



## Research article

# Evaluation of the Wheeler-Jonas parameters for biogas trace compounds removal with activated carbons



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## ABSTRACT

A practical and feasible solution to reduce the global impacts from fossil fuels is represented by the locally distributed micro-cogeneration systems with high temperature solid oxide fuel cells (SOFC) fed by biogenous fuel coupled in an energy distributed system. One of the main drawback is the low tolerability towards certain fuel impurities, mostly sulfur, chlorine and siloxane compounds. The opportunity to predict the breakthrough time of a gas cleaning section with a high precision level is mandatory to meet SOFC requirements. The reaction kinetic equation called the Wheeler-Jonas equation is adopted to estimate this breakthrough time. Two different commercial activated carbons were studied estimating the breakthrough time varying the operating temperature, the pollutant concentration (single and multiple effects) and the relative humidity. Results showed how relative humidity content affects inversely the removal performance for both sorbents. The Carbox sample, below RH 20% showed interesting results due to its metals content and microstructure. Here, relative humidity promoted the best condition to remove organic vapors from the biogas stream. Multiple contaminant conditions for both sorbent materials decreased the removal performance ( $t_b$ ). This decreasing for the Carbox sample ranged from a minimum of 44% to a maximum of 50% for H<sub>2</sub>S, and 70% for HCl with wet and dry conditions respectively.

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## 1. Introduction

Nowadays the exploitation of some fossil fuels such as diesel or gasoline fuel, natural gas and coal, satisfy the majority of the growing world energy demand. Due to the rapid population growth coupled to the industrial increasing requirements, the emissions from fossil fuels are destined to run out relatively quickly [1]. In addition, their global impacts are extremely harmful, such as the greenhouse effect, ozone layer depletion, acid rains and generalized environment pollution (PM<sub>2.5/10</sub>, NO<sub>x</sub>, SO<sub>x</sub> and CO). In order to reduce or to narrow down this problem, the renewable fuels employment in energy generation systems turns out to be necessary. A practical and feasible solution to reduce the global impacts from fossil fuels is represented by the locally distributed micro-cogeneration systems, with high temperature solid oxide fuel cells fed by biogenous fuel coupled in an energy distributed system [2,3]. These systems couple the high fuel conversion values of SOFCs, due to the electrochemical reactions instead of fuel combustion [4] and their remarkable fuel flexibility [5]. Biogas from a biomass source is a viable alternative to fossil fuels in terms of energy generation performance and pollutant emissions [6]. This fuel is appropriate to be used in SOFC

systems as reported in previous studies [7–10]. Biogas is produced from organic matter digestion with methane and carbon dioxide as principal gas mixture elements. Next to these main constituents a wide and variable range of trace compounds are contained in biogas [11]. One of the main drawback for SOFCs fed by biogas is the low tolerability towards certain fuel impurities, mostly sulfur, chlorine and siloxane compounds that may decrease cell efficiency and degrade the fuel cell [12–17]. In order to achieve the biogas quality requirements for SOFC applications, there are numerous techniques available which can be classified as biological, physical and chemical processes [18]. Biological processes are commonly used to reduce the pollutant emissions contained in the biogas. These treatments have significant economic advantages over other pollution control technologies. Biofiltration is a process by which contaminated gases pass through the biofilter and pollutants are transported into the biofilm, where they are utilized by microbes as a carbon and energy source [19,20]. It is a process adopted to roughing the starting pollutant concentration from thousands of ppm(v) to hundreds of ppm(v). Physicochemical processes can be classified as absorption (caustic washing), reactive (chemical oxidation) and adsorption techniques (iron and metal adsorbents, activated carbons). They are mainly appropriate with low gas flow rates and low H<sub>2</sub>S concentrations (hundreds of ppm(v)). Adsorption is the only technique which can reduce the concentration of contaminants to the stringent extent of fuel cell's specifications [21].

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## Nomenclature

$A_{\text{tot}}$	total water that can be adsorbed ( $g_{\text{water}}/g_{\text{coal}}$ ),
$B$	structural carbon constant ( $K^{-2}$ ),
$C_0$	initial concentration ( $g/cm^3$ ),
$C_s$	contaminant concentration at the saturation pressure ( $g/cm^3$ ),
$C_w$	water concentration in the gas stream ( $g/cm^3$ ),
$C_x$	breakthrough concentration ( $g/cm^3$ ),
$D$	unit of dipole moment (debye),
$d_b$	bed depth (cm),
$d_l$	liquid density of the organic vapor ( $g/cm^3$ ),
$d_p$	carbon particle average diameter (cm),
$H_2S$	hydrogen sulfide,
$HCl$	hydrogen chloride,
$k_v$	overall adsorption rate coefficient ( $\text{min}^{-1}$ ),
$k_{v,\text{wet}}$	adsorption global coefficient with humid condition ( $\text{min}^{-1}$ ),
$m$ and $k$	Langmuir constant,
$MW$	molecular weight (g/mol),
$p_1^0$	$p_2^0$ model parameter,
$PDMS$	polydimethylsiloxane membrane,
$PID$	Proportional Integrative and Derivative controller,
$P_s$	saturation pressure of the pollutant (mbar),
$P_w$	saturation pressure of the water (mbar),
$Q$	volumetric flow rate ( $\text{cm}^3/\text{min}$ ),
$RH$	Relative Humidity,
$SOFC$	Solid Oxide Fuel Cell,
$T$	operating temperature (K),
$t_b$	breakthrough time (min),
$TPV$	total pore volume,
$v_l$	linear velocity through the bed (cm/s),
$VOCS$	Volatile Organic Compounds,
$W$	sorbent weight (g),
$W_0$	micropore volume ( $\text{cm}^3/g_{\text{coal}}$ ),
$W_e$	equilibrium adsorption capacity ( $g_{\text{pollutant}}/g_{\text{coal}}$ ),
$W_{e,1-2}$	equilibrium adsorption coefficient of the component 1 or 2 ( $g_{\text{pollutant}}/g_{\text{coal}}$ ),
$W_{e,\text{tot}}$	equilibrium adsorption capacity of the binary mixture ( $g_{\text{pollutant}}/g_{\text{coal}}$ ),
$W_{\text{pre}}$	volume occupied by the pre-adsorbed water from the carbon ( $\text{cm}^3/g_{\text{coal}}$ ),
$y_1$	molar fraction of the component,
$\Delta W_{\text{gas}}$	volume occupied from the gas flow ( $\text{cm}^3/g_{\text{coal}}$ ),
$\Delta W_s$	volume of water adsorbed and replaced from the pollutant ( $\text{cm}^3/g_{\text{coal}}$ ),
$\beta$	organic vapor coefficient,
$\rho_b$	bulk density of the carbon bed ( $g/cm^3$ )

VOCS removal from activated carbon adsorption represents a valid and economic solution [22]. The main properties of activated carbon filters for the effective removal of VOCS are high porosity, high superficial area, high volume, pore distribution [23] and treatment with metal ions. Physisorption and chemisorption are the two underlying physical phenomena of adsorption. The first one deploys weak Van der Waals forces, whereas the second one involves the stronger covalent and ionic bonds. Van der Waals forces require polar or polarizable compounds. Typically, compounds with no dipole moment (0 D) are classified as non-polar, and else as polar.

Activated carbons are usually impregnated with metal ions because of the higher chemical affinity for organic compounds. Typical metals deployed are iron, copper, silver and chromium. These substances are able to promote the bond between carbon and polar molecules, such as sulfur, aromatic and carbonyl compounds [24–27]. In order to work

with a SOFC generator fed by a biogenous fuel, the possibility to predict the breakthrough time with a high precision level is mandatory, especially with measurable variables. The reaction kinetic equation, called the Wheeler-Jonas equation is adopted to estimate the breakthrough times of filters against organic vapors [28–30]. Several research works were accomplished in order to investigate the predictability of different sorbent materials against different organic vapors [29–32]. Some organic vapors were investigated, for example: chlorine compounds, hydrocarbons (heptane, cyclohexane), ethanol and acetone. This equation has been adopted to extrapolate single and multiple laboratory results by simply varying the independent variables of the equation, for example the weight of the carbon bed, the carbon material adopted, the initial concentration and the volumetric flow rate [28,33,34]. The Wheeler-Jonas equation is simple, fast with low computational cost to be used for the comparison of model and experimental results.

This equation is the result of a semi-empirical approach based on solely measurable and readily available macroscopic parameters. It is originally based on a continuity equation of the mass balance between the vapor entering the carbon bed and the sum of the amount adsorbed by this bed plus the amount penetrating through it.

This model requires several assumptions:

- the flow pattern is a perfect plug flow;
- only physisorption in the micropores is considered;
- the kinetics of the reaction are of a pseudo-first order.

A perfect plug flow implies the absence of any axial dispersion and/or wall effects. In a first approximation this assumption, can be adopted because the WJ equation is not focused on individual channels study. Only physisorption in the micropores excludes any chemisorption and/or interactions with other gases or vapors present on the carbon. In the Wheeler-Jonas equation it is not crucial which step in the adsorption process is rate controlling, as long as the rate constant is of a first order with respect to the number of gas molecules. This is only true in the first, convex part of the sigmoidal breakthrough curve, i.e. for values of  $C_{\text{out}}/C_{\text{in}} < 4\%$ . As the breakthrough criterion used in this study is 0.1–0.4%, this equation is valid. In summary, there is a strong possibility that the Wheeler-Jonas equation can be applied to these experiments. Lack of the literature studies about the estimation of the breakthrough time for sulfur and chlorine compounds needs a specific investigation. In this work, the Wheeler-Jonas equation was adopted to investigate and to estimate the breakthrough time, the adsorption capacity and the overall adsorption rate capacity for two different commercial activated carbons. The operating conditions considered were temperature, the pollutant concentration (single and multiple effect) and the relative humidity.

## 2. Materials and methods

In this section are reported the description of activated carbon materials, the experimental set-up, and the mathematical model used. The Wheeler-Jonas equation is the result of a semi-empirical approach based on solely measurable and readily available macroscopic parameters. It is originally based on the continuity equation of the mass balance. To consider the effect of humidity it was considered the work made by Lodewyckx and Vansant (1999) [33]. Here, the approach followed by Grant and Manes was developed with the static adsorption coefficient determination in the presence of a constant relative humidity. The adsorption was based on the Polanyi model of pore filling [35,36].

The interaction between water vapor and contaminant molecules in porous media includes mainly three pathways, [37]:

- (1) the competition for active sites between water vapor and contaminant molecules at the exposed pore surface;
- (2) the capillary condensation of water vapor in micropores, which would reduce the amount of exposed surface area for pollutant molecules;

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