



# Removal of highly concentrated toluene from flue gas by an anode-supported solid oxide fuel cell reactor to generate electricity



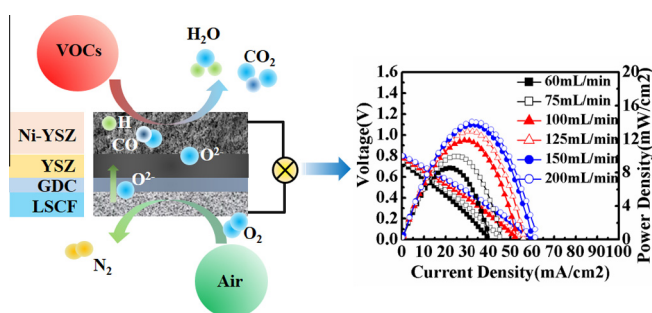
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## HIGHLIGHTS

- SOFC to remove highly concentrated toluene.
- Excellent power performance is obtained at an intermediate temperate of 650 °C.
- The power performance increases when flow rate and toluene concentration increase.
- The generating efficiency of the cell with toluene as the fuel is evaluated.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Toluene, a typical volatile organic compounds (VOCs), is raising growing concern due to its harm to human health and atmospheric environment. However, it also can become a fuel for that C–C and C–H bonds contain considerable chemical energy. In here, we utilize a solid oxide fuel cell (SOFC) reactor to degrade toluene as an environment pollutant and to take full advantages of its chemical energy simultaneously to generate electricity. The scanning electron microscopy (SEM) results show a uniform three-layer structure of the cell and the porous morphology of the anode. Energy dispersive spectroscopy (EDS) illustrates the atomic ratio of the fuel cell and reveals a diffusion layer of the cell between the cathode and the electrolyte with an atomic ratio of Ce:Gd:O = 5:26:69. In the reactor, the toluene can be removed from a low temperature of 600 °C. It can be totally removed under 650 °C and obtain a power density of 14 mW/cm<sup>2</sup>. The removal efficiency can reach 94.19% under high toluene concentration of  $1.874 \times 10^5$  ppmv and a temperature of 650 °C and obtain a power density of 14 mW/cm<sup>2</sup>. However, it decreases as the flow rate increases to 200 mL/min. With the initial concentrations varying from  $1.243 \times 10^5$  ppmv to  $5.472 \times 10^5$  ppmv, the cell efficiency declines from 29.93%, 20.89%, 19.13% and 19.61% to 16.06%, 5.030%, 8.297% and 7.874% as the flow rate increases from 50 mL/min to 200 mL/min. The impedance spectra are analyzed for the mechanism of cell performance. The economic analysis shows that it can save energy not only in that it generates electricity while removing toluene but also in that the heat it releases can well help preheat the cathode air.

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## 1. Introduction

Volatile organic compounds (VOCs) are defined as a group of carbon containing compounds that readily evaporate at room temperature (USEPA, 2012). They are a group of air pollutants that

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## Nomenclature

VOC	volatile organic compound	$R_e$	electrode polarization resistances, $\Omega/\text{cm}^2$
SOFC	solid oxide fuel cell	$P^*$	saturated pressure at a certain pressure, mmHg
SEM	scanning electron microscope	$p^b$	standard atmospheric pressure, 101.325 kPa
ppmv	parts per million by volume	$t$	temperature, $^\circ\text{C}$
EIS	electrochemical impedance spectroscopy	$\eta$	generating efficiency, %
TPB	Three-phase boundary	$\Delta H_c$	combustion enthalpy of toluene, $\text{kJ mol}^{-1}$
$C_{\text{Toluene,in}}$	inlet toluene concentration, $\text{mol L}^{-1}$	$H_{\text{reac}}$	formation enthalpy of the fuels, $\text{kJ mol}^{-1}$
$C_{\text{Toluene,out}}$	outlet toluene concentration, $\text{mol L}^{-1}$	$H_{\text{prod}}$	formation enthalpy of the products, $\text{kJ mol}^{-1}$
$H_{\text{fuel}}$	enthalpy change of the fuel, $\text{kJ mol}^{-1}$	$\Delta h_c$	calorific value of toluene, $\text{kJ mol}^{-1}$
$R_0$	ohmic loss, $\Omega/\text{cm}^2$		

are widely present in both indoor and outdoor air because they come from natural emissions and possibly from anthropogenic activities, including motor vehicles exhaust and solvent utilization [1] and are also contained in biogas from the anaerobic digestion of organic waste, as reported by Papurello [2]. VOCs including benzene and chloroform are reported to be carcinogenic, mutagenic, and teratogenic to humans [3]. Due to stringent regulations on VOCs emissions, removal of VOCs from flue gas streams became an important issue for engineers and researchers. Toluene is a typical VOC as it is emitted in high levels and is difficult to abate [4]. It is reported that inhalation of toluene brings about nervous system disorder, such as reduction in thinking, memory and muscular abilities, as well as some level of loss in both hearing and color vision [3]. Toluene has found its place on the priority pollutant list of the USEPA due to its high concentration in gasoline.

Researches have been conducted on toluene removal strategies, which are generally classified as either destructive [5,6] or recuperative [7,8]. The most common destructive technology catalytic oxidation as Qian [9] has conducted to apply nanoscale  $\text{CaO}_2$  to the removal of toluene contaminants has demonstrated a superior performance in the degradation of toluene, which could be eliminated in 3 d at pH 6. Zubair Ahmed [10] adopts yeast strain *Candida tropicalis* enhanced by granular activated carbon to remove toluene, obtaining a maximum elimination capacity with a toluene loading of 291  $\text{g}/\text{m}^3/\text{h}$ . These destructive technologies prove to be effective but suit better for lower concentration, and furthermore they cannot make best use of the chemical energy of toluene. Other recuperative [8,11] technologies use methods such as adsorption or condensation to transfer the toluene from one phase to another rather than to eliminate them, but these technologies are not economical [12]. Therefore, it is of great importance to search for a technology that will not only remove toluene but also take advantages of its chemical energy to save energy.

Solid oxide fuel cell (SOFC) is a promising all-solid-state energy conversion device [13] and has attracted considerable attention in recent years for their flexibility in fuel selection. Oxygen anions are transported through a membrane [14], which is different from that used in other fuel cells such as proton exchange membrane fuel cell (PEMFC) and thus it requires high operating temperatures. Although considerable researches have been done on SOFC, most researches on SOFC fuels have focused on hydrogen [15–18] or alkanes with less than five carbons [15,19,20] and at high temperatures above 700  $^\circ\text{C}$  [21–25] and there lack of studies on complex organic like benzene series. Thus application of SOFC to removal of toluene will not only help solve the problem of degradation of benzene series which take up a remarkable place in the most common VOCs pollutants [26], but also make best use of their chemical energy. The main objective of this study is to investigate the possibility of removing toluene by feeding it as the anode fuel of the SOFC reactor and estimate the power efficiency of this application.

## 2. Experimental section

### 2.1. Chemical and materials

The purity of toluene (Shanghai Lingfeng Chemical Reagent Co., Ltd) is 99.9%. The SOFC is supplied by Shanghai Institute of Ceramics, Chinese Academy of Science. The toluene generator is composed of a water bath (Jintan Chengxi Zhengrong Experimental Instruments Plant) and a homemade glass columnar container (6 cm in diameter and 20 cm in height). The electric heating furnace and the temperature controller instrument are purchased from Shanghai Yuanci Environmental Technology Co., Ltd.

### 2.2. Experimental setup and reactor

As is shown in Fig. 1, the experiment is carried out in a single SOFC setup. The three-layer solid oxide fuel cell is 2.5 cm in diameter and the cathode layer is 1.0 cm in diameter. A Platinum mesh (80 meshes) and silver rod (0.5 mm in diameter) as current collectors are attached to one side of a ceramic tube with silver paste and then a heat treatment of 120  $^\circ\text{C}$  for 30 min. Then the anode side of the cell is sealed to the Platinum mesh and more silver paste is employed to fill the gap before another heat treatment of 120  $^\circ\text{C}$  for 60 min to finish the solidification. The cathode side is exposed to stagnant air.

A temperature controller is used to keep the reactor temperature stable. When setting the furnace to an aimed temperature and time, it will heat up to the aimed temperature with a heating rate of 10  $^\circ\text{C}/\text{min}$ , and remain constant for a preset time. The cooling rate is set as 1  $^\circ\text{C}/\text{min}$  so as to avoid dehiscence of the cell after the experiment. An appropriate flow of nitrogen is pumped in through the toluene solution, the temperature of which is kept constant in a water bath so that a mixture of nitrogen and toluene vapor with a stable concentration could be introduced to the anode surface. Static air served as an oxidant in the cathode chamber. And a heater band is employed to keep a gaseous state of the wastes. The waste gases are directly diluted with high purity nitrogen and then introduced to the total hydrocarbon analyzer to be detected.

The current–voltage curves and electrochemical impedance spectroscopy (EIS) are obtained using an electrochemical workstation (Chenhua, China) with an amplitude of 0.01 V over the frequency range of 0.1 Hz to 1000 kHz under open circuit voltage, and the concentrations of the toluene during the reaction are recorded by a total hydrocarbon analyzer (THC, Thermo Fisher Scientific, USA).

### 2.3. Experimental procedure

The temperature controller is set to the reaction temperature, and nitrogen with a flow rate of 50 mL/min is introduced into

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