0.1–0.5%.

In the same year (2017), (Sher et al., 2017), a 20 kW bubbling fluidised bed combustor burning domestic & industrial wood, miscanthus, straw, and peanut shell pellets was investigated. They measured CO with ABB, EL3020 analyser and observed only 0.1 – 0.6 vol% as CO emissions. Still, in 2017, a hybrid stove burning biomass was able to reduce CO emissions by 65% (Lamberg et al., 2017). In 2018, a 140-kW fixed bed biomass boiler (Caposciutti and Antonelli, 2018) was used for burning biomass and they observed that low CO emissions were obtained for the split ratio of 0.4 and air

excess of 1.5 –2.5. In the same year, an OP 230 boiler - two-pass drum boiler burning pulverized hard coal was used to investigate an air-staging system (Hernik et al., 2018). They observed that low CO emissions were achieved where overfire air was perpendicular to the surface of the nozzles.

4.2.2. Improved boiler systems

The improved boilers work by reducing incomplete combustions through the optimization of the combustion system due to the improvement of boiler efficiency and CO emissions reduction by boiler manufacturers with or without the automatic airflow control system. They may have continuous or periodic feeding operations (Johansson et al., 2004). The materials used for such boiler systems range from sewage sludge, coffee husks, wheat straw, briquettes made from; corn stover, rye straw, miscanthus, cherry stones, cylindrical briquettes, hay and sunflower husk pellets, pellets mixture (deciduous and coniferous wood). The feedstock could also be basket willow-chips, rape straw-briquettes, Wood waste-pellets, Poplar firewood-chips, Sawdust, Oak bark shavings, from a sawmill, Rape cake, coal. Wood pellets, wood briquettes, wood logs and oil, grain whole crop (triticale), hay, and forest residue wood could also be used. The woods could be of particle length of 1 to 2 cm, while the herbaceous fuels could be as both, pellets (12 mm diameter, 25 mm mean length) and chopped material (20 mm mean chopping length).

In the year 2000, a 15 and 50 kW modern multi-fuel furnace (Launhardt and Thoma, 2000) that burnt biomass and wood pellets were investigated for their effectiveness in reducing CO emissions. They observed that CO-emissions were all around 200 mg/m³; related to the fuel type. Four years later, CO emissions from Old-type wood boilers (6-24 kW), Modern wood boilers (12-34 kW), Pellet burners (3-11 kW), and Oil burners (18-21 kW) (Johansson et al., 2004) were also analysed. They observed that long residence time, high temperature and adequate mixing of air & combustible gases reduced CO emissions.

Using a 50 kW Arimax 340 fixed retort system biomass boiler (Bignal et al., 2008), the full flame boiler conditions had low CO while the slumber mode had high CO emissions. Figure 9, shows the relationship between the total PAHs and CO emissions for the different boiler operating conditions. A year later (Ozil et al., 2009), two 13 kW domestic wood heating fireplaces with catalyst systems: cordierite honeycomb monolith and metallic corrugated structure in their chimney were investigated. They observed that a switch of normal to low-charge phase increased CO emissions. Having a catalyst in the chimney reduced CO emission by up to 80% at ignition and to 90% in the low-charge phase. In 201, a 6 kW pellet stove; a 40 kW biomass boiler; a 6.5 kW simple logwood stove and a 6 kW logwood stove (Schmidl et al., 2011), were investigated for their effectiveness in minimising CO emissions. It was observed that full load conditions in automatically fired systems exhibited very low CO emissions compared to manually fired systems.

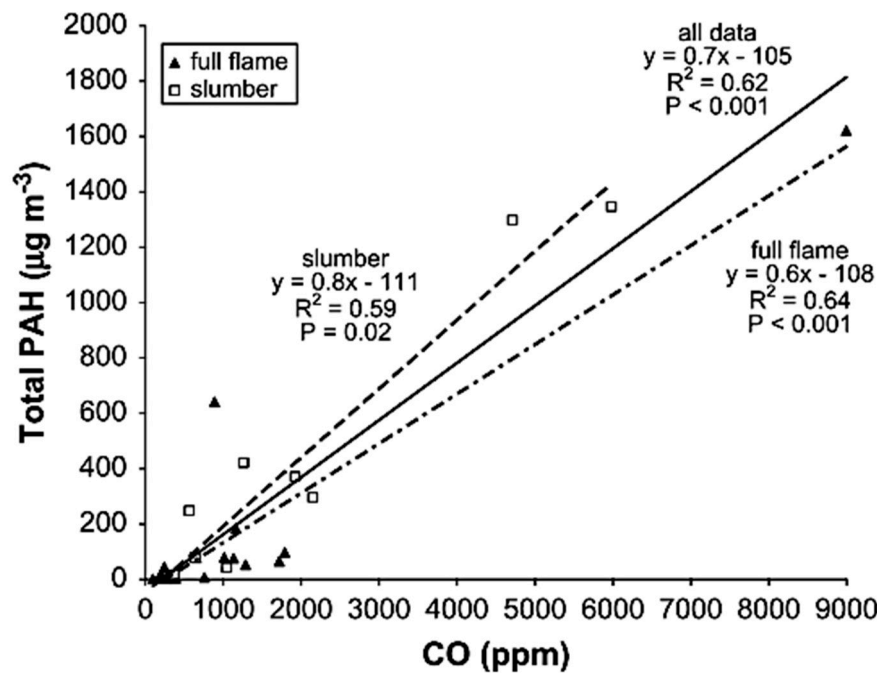


Figure 9. Release of polycyclic aromatic hydrocarbons, carbon monoxide and particulate matter from biomass combustion in a wood-fired boiler under varying boiler conditions (Bignal et al., 2008).

A year later (2012), a 15-kW horizontal-feed furnace (Juszczak and Lossy, 2012), was investigated having been installed in a down-draft wood log boiler. They observed that the higher the proportion of wood pellets in the mixture the lesser slag and the lower CO in the flue gas. In the same year, a vertical laboratory-scale furnace (Jeguirim et al., 2012) that burnt date palm residues was used for CO investigations. They used a Rosemont IR analyzer for CO analysis. It was observed that secondary air supply and proper air mixing reduced CO emissions. Again in 2012, the use of a 20 kW wood pellet boiler, and a pellet stove combined (Win et al., 2012) with a solar heated buffer store were analysed. They observed that the pellet stove with efficient air staged combustion produced the lowest CO.

In 2013, (Rabaçal et al., 2013), observed that a 22 kW domestic wood pellet-fired boiler could reduce CO emissions if operated at high temperature and high air excess. Juszczak (Juszczak, 2014), in 2014 observed that a 25 kW water boiler equipped with an over-fed wood pellet furnace conditions burning ECOPELLET wood pellets reduced CO emissions as oxygen in the system was increased. In the same year, (Win and Persson, 2014), using a 12, 20, and 25-kW wood pellet boilers and a pellet stove of the nominal power of 12 kW observed that the CO emissions increased with decreasing combustion power.

In 2015, (Calvo et al., 2015) used a manually operated wood stove with handheld control of combustion air, and a traditional Portuguese brick open fireplace to investigate toxic combustion emissions. They observed that CO emissions from the fireplace were higher than those observed for the stove. The following year, (Juszczak et al., 2016) investigated a 20-kW full-scale heat station connected with district heating network burning biomass pellets and Coal for emission control. They observed that increased temperatures and oxygen availability reduced CO emissions. Again in 2016, (Tschamber et al., 2016) performed experiments with an airtight cast-iron stove that burnt wood logs. Low CO emissions were recorded with the airtight cast-iron stove in comparison to other stoves and fireplaces of the 1990s or new ones without post-combustion system. In the same year, (Mantanant and Patumsawad, 2016) used a lab-scale fixed-bed combustion system burning Thai lignite and agricultural residues, rice husk, rice straw, and bagasse to investigate the toxic combustion emissions. They observed that CO emissions reduced considerably as secondary air/total air ratio was increased.

In 2017, (Zajac et al., 2017) investigated a 32-kW boiler with automatic fuel load burning wood pellets with UL- TRAMA T 23 analyser for emission analysis. They observed that temperature and fuel type influenced CO emissions. In the same year, (Wielgosinski et al., 2017) used an electric resistance furnace and a pipe furnace with a horizontal working chamber to analyse emission reduction effectiveness. They found out that increasing aeration decreased the amount of CO, especially at elevated temperatures. More recently, (Byul et al., 2018) used a 3.0 MW KFS subsidized industrial wood pellet boiler to monitor emission reduction for 3 years. They observed that the system was able to achieve a 58.9 % CO reduction between 2012 to 2015. In the same year, (Pałaszynska and Juszcak, 2018) used a 50 kW boiler type Bio-warmer that burnt sewage sludge, coffee husks, wheat straw, biomass briquettes, and miscanthus to investigate combustion emissions. They observed that except for sewage sludge pellets, CO did not exceed 3000 mg/m³.

The main challenges facing cooking and heating systems are variation in fuel quality and proper mixing to optimize the available air for enhanced oxidation. Feedstocks are obtained from different locations and their growth patterns, soil conditions, nutrient availability and climate dictate the final quality (moisture content, volatiles, fixed carbon, mineral content, among others). Applying different recipes like single biomass species batches, mixed biomass, co-firing, single coal batches, mixed coal batches have all been applied to solve this challenge. With optimizing air mixing, self-adjusting controls that use sensors have been used for CO, temperature and air-excess (Nussbaumer, 2003). But still, the mixing of combustibles with air is a challenge. The application of computational fluid dynamics for extended simulation and modelling is being used to solve the mixing challenge.

4.3. Catalytic oxidation of CO

4.3.1. Carbon monoxide and oxygen chemisorption on metals

Carbon monoxide

The adsorption and reactivity of carbon monoxide on oxides and metals is a result of CO having three resonance structures. By donating electrons through the 5s orbital and accepting electrons through the antibonding 2p* orbital makes it possible for carbon monoxide to be coordinated to one or numerous species. Five spectral ranges of CO adsorption bands can be detected depending on the number species it is bonded to 1700–1800, 1800–1920, 1860–2000, 2000–2130, and 2130–2200 cm⁻¹. The particle size and the nature of the metal site will determine the IR band position. Platinum, rhodium, and palladium have been used to oxidise CO and have given infrared vibration bands at different positions depending on the orientations (mirror indices) of these metals. Chemisorption behavior of carbon monoxide on metals also varies considerably: for Pt, Rh, and Ru, the heat of chemisorption is directly proportional to the metal surface density. Also, the heats of chemisorption are inversely proportional to CO coverage (q_{CO}) with platinum having the highest sensitivity (Royer and Duprez, 2011).

Oxygen

For CO oxidation to be fully understood, the chemisorption of oxygen on catalyst surfaces must be understood as well. Unlike CO, adsorption of oxygen is dissociative. The heat of adsorption on most non-noble transition metals is mostly not affected by oxygen coverages. However, noble metals (Rh, Pd, Pt) have inverse relations of the heat of chemisorption with oxygen coverage. This is due to differences in the metallic radius of the metals Figure 10.

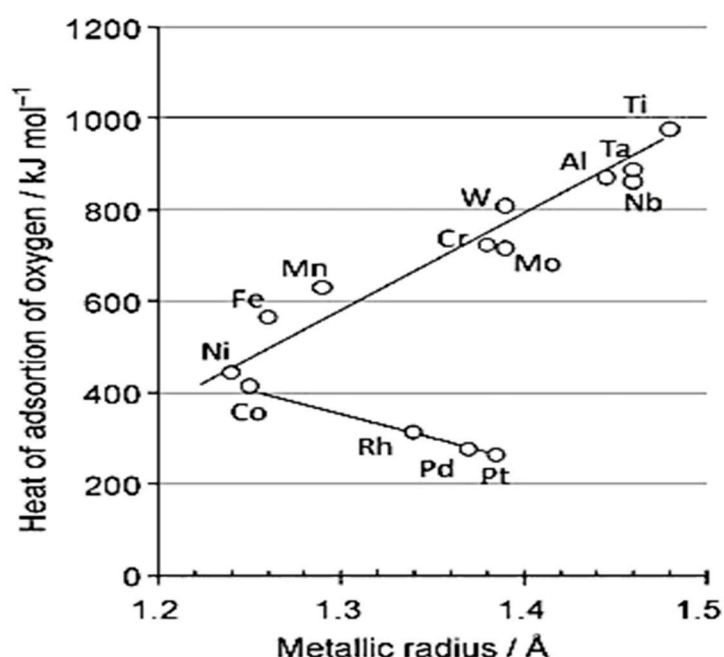


Figure 10. Heat of adsorption values of metals, as a function of their metallic radius. Adapted from (Royer and Duprez, 2011).

4.3.2. Laboratory oxidation of CO over metals

Supported metal catalysts

The presence of catalyst support may have a significant effect on the catalyst activity and the overall reaction. The final reaction usually occurs on the catalyst or at the catalyst/support interface. Secondly, the oxidation/reduction state of the metal catalyst has a major implication on CO oxidation (Royer and Duprez, 2011).

CO oxidation over simple oxide catalysts

Much as noble metals are very active for oxidation of CO, some oxides especially Cu and Co have shown remarkable activity as well for CO oxidation. The reactivity of noble metals and base metal oxides for oxidation of 1% CO in excess oxygen and 300 °C follows the order; Pd > Pt > Co₂O₃ > CuO > CuO/Cr₂O₃ > LaCoO₃ > Au > MnO₂ > Fe₂O₃ > Cr₂O₃ > NiO (Royer and Duprez, 2011).

Gold-based catalysts

The ability to change oxidation states by gold and ceria has played a great role during the catalytic oxidation of carbon monoxide (R. Zhang et al., 2017). This is also due to their oxygen vacancies, and high oxygen storage capacity of this catalyst system. The shape of ceria as support also influences the catalytic activity of gold for CO oxidation. Carltonbird *et al.* (Carltonbird et al., 2018), observed that the order of reactivity of gold catalyst in relation to the shape of ceria support was; rod-shaped > polyhedral > cube > octahedral shaped CeO₂. However, the addition of iron to the Au/CeO₂ system improved the catalytic activity for oxidation of CO and benzylamine compared to Au/CeO₂ alone (Sudarsanam et al., 2014). A gold/titania-based catalyst is a unique combination in that on top of achieving 100% conversion of CO at room temperature, it displayed remarkable CO oxidation at 120 K (Lee and Zaera, 2014). Other researchers have reported a gold catalyst supported on various supports for the oxidation of CO and alcohols. Such systems included Au/activated carbon, Au/boron nitride, Au/titanium dioxide, and Au/mesoporous silicate-based (SBA-15) (Song et al., 2017). Perovskites (LaMnO₃, LaFeO₃, LaCoO₃, and LaCuO₃) have also worked excellently as

supports for gold. With only 1wt% of gold, superior activity was registered for CO oxidation (Mokoena et al., 2016). Gold supported on three dimensional Mn_2O_3 was used for CO, and toluene oxidation (Xie et al., 2014). At only 15 °C, up to 90% conversion was achieved. This was attributed to its low-temperature reducibility, high oxygen adsorption capability, high porosity and the strong interaction between gold nanoparticles and the three-dimensional Mn_2O_3 . In other studies, a gold-copper alloy supported on MgO or graphene (Koizumi et al., 2015) has been used for efficient oxidation of CO. This combination was preferred for its effectiveness and commercial affordability compared to other gold-based catalysts.

Copper-based catalysts

Water-resistant SnCu30 (30wt%Cu) catalysts have been prepared and used for oxidation of CO (Bai et al., 2017). The high activity was attributed to the lattice surface oxygen and the highly dispersed Cu^+ species within the Cu-SnO₂ catalyst matrix which provided active sites for adsorption of CO. Un-supported CuO uses the surface area advantage for oxidation of CO. A decrease in specific surface area of CuO from 90 to 8 m²/g resulted in a four-fold increase in the specific catalytic rate (Svintsitskiy et al., 2016). However, a 20-fold decrease in the specific catalytic rate was observed when the catalyst specific surfaces area was reduced to 1 m²/g. Activated-red-mud as support on CuO produces a well-structured porous and high surface area catalyst (Hu et al., 2016) for CO oxidation. However, the CO oxidation is dependent on CuO loading and the catalyst calcination temperature. Copper-Manganese (commercially known as hopcalite) prepared by flame spray pyrolysis produced a superior hydrophobic catalyst that was resistant to humidity levels as high as 75% (Biemelt et al., 2016). This was achieved by applying 2-ethyl hexanoate as precursors achieving a high specific surface area catalyst (180 m²/g) composed of $\text{Cu}_{1.5}\text{Mn}_{1.5}\text{O}_4$ as the main phase. A more sophisticated Cu-based catalyst composed of Cu-Pd/CeO₂ (Du et al., 2017) achieved excellent activity for CO oxidation. This was attributed to full oxidation of the Cu and Pd by the ceria support which dominated the catalyst system providing an excellent metal/support interaction.

Cobalt catalysts

The catalyst preparation method greatly impacts its performance. Dispersion-precipitation synthesis of Co_3O_4 produced a better catalyst for oxidation of carbon monoxide and propane than alkali-induced precipitation (W. Zhang et al., 2017). Dispersion-precipitation produced a more reducible Co_3O_4 catalyst with high numbers of active surface oxygen species. The activation energy for CO and C₃H₈ oxidation were reduced by 38% and 31% respectively with the dispersion-precipitation synthesised Co_3O_4 compared to alkali-induced precipitation synthesised Co_3O_4 . Titania supported Co_3O_4 catalyst prepared by incipient wetness impregnation had a superior activity for CO and hydrocarbon oxidation in vehicle exhaust emissions compared to $\text{Co}_3\text{O}_4/\text{TiO}_2$ catalyst prepared by wet impregnation method (W. Ahmad et al., 2017). Similarly, the activity of hollow type Co_3O_4 spheres prepared using silica templates for CO oxidation was dependent on calcination temperature with 623 K producing the best activity (Umegaki et al., 2016). These hollow sphere Co_3O_4 catalyst also showed superior activity compared to supported Co_3O_4 as well as Co_3O_4 nanoparticles. In a similar situation, Cao *et al.* (Cao et al., 2014) observed that the wormhole-like-mesoporous Co-Fe-O catalyst activity for CO oxidation was dependent on Co loading (Figure 11), the surface area, the pre-calcination temperature, and the particle size.

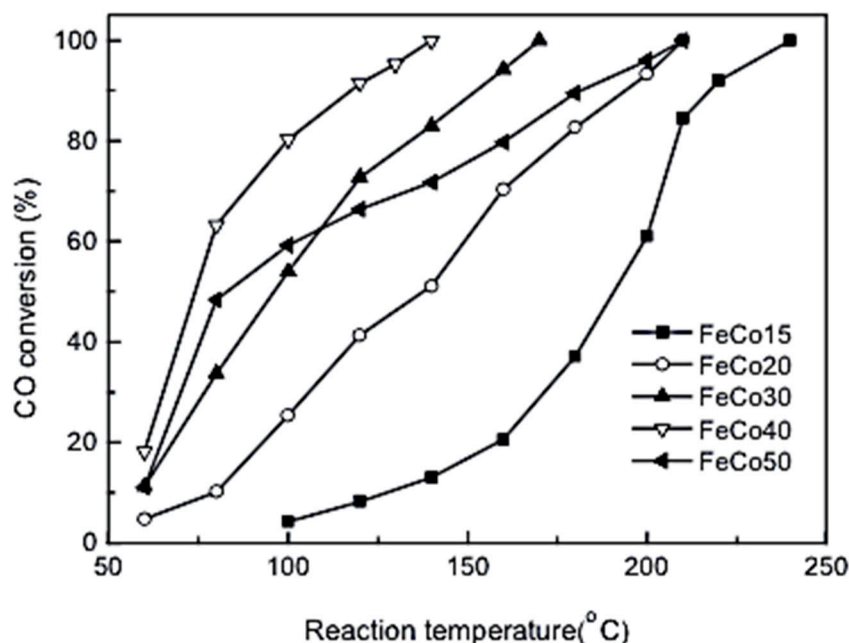


Figure 11. Mesoporous Co-Fe-O nanocatalysts: Preparation, characterization and catalytic carbon monoxide oxidation (Cao *et al.*, 2014).

Platinum and Palladium catalysts

Being noble metals, a small concentration is needed in addition to a support for efficient CO catalytic activity. With only 0.5wt%Pd/manganese oxide catalyst, maximum CO, toluene, and ethyl acetate oxidation were achieved at 55°C compared to un-supported Pd catalyst. This excellent activity was attributed to high oxygen adsorption and low-temperature reducibility of the 0.5wt%Pd/manganese oxide catalyst. With LaMnO₃ Perovskites, palladium loading was directly proportional to CO catalytic activity (Kucharczyk, 2015). The activity was seen to increase with the temperature especially in the range 650 to 800 °C.

Pt/CeO₂ catalysts prepared by impregnation-reduction (Hong and Sun, 2016) had the best activity for complete oxidation of CO at room temperature compared to the same catalyst prepared by impregnation, and deposition-precipitation methods. The activity of the impregnation-reduction prepared Pt/CeO₂ catalyst was attributed to possessing highly numbers of negative Pt species for efficient adsorption of oxygen for CO oxidation. Similarly, acid treatment followed by thermal activation of palladium-copper complexes affected the catalyst's activity for CO oxidation (Rakitskaya *et al.*, 2016). The best results were obtained when the bimetallic Pd(II)-Cu(II) complexes were treated with 3M HNO₃ for 0.5 and heat-treated for 60 minutes.

Ceria based catalysts

Ceria being a base metal oxide works better with a support and at intermediate temperatures. It was reported that ceria/alumina could achieve complete oxidation of 7739 ppm of CO at 400 °C in excess oxygen compared to un-supported ceria (Wilklow-Marnell and Jones, 2017). Similarly, a ceria-zirconium catalyst system doped with low amounts of Nd (<0.1wt%) was reported to enhance the surface area of the catalyst thereby improving the catalytic activity for oxidation of CO (dos Santos Xavier *et al.*, 2015). High Nd content (0.2 – 0.3) promoted sintering and retarded the catalytic activity of Ce-Zr-Nd catalyst for CO oxidation.

Other catalysts

Aromatic organic chemical tetrapyrrole (commonly known as corrole) combined with transition metals (corX; X standing for the transition metal) have also been used for catalytic oxidation of CO.

In one study, the metals Al, Ga, Si, Ge and As were tested (Mohajeri and Hassani, 2018). The best results were obtained from the CorAl and CorGa while CorSi and CorGe formed very stable carbon-like intermediates that hindered CO oxidation. Other researchers have used Li_5FeO_4 and LiFeO_2 (Lara-García et al., 2017), V/graphene (Tang et al., 2018), Cr/graphene (Dai et al., 2018), and ZrO_2/MgO for oxidation of CO.

The ideal catalyst for CO oxidation should have high selectivity, high activity, thermally stable and long life, all of which come at a high cost. Noble metals are the best choice, but they are very expensive. Research into new materials such as Mn, Fe, Co, Ni, Cu or their combinations has been extensive (Rola Mohammad et al., 2018), however, improving their stability is still a challenge. Even the reaction itself between CO and O_2 on catalyst surfaces has had over 20 different proposed mechanisms with the Langumir-Hinshelwood (LH) being the best. Understanding the dynamic reactions at microscopic level on surface and the bulk of the catalyst under heating and cooling is the task at hand for researchers.

4.4. CO detection technologies/sensors

4.4.1. Sensors

The development of CO sensors started in the mid-1900s, but the actual reported sensors became operational in the 1970-1980s (Windischmann and Mark, 1979). The early designs had chemical pads that changed to a brownish or blackish color when exposed to CO. As CO related deaths became rampant, audible alarms became the standard. The alarm response on the CO detector uses a concentration-time function. Lower CO concentrations would not trigger the alarm for a given period, however, higher CO levels would cause the alarm to go off in a few minutes. This concentration-time function mimics the CO assimilation in the body while preventing false alarms due to common sources of CO like cigarette smoke.

Colorimetric CO sensors

Several sensors have been designed especially using mono, di, and tri-rhodium complexes. Binuclear rhodium complexes $[\text{Rh}_2\{(\text{XC}_6\text{H}_3)\text{P}(\text{XC}_6\text{H}_4)\}_n(\text{OCR})_2]$, when exposed to 100-300 ppm CO would change from purple to red within 20 seconds at room temperature (Pannek et al., 2018). A potassium-palladium complex ($\text{K}_2\text{Pd}(\text{SO}_3)_2$) with a high response at room temperature would have a detection limit of 1 ppm (Lin et al., 2018). Similarly, a Rh complex activated with ethanol and tributyl phosphate would also change from purple to red upon exposure to only 10 ppm CO within 30-60 seconds. Some designs are even more sensitive that they can detect CO levels as low as 5 ppb at room temperature (Toscani et al., 2015).

Ruthenium (II) complexes react extensively with small-donor ligands such as CO. This concept was used to design CO colorimetric probe which expresses both chromo- and fluorogenic response. At low CO levels as 5 ppb, a color response could be observed by the naked eye with clear changes from orange to yellow seen at 100 ppm of CO in air. When the turn-on emission fluorescence was increased, substantial sensitivity to 1 ppb of CO could be achieved in air and in solution. The system is insensitive to moisture and other gases (Moragues et al., 2014).

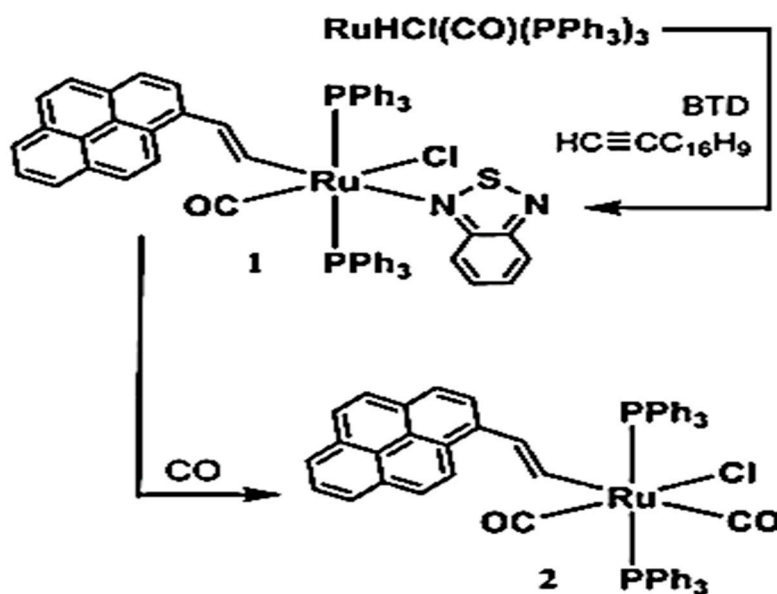


Figure 12. Formation of the Complexes $[\text{Ru}(\text{CH}=\text{CHPyr1})\text{Cl}(\text{CO})(\text{BTB})(\text{PPh}_3)_2]$ (1) and $[\text{Ru}(\text{CH}=\text{CHPyr1})\text{Cl}(\text{CO})_2(\text{PPh}_3)_2]$ (2) (Moragues *et al.*, 2014).

Electrochemical

These fuel cells produce a signal current that is proportional to the concentration of the target gas (i.e. CO). The electrochemical cell has a container, the working and counter electrodes, connection wires and an electrolyte – usually an acid. The oxygen available at one electrode is used to oxidise CO at the other electrode to carbon dioxide which is detected. Electrochemical cells have high accuracy and linear output to CO concentration, require minimal power, are operated at room temperature, and have a long lifetime (i.e. ≥ 5 years).

At the onset of the 1980's, (Okamoto *et al.*, 1980) developed an electrochemical sensor based on Yttria-stabilized zirconia as the electrolyte and Pt electrodes. For 160 ppm of CO, it took 300 seconds for the sensor emf to reach 90% of its saturation at 300 °C. Ten years later, (Otagawa *et al.*, 1990) used a Nafion® membrane as an electrolyte with Pt electrodes to design a CO sensor that could detect 500 ppm of CO within 100 seconds at 20-22 °C. Towards the end of the 20th century, (Tess and Cox, 1998) developed a silica electrolyte based sensor with Pt electrodes that could detect 67 ppm of CO within 2-50 seconds at 20-22 °C.

In 2007, (Santhosh *et al.*, 2007) showed that a multiwall carbon nanotubes and poly-diphenylamine on a glassy carbon working electrode and Pt counter electrode would work with HClO_4 , H_2SO_4 , HCl and H_3PO_4 electrolyte to detect 100 ppm of CO within 2 seconds at room temperature. Two years later, (Paul *et al.*, 2009), used a polypyrrole/Au/standard calomel working electrode and Pt counter electrode to design CO electrochemical cell with pyrrole monomer dissolved in acetonitrile and tetrabutylammonium perchlorate as an electrolyte. The sensor could detect 300 ppm of CO at room temperature within <1 second. (Zhi *et al.*, 2012) observed that a cell constructed from $\text{La}(\text{CH}_3\text{COO})_3 \cdot 1.5\text{H}_2\text{O}$, $\text{Sr}(\text{CH}_3\text{COO})_2 \cdot 0.5\text{H}_2\text{O}$ and $\text{Mn}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}$ (Pt) as working electrode and Yttria-stabilized zirconia as electrolyte could detect 200-500 ppm CO at 550 °C within 65-80 seconds (Figure 13). In the same year, (Phawachalotorn *et al.*, 2012) observed that a RuO_2 (Pt) working electrode cell with Au (Pt) as counter electrode and $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.2-x}\text{Fe}_x\text{O}_3$ and $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{1-x}\text{Fe}_x\text{O}_3$ as electrolyte could be used to detect 1000 ppm of CO at 400 °C within just 120 – 180 seconds.

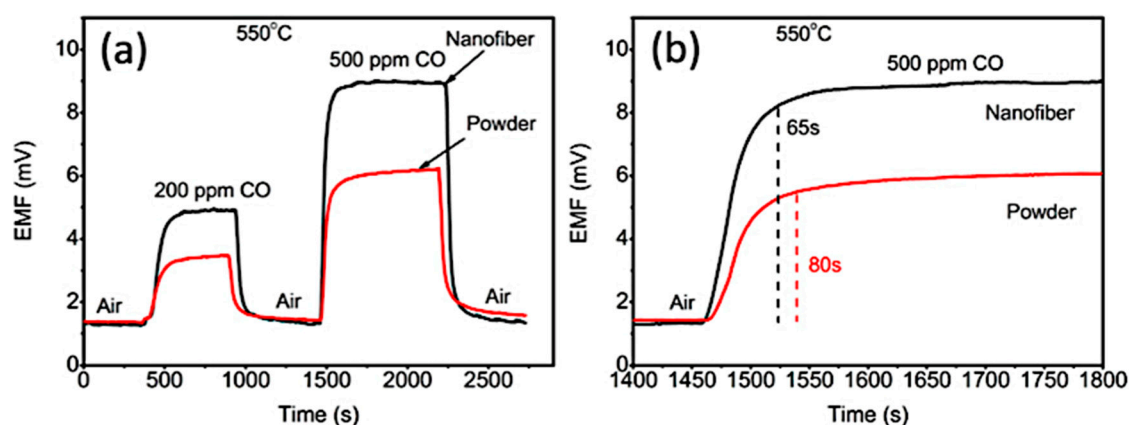


Figure 13. Response to change in CO concentration at 550 °C for the nanofiber sensor and the powder sensor (a) and response time (b) (Zhi et al., 2012).

Using pure single-walled carbon nanotube (SWCNT), CuCl-doped SWCNT, and pure CuCl (Zhang et al., 2013) as working electrodes and Au as a counter electrode with SiO₂/Si as the electrolyte, a CO electrochemical sensor was constructed. They observed that 20-100 ppm CO could easily be detected at room temperature. (Zhu et al., 2015) realized that PdO powder & α -terpineol as working electrode and Pt as a counter electrode with yttria-stabilized zirconia could be a good combination for detection of CO. They observed that 20-200 ppm of CO could be detected in 118 seconds at 450 °C. In 2016, (Izu et al., 2016) used Pt for both working and counter electrodes with zirconia-doped ceria (Ce_{0.9}Zr_{0.1}O₂) as the electrolyte and observed that 1000-5000 ppm of CO could be detected in 2-4 seconds at 400-500 °C. In the same year, (Shimizu et al., 2016) used Au/SnO₂ as working electrode and Au as a counter electrode with anion-conducting polymer as the electrolyte for a CO sensor. They observed that 10-3000 ppm of CO could be detected in 14-300 seconds at 30 °C. All these developments show a remarkable stride in improving the detection of CO with electrochemical sensors.

Semiconductor

The circuit system of semiconductor sensors consists of thin wires of the semiconductor material on an insulating ceramic base. This sensing element is heated to an appropriate temperature for proper operation. Exposure to CO would reduce the resistance while oxygen would increase it. The integrated circuit works by monitoring the resistance of the sensing element (Bornand, 1983). Semiconductor sensors have a longer shelf life (i.e. >10 years), however, they require higher power compared to electrochemical sensors.

In 1983, (Bornand, 1983) developed a tin oxide sensor with Ag & Au electrodes to detect 1750 ppm of CO within 100 seconds at 400-600 °C. Five years later, (Formosa et al., 1988) developed a Pd-gate sensor that could detect 10,000 ppm of CO at <200 °C within 60 seconds. In 1994, (Cantalini et al., 1994) developed an α -Fe₂O₃ microporous ceramic sensor that was very sensitive to 300 ppm of CO at 300-400 °C. (Davis et al., 1998) investigated the effects of crystallite growth and dopant migration on the CO detection properties of nanocrystalline SnO₂ sensor in 1998. They observed that 300 ppm of CO could be detected very fast if the sensor was tuned to 200 – 400 °C temperature.

In 2002, (Sin et al., 2002) also investigated the sensing properties of SnO₂-Cu/Pt/SiO₂ for 1000 ppm of CO. The developed sensor could detect CO within 5 seconds at 270-320 °C. In 2006, (Wang et al., 2006) used Au/Sn sensors-based gold/tin dioxide, to detect 500 ppm of CO within 30-70 seconds at 81-210 °C. In 2011, (Blondeau-Patissier et al., 2011) investigated the surface acoustic wave devices in combination with Cobalt corrolles to detect 4.5 ppm CO at room temperature. The sensor was able to detect the low-level CO within 100 seconds. (Anastasescu et al., 2016), found out that a SnO₂-ZnO sensor could detect 50-2000 ppm of CO within 120 seconds at 210 – 300 °C. More recently (P. Kumar et al., 2018) found out that ZnO and Fe-ZnO thin films could detect CO at 100 – 600 °C within 3-5

seconds. This year (2019), (Ortega et al., 2019), observed that europium doped ceria sensors (Figure 14) could detect CO at 100 – 400 °C within 56 seconds. More semi-conductor sensors are given in Appendix A - Table 1.

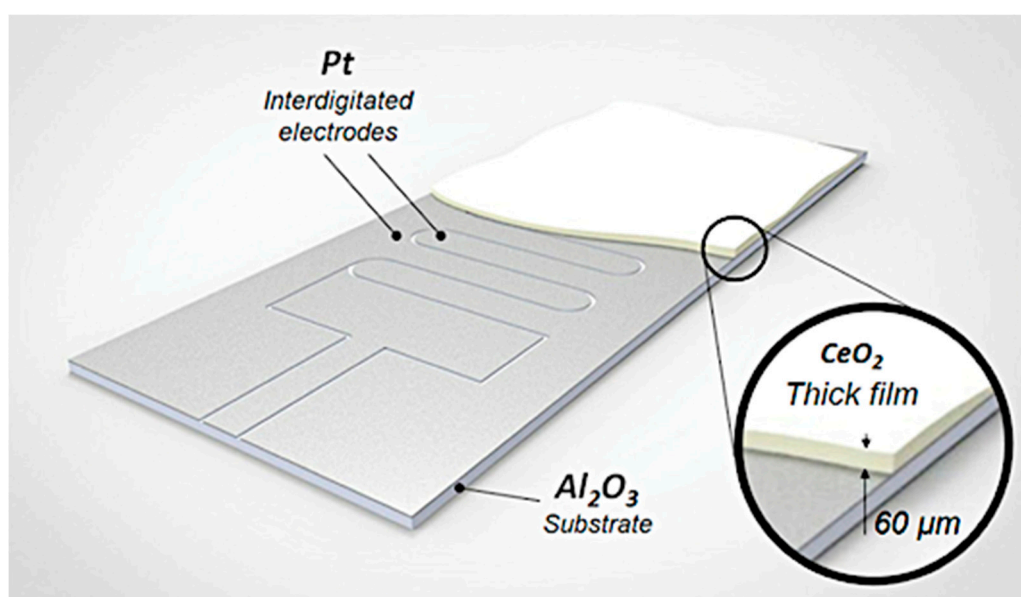


Figure 14. Illustration of the deposited ceria film on alumina substrates with interdigitated Pt electrodes (Ortega et al., 2019).

Infrared sensors

These sensors work by analyzing the absorption signature wavelength of the target gaseous sample in the electromagnetic spectrum (Li et al., 2012). Carbon monoxide has strong absorption in the 4.6 mm region. (Dong et al., 2017) observed that using a dual-channel pyroelectric detector with a self-developed digital signal processor-based orthogonal lock-in amplifier, they could detect up to 1000 ppm of CO within only seven seconds. (Qiu et al., 2019) used a similar system but with a 32-bit microcontroller and detected 125 ppm of CO within 110 seconds.

Continuous-wave mode distributed feedback quantum cascade laser with optical length, and self-development electronic modules (Dang et al., 2018), have also been used for detecting low levels of CO at ambient temperature (Vanderover and Oehlschlaeger, 2010) with quick responses of up to 3 seconds. Similar quantum cascade laser utilizing mid-infrared (Mulrooney et al., 2008), (Barron-Jimenez et al., 2006) wavelength modulation spectroscopy absorption sensor (Vanderover et al., 2011) and very low limits of detection as 0.03 ppm have also had very quick responses to CO.

Laser diodes with thermoelectric coolers as radiation source (Shanying et al., 2010), (Thurmond et al., 2016) have detected 50-5000 ppm of CO within 20 seconds. Some of these have slightly high detection limits of up to 50 ppm (Zhu et al., 2010). Others use a tunable diode laser spectroscopy sensor with a vertical-cavity surface-emitting laser (Hangauer et al., 2014).

Photoionization detectors (PIDs) with UV-assisted Cataluminescent (CTL) sensor based on g-C₃N₄ (Li et al., 2018) and photons platform using a distributed-feedback laser diode operating at room temperature (Thomazy et al., 2010) have been of much success with low detection limits 0.08-3 ppm and response times as a second to as high as 140 seconds.

4.4.2. Wireless systems

Recently, remote warning handsets or strobes, and vibrating pillow pads have been introduced as linkages to wireless safety solutions in homes. These provide warning signals to those with impediments like the deaf, blind and deep sleepers to get up quickly and escape a suspected CO incident in any facility. Metal-oxides like tin oxide (SnO₂) semiconductor sensor, a data acquisition system, and a communication system from remote terminal unit to a web server have been used for

CO detection with an average error of 7.73 ppm, and a mean absolute percentage error of only 2.81% (Suryono et al., 2017). With a three-lead electrochemical CO sensor without battery (2.5 W), and the power consumption of only ~13 mW, a wireless system was set up and used with a Nordic Bluetooth dongle low power protocol to send a trigger to activate Wi-Fi alarms and notifications to the mobile device through the Internet (Chen et al., 2016).

Using sensor nodes consisting of an LM35DZ temperature sensor, a humidity sensor HSM 20-G and a CO sensor TGS 2600, can be used to achieve an average error of 4.414% when compared to the dataset and 2.12% (if the data outliers are eliminated) (Firdaus et al., 2016). Chen, Shi, and Guo used Wireless Sensor Network node, low-power microprocessor C8051F930, with CO metal oxide semiconductor sensor to detect CO in the range of 30 to 1000 ppm with a data transmitting distance of 200 m (Chen et al., 2013). (Bicelli et al., 2009) used a similar system but with TGS2442 (Figaro, Inc.) metal-oxide CO sensors to detect CO in the range of 30 to 300 ppm. The sensitivity obtained was <1s, at room temperature.

The challenge with CO sensor systems are mainly false detection due to age, malfunctions or chemical substance with a similar signature as CO (Ryan and Arnold, 2011). Fitting new sensors, and continuous research and development is the only way to solve this challenge.

5. Catalyst impregnation on to solid fuels

5.1. Enhancing pyrolysis and char gasification

When secondary reactions are triggered during biomass pyrolysis by either alkali or alkali earth metals (Anca-Couce et al., 2017), the resulting char has varying reactivity. This could be due to blocking or deactivation of active sites for oxygen attack. (Kirtania et al., 2017) observed that impregnation of sawdust with K_2CO_3 , Na_2CO_3 , NaOH and NaCl, followed by devolatilising, and pyrolysing the resulting char at different temperatures (750–900 °C) under CO_2 , three classes of chars were produced. Those chars that were highly influenced by catalysts were swollen and had molten surfaces. The moderately influenced chars were wood-like while the least affected had salt deposits. Potassium carbonate (K_2CO_3) gave the best catalytic activity for gasification. When iron or nickel were impregnated on cellulose, the yield and composition of fast pyrolysis products were different compared to un-catalysed cellulose (Collard et al., 2015). With only 1.5wt% Fe or 1.7wt% Ni catalyst loading, cellulose depolymerization was inhibited; char, moisture, and CO_2 yields were increased while tar and CO yields were reduced.

(Yu et al., 2017), observed that bread waste could be turned into hydroxymethylfurfural (HMF) by impregnation of $SnCl_4$, $AlCl_3$, and $FeCl_3$ followed by pyrolysis. The highest HMF yield (30 mol%) was achieved using $SnCl_4$ as the catalyst (Figure 15). The polymerization-induced metal-impregnated high-porosity carbon was a possible precursor of the biochar-based catalyst.

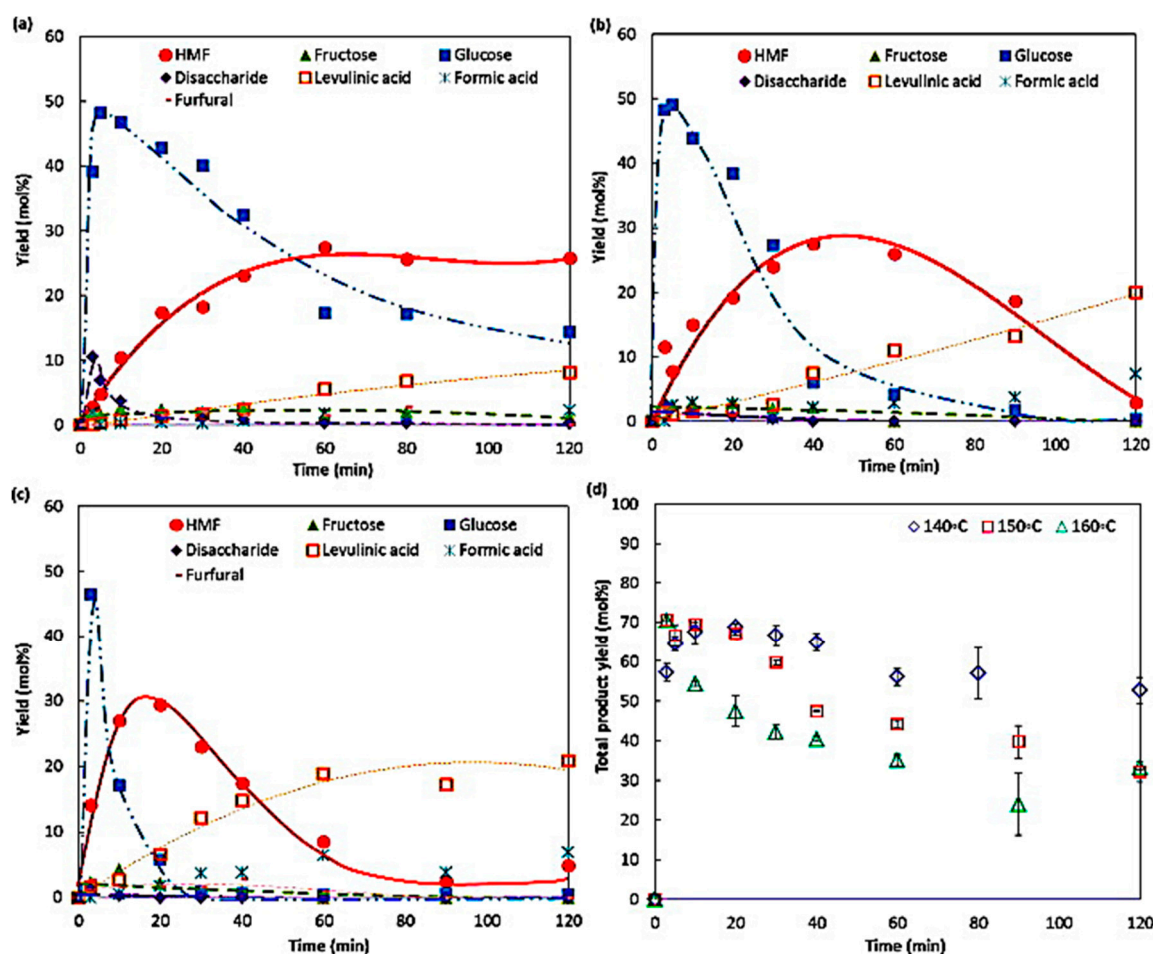


Figure 15. Product yields during the SnCl_4 -mediated conversion of bread waste under heating at (a) 140 °C, (b) 150 °C, and (c) 160 °C; and (d) total product yield at the three temperatures (conditions: 5 wt/v% substrate and 55.5 mM metal chloride in Dimethyl sulfoxide/water (1:1v/v); yield = $\text{product}_{\text{Cmol}}/\text{substrate}_{\text{Cmol}} \times 100\%$) (Yu et al., 2017).

Copper and lead impregnated on biomass had little effect on kinetic parameters that describe the pyrolysis process (Martín-Lara et al., 2016), (Martín-Lara et al., 2018). However, silicon compounds resulted in glassy shelled char (Trubetskaya et al., 2016) when impregnated on rice husk biomass. The particle size and shape of the resulting chars were preserved. However, the reactivity of the char is more influenced by alkali content than the silicon oxides. The presence of K, in the raw biomass increases the reactivity and favoured the heat produced during low-temperature oxidation of the chars. This implies that the self-heating tendency of the char could be reduced by removal of inorganic matter through leaching of the biomass (Fan and Sheng, 2016).

5.2. Other applications of catalyst impregnated solid fuels

5.2.1. Improving char properties and removal of pollutants

Charcoal has been used as a support in several studies and has worked excellently. The addition of Co, Ni, and Fe to charcoal followed pyrolysis under helium enhanced the thermal stability of charcoal for catalytic combustion of volatile organic compounds (Liao et al., 2016). Cobalt gave the best results transforming the amorphous carbon into a graphite-like structure and also served as an active phase in the toluene and ethyl acetate oxidation reaction. Copper impregnated on activated carbon produced from oxytetracycline bacterial residue (Zhou et al., 2015) gave excellent activity for adsorption and oxidation of sulphur dioxide than the activated carbon alone. Similarly, hydrated manganese oxide supported by biochar effectively sequester Pb(II) and Cd(II) than biochar alone

(Wan et al., 2018). Interestingly, the catalyst system could be regenerated and re-used several times. The catalyst activity was attributed to specific inner-sphere complexation of the manganese oxide and pre-enrichment and permeation of Pb(II) and Cd(II) cations into the pore channels of biochar enhanced by the oxygen-containing groups.

Coloured pigments are easily removed when char is impregnated with some metal catalysts. (Park et al., 2018) observed that Fe-impregnated sugarcane biochar could achieve a 99.7% azo dye Orange G (OG) efficiency from solution within 2 hours. The Fe- sugarcane-biochar is more economical, efficient, and recyclable. In similar studies, 96% MB dye removal efficiency was achieved using Ni-impregnated powder activated carbon (Alayan et al., 2018) while RB5 azo dye was completely degraded using grape marc activated carbon/TiO₂ hybrid (Bourahla et al., 2018) catalyst. The degradation species including carboxylic and sulfate groups were observed on the surfaces of the hybrid catalyst using FT-IR and UV-visible analysis.

(Poggi and Singh, 2016) observed that impregnation of Fluid Cracking Catalyst (FCC) on modified biochar achieved a higher thermal degradation capability of acetic acid than FCC alone. With no catalyst, only 5.55% conversion was achieved. With biochar-Ni, a 15.21% conversion was achieved while FCC achieved a 12.41% conversion. The modified bio-char (Biochar-Fe) showed the highest conversion of 40.66%.

5.2.2. Fuel cell performance enhancement

The use of coal char impregnated with iron oxide or calcium oxide for the anode and Y/Zr with La_{0.8}Sr_{0.2}MnO₃ as cathode enhanced the performance of the fuel cell even without a catalyst (Jiao et al., 2015). There was a two-fold increase in peak power density with Fe_mO_n-alkaline metal supported coal char compared to a cell fuelled with pure coal char alone.

6. Conclusions and ideas for future work

This work has discussed solid fuel processing, kinetics and the major advances for minimizing human exposure to carbon monoxide from solid fuel combustion. In summary;

- Solid fuel processing may be achieved by combustion (smoldering or flaming), thermochemical (torrefaction, flash carbonization, pyrolysis, gasification, hydrothermal or liquefaction), and biochemical (anaerobic digestion, fermentation and photobiological processing),
- The general emission products from combustion of solid fuels may be organic and could include more than 15 inorganic elements. Carbon monoxide is released from oxygenated surface functional groups followed by free edge and zig-zag site reactions on PAHs molecules reacting with oxygen until the entire structure is oxidized.
- The methods used to determine kinetic parameters include among others, Friedman, Gupita, KAS, FWO, Starink, Boswel, Coats and Redfern, ASTM methods, Karaosmanoglu and Cif, isothermal methods, iterative methods and Kissinger. There are also methods for determining entropy (ΔS), pre-exponential factor (A), Gibbs free energy (ΔG), and enthalpy change (ΔH).
- Carbon monoxide has been a silent killer since the paleolithic era and has continued to threaten human lives until today.
- Improved cook-stoves offer high heat conservation with less or no smoke at all. They are highly efficient with natural and forced draft air systems and some employ catalyst layers hence ensuring the safety of users from toxic combustion emissions
- Heating systems with two-stage combustion and improved biomass boilers with/without catalysts have also been very crucial in reducing CO exposure. They achieve up to 80% CO conversion and offer multi-solid-fuel usage designs. They have low energy requirements and could be operated as domestic or for district heating systems

- Direct catalytic oxidation of post-combustion pollutants are highly efficient and can achieve 100% CO conversion at very low catalyst loading and ambient temperature. They can be used in conjunction with other systems like cooking and heating. There are various combinations of catalysts available commercially and many more to be designed in the future. Their success is attributed to their high affinity for oxygen and toxic pollutants onto their surfaces.
- The CO sensor systems are various and offer the portability advantage. They have very low detection limits and quick response times. They operate at ambient temperatures enabling usage in various environments. Lately, they can be incorporated with wireless systems allowing CO detection remotely.

Although substantial safety from CO has been provided by the minimization systems described above, there are still several CO related deaths recorded worldwide. All the systems used to minimize CO exposure apply the solid fuel in their original form and during combustion, such solid fuels continue to release CO depending on the prevailing conditions without being impeded. Should there be any malfunction, leakage or pathway for CO, such systems would still expose the user to toxic amounts that may be lethal or cause lifelong injuries.

Altering the composition of the solid fuels by impregnation of chemical catalysts has been used to trigger desired reactions and achieve target products. Using such an approach on solid fuels used for cooking and heating systems would ensure that the solid fuel release minimal amounts of CO. Hence, the amounts of CO released would be rendered none lethal acutely. This coupled with other technologies like sensors/detectors would ensure a safer environment for solid fuel users.

The challenges envisaged would be the costs added to the solid fuel by impregnation of catalysts. Such products if not well marketed may not compete favorably with non-catalyst impregnated products. Furthermore, there are many other considerations from the health and manufacturing perspectives that would also dictate the final pricing. From a health perspective: how much of the catalyst would end up on the food during cooking from the briquette dust, material transfer via contact, and sparks?; would such amounts be within the recommended daily intake as per local and international health standards?; how about the management of ash from the briquettes after cooking – would this ash be environmentally friendly if discharged without caution?. From the manufacturing perspective: making such solid fuels would involve more than just the addition of the catalysts - it may involve accelerants for quick start-up, white ash to show that the briquettes are hot enough and ready to cook food, binders for holding together all the briquette ingredients, press release for easy removal of briquettes from the mould during production, and fillers that add mass to the briquettes.

All the above parameters would add costs to the final product, but the biggest question is how much does someone's life cost? Ultimately, a safe solid fuel is always less expensive than a life. National governments can also assist such projects by waiving off certain taxes on the raw materials used during manufacturing.

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Declaration of interest: None.

Appendix A

Appendix A - Table 1: Semi-conductor sensors

Working/sensing material	CO conc. (ppm)	Response time (s)	Operating temp (°C)	Ref.
Ag-Co ₃ O ₄	5–1500	10-30	50–200	(Molavi and Sheikhi, 2018)
Cobalt oxide nanosheet and carbon nanotube film	200	23	Room temp.	(Dai et al., 2010)
Au-doped CoOOH	1000	40	60 – 110	(Zhuiykov, 2008)
Cobalt oxide (CoOOH)	1000	60	80	(Wu et al., 2006)
Ni and Zn doped SnO ₂	500	5-7	280	(Tang et al., 2017)
Pd on gallia: tin oxide	30	10	300–500	(Kundu et al., 2018)
n-type Zn ₂ SnO ₄	200	Quick response	50	(Chen et al., 2018)
SnO ₂ /CMOS	200	Quick response	375	(Lackner et al., 2017)
Ca-SnO ₂	1	10-12	350	(Ghosh et al., 2014)
Pd ²⁺ /SnO ₂ /CNT	500	2	100	(Hu et al., 2014)
hydroxypropyl cellulose with Pd/SnO ₂	6-18	Quick response	60	(Kim et al., 2013)
V-SnO ₂ and Au/V-SnO ₂	50-1000	5-20	125 – 175	(C. T. Wang et al., 2013)
V-SnO ₂	50-500	14-19	175	(Wang and Chen, 2010)
Si-B-C-N-coated SnO ₂	10 – 120	20-60	350 – 530	(Prasad et al., 2010)
ultrathin SnO ₂ -films	300	20-30 LOD <5ppm	250-400	(Tischner et al., 2008)
Pt/SnO ₂ /i- diamond/p ⁺ -diamond CAIS (Catalyst/Adsorptive Oxide/Insulator/Semiconductor)diode	0.4 – 5.4 torr	24-28	50-500	(Gurbuz et al., 1998)
CaO/Nb ₂ O ₅ /SnO ₂	30-2800	Quick response	100-230	(Tsai et al., 1995)
Si/SnO ₂ , Pd/SnO ₂ , Borosilicate glass/SnO ₂	40-200	Quick response	200-400	(Van Geloven et al., 1991)
SnO ₂ thin film	1-100	Quick response	350	(Windischmann, 2006)
SnO ₂ /La ₂ O ₃ , SnO ₂ /Sb ₂ O ₅ +La ₂ O ₃ and SnO ₂ /Pt+Pd	200	Quick response	100-600	(Malyshev and Pisyakov, 2008)
SnO ₂ thin film	1000	5-20	50-250	(Salehi, 2003)
Ti-doped SnO ₂	300	18-20	150-450	(Z. Wang et al., 2013)
CoxOy/SnO ₂	125-2500	4.9-40.5	Room temp.	(Oleksenko et al., 2013)
Bismuth ferrite (BiFeO ₃)	5-30	25	270-450	(Chakraborty and Pal, 2018)

Prism/Au/ZnO	0.5-100	Quick response	Room temp.	(Paliwal et al., 2017)
SnO ₂ /ZnO	100-1000	120-240	470-510	(Zaikin et al., 2002)
pyridyl-functionalized single-walled carbon nanotubes (F-SWCNTs) and iron porphyrin (Fe(tpp)ClO ₄)	50-200	30-60	Room temp.	(Savagatrup et al., 2017)
ZnO nanoparticles onto 3D graphene Oxide	1000	7	200	(Phuong et al., 2017)
TiO ₂ (Au- TiO ₂) thin films	60-125	20	230-320	(Joy et al., 2006)
TiO ₂ thin films	250	Quick response	550	(Dutta et al., 2005)
Nb-TiO ₂	1000	Quick response	550 – 850	(Anukunprasert et al., 2005)
TiO ₂ , TiO ₂ /La ₂ O ₃ , TiO ₂ /La ₂ O ₃ /CuO and TiO ₂ /CuO	500	Quick response	600	(Savage et al., 2001)
(La _{2-x} A'x Cu _{1-y} B'y O ₄ ; A' = Sr, Ba, Ce; B' = Zr, x, y = 0-0.2)	50-600	Quick response	300-600	(Shimizu et al., 2017)
(x)NiFe ₂ O ₄ (spinel)(1-x)	10-500	60-360	125 – 175	(Maity et al., 2016)
La _{0.8} Pb _{0.2} Fe _{0.8} Co _{0.2} O ₃ (0≤x≤1)	0-50,000	Responsive	Room temp.	(Ho et al., 2011)
NdFeO ₃	10-40	~ 60-360	200-300	(Salker et al., 2005)
LaCoO ₃ -In ₂ O ₃ , LaCoO ₃ -Bi ₂ O ₃ and LaCoO ₃ -PdO	500	10-20	30-160	(Hosoya et al., 2016)
Pt/La _{9.0} Si _{5.8} Mn _{0.2} O _{27-δ}	500	181	100 - 550	(Ding et al., 2015)
LaCoO ₃	500	20-40	130	(Hosoya et al., 2015)
La _{0.87} Co _{1.13} O ₃ -loaded Ce _{0.67} Zr _{0.18} Sn _{0.15} O _{2.0}	500	0.5-1	80-180	(Yang et al., 2019)
Pt/Ce ₄ La ₆ O ₁₇	10,000	10-60	150	(Jelley and Maclay, 1987)
Pd/SiO ₂ /Si	0-5000	180-1020	350-500	(Azad, 2006)
MoO ₃ , MoO ₃ /ZrO ₂ and MoO ₃ /Pd	0-50,000	<120	550 – 950	(Lampe et al., 1995)
BaSnO ₃	20-30	10-60	300-600	(Ghosh et al., 2016)
LuFe ₂ O ₄	0-500	Quick response	23-300	(Guillén-Bonilla et al., 2016)
MgSb ₂ O ₆	100	1.5-32	120-290	(Nikfarjam and Salehifar, 2015)
TiO ₂ /Cu under UV illumination	10-8000	55	Room temp	(R. Kumar et al., 2018)
Cu doped cryptomelane octahedral molecular sieves (Cu-OMS-2)	Up to 10000	<300	50-250	(Nagai et al., 2013)
Au, Pd, and Pt on CoO and Co ₃ O ₄	5-100	8	200-500	(Chen et al., 2012)
Nano-SnO ₂ powder	600	Quick response	25-350	(Kocemba and Rynkowski, 2011)
Pt/SnO ₂				

Pt/SiO ₂ /SiC transistor sensor	0-1250	Quick response	100-400	(Becker et al., 2011)
Co-Ce oxide	3vol%	72	90-125	(Xu et al., 2008)
Ag-doped SnO ₂	100-500	10-17	200	(Petruk and Kravets, 2007)

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