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High-pressure C-H-O diagrams: Fuel composition, carbon deposition, and open circuit voltage of pressurized SOFCs

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ABSTRACT

Solid oxide fuel cells (SOFCs) can operate by using various fuel species. Pressurized SOFCs with gas/steam turbine(s) may achieve higher power generation efficiency as hybrid or triple-combined power generation systems. In this study, fuel gas composition is systematically investigated by thermochemical equilibrium calculations on the anode side of SOFCs, pressurized up to 30 bar over a wide temperature range, up to 1000 °C. Since conventional hydrogen-containing fuel gas consists mainly of carbon, hydrogen, and oxygen, high-pressure C-H-O equilibrium diagrams are numerically obtained. It is revealed that the carbon deposition region contracts in the hydrogen-rich area and expands in the oxygen-rich area with increasing total pressure. The molar fraction of each gas component, described in such C-H-O diagrams, also depends on the total pressure. The theoretical open circuit voltage (OCV) increases by pressurization. The effect of nitrogen in high-pressure SOFC fuels is also considered, which is important especially for air-blown coal gas. The minimum amount of H₂O, O₂, and CO₂ required to prevent carbon deposition in steam reforming, partial oxidation, and CO₂ (dry) reforming, respectively, is also derived up to 30 bar. The high-pressure C-H-O diagrams are also applicable to various high-temperature/high-pressure energy systems such as solid oxide electrolyzer cells (SOECs) and reversible fuel cells.

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Introduction

Various chemical species can be used as fuels in SOFCs for power generation [1–4], including hydrogen, methane, alcohol [5], coal gas [6–8], and biogas [9,10]. However, using different fuel species can cause technological issues such as cell performance degradation and carbon deposition [11,12]. The power generation and efficiency of SOFCs combined with a micro gas turbine (MGT) have been previously studied *e.g.* for methane- and methanol-based fuels [13]. The changes in cell performance caused by using various fuels is related to the cell voltage [12,14], the overvoltages, and thus the exchange current density [15,16]. Furthermore, fuels are often reformed by adding steam and/or ambient air (*i.e.*, oxygen) in order to prevent carbon deposition [2,17–20]. The effect of this addition depends on the fuel gas composition, so that it is necessary to determine the appropriate reforming conditions [18,21].

SOFCs usually operate at high temperatures, from around 600 to 1000 °C. They can be applied in extremely efficient triple-combined-cycle large-scale power generation systems [7,8,22], incorporating a gas turbine and a steam turbine, operated at high pressure (around 20 bar). The fuel for such triple-combined-cycle systems is generally natural gas (consisting mainly of CH₄) and coal gas, after air-blown or oxygen-blown gasification [23–25]. Both of these gasification methods are still under development in parallel, since they each have their own advantages and disadvantages [23–27]. Fig. 1 shows

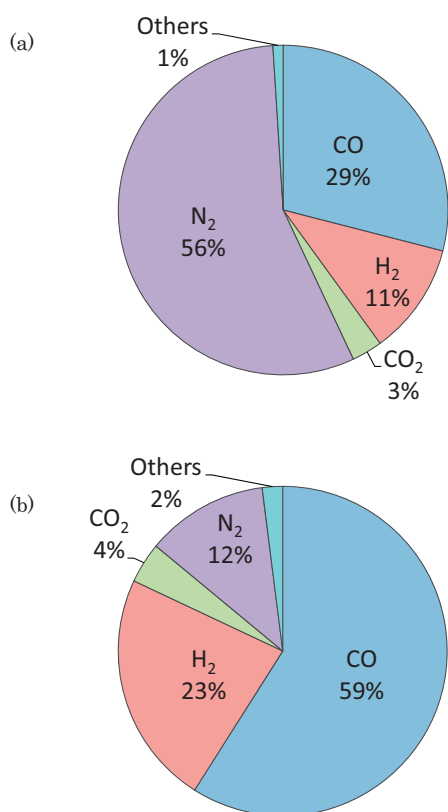


Fig. 1 – Typical fuel composition of (a) air-blown coal gas, and (b) oxygen-blown coal gas, by volume ratio [27].

the typical compositions of air-blown and oxygen-blown coal gas, by volume ratio [27]. Whilst coal gas may also contain nitrogen, the fuels mainly consist of carbon, hydrogen, and oxygen, and the specific composition determines SOFC performance. Fig. 2 shows the positions of these different SOFC fuels in C-H-O diagrams. The positions of natural gas (CH₄) and coal gas are also shown, and in addition their positions after steam reforming with typical steam-to-carbon ratios (*S/C*) of 2.0/3.0 for natural gas and 1.0/1.5 for coal gas [28,29]. As the electrochemical reactions proceed, the fuel composition shifts towards the oxidizing direction, as depicted by red arrows in Fig. 2.

According to previous studies in the literature, pressurized SOFCs have various advantages, including higher cell voltage, which is associated with lower electrode overvoltage [8,30], and lower costs of the produced electricity [31]. In particular, SOFC performance has been successfully demonstrated to increase by pressurization up to 15 atm, as shown by Virkar, Fung, and Singhal [30]. Riensche *et al.* revealed that pressurization up to 10 bar led to a 100 mV increase in cell voltage [32]. Such changes induced by pressurization also depend on operation temperature [33]. Fuel composition at lower temperatures should also be taken into account, especially near the fuel inlet and near fuel cleaning systems, or within specific low-temperature SOFC systems [34,35]. Willich *et al.* investigated the pressure dependency of the open circuit voltage (OCV) using three different types of fuels; a H₂/N₂ mixture and two different types of reformed gas, between 1 and 8 bar [36]. Wendel *et al.* analyzed the operating conditions of reversible SOFC/SOEC systems using a thermodynamic approach, considering the effect of pressure on fuel composition from 1 to 25 atm [37]. However, fundamental characteristics such as carbon deposition, equilibrium gas composition, and theoretical OCV at high pressure have not yet been systematically clarified in the literature.

Since most fuels in energy systems contain elemental carbon, hydrogen, and oxygen, the C-H-O diagrams [19] have been widely used as general and practical guidelines to take into consideration in the design of energy systems. Thermochemical equilibria have been taken into account in evaluating various energy systems, for example: SOFC systems coupled with fuel processing for power-to-fuels by Hansen [38]; with anode off-gas recycling by Lee *et al.* [39]; with biogas treatment by Mermelstein *et al.* [40]; hybrid SOFC-Stirling engine plants by Rokni [41]; single chamber SOFCs by Buegler *et al.* [42]; tubular SOFCs by Zhu *et al.* [43]; reversible SOFCs by Wendel *et al.* [37,44]; regenerative SOFCs for energy storage by Kazempoor *et al.* [45]; combined SOFC and PEFC systems by Rabbani *et al.* [46]; and electrolysis of steam and CO₂ by Ebbesen *et al.* [47,48]. The C-H-O diagrams are helpful in considering fuel compositions, as mentioned *e.g.* in review papers by Gür [49,50] and in a theoretical paper by Jaworski *et al.* [51]. Equilibria in fuel gases have been considered for various fuels including methane [18,43,50,52], alcohols [53–55], biogas [40,56,57], dry CO [58], hydrocarbon fuels [18,59–61], solid carbon fuel [49], and various impurity-containing fuels [12,62–64], where most of these studies referred the C-H-O diagrams.

It should be mentioned, however, that both thermodynamics and kinetics can dominate fuel compositions, as is

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