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A catalytic combustion based flexible thin-film thermoelectric power generator with fuels of methanol and ethanol

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ABSTRACT

Flexible thin-film micro combustion-thermoelectric coupled (tf-MTC) device which composed of annular thin-film thermoelectric generator (ATTEG) and catalytic combustor based on Pt nanoparticles shows the potential to provide power for the portable electronic devices. The tf-MTC with in suit heat source directly attached on the ATTEG shows a great reduction of heat loss without the mechanical contact between TEG and heat source. The device shows higer power output for the higher quality temperature input of the catalytic combustion when compared with the flexible thermoelectric device without in situ heat source, and is suitable for various application scenarios for the flexibility and small size. Methanol and ethanol were chosen as the fuel of catalytic combustion for the study of the output performance of the coupled device to find out the appropriate application scenarios. This paper mainly explored the difference of output performances of the coupled device with methanol and ethanol. With the consideration of the characteristics of methanol and ethanol during catalytic combustion based on Pt nanoparticles. Multicycle test with on and off state of the reactant mixture was applied to methanol, and single cycle test with long time use was applied to both methanol and ethanol. The results of multicycle test showed similar repetitive typical temperature and output performance of the tf-MTC device. An obvious increase of output performance was obtained during single cycle test of the tf-MTC device with ethanol when compared with methanol. The output voltage and the maximum output power reached 0.201 V and 2.38 μW, respectively. The results indicates both methanol and ethanol are suitable for the tf-MTC device as power generator, and different output performances paly a guiding role of the choice of the fuel for different application scenarios.

Keywords: the coupled device; catalytic combustion; methanol; ethanol

NONMENCLATURE

Abbreviations	
ATTEG	Annular thin-film thermoelectric generator
C-C	Catalyst combustor
TE	Thermoelectric
TEG	Thermoelectric generator
tf-MTC	Flexible thin-film micro combustion-thermoelectric coupled
Symbols	
P_{max}	The maximum output power
R	Internal resistance
V	the output voltage
α	Seebeck coefficient
ΔT	Temperature difference

1. INTRODUCTION

Microreactors show great potential for power generation[1]. Thermoelectric generator (TEG) can directly convert thermal power into electrical power for the Seebeck effect of thermoelectric (TE) materials, which shows the ability of generate voltage when the TE leg is exposed to a temperature gradient[2,3].

In order to improve the output performance of the power generator which composed of TEG and heat source. Both the heat source quality and thermal management[4] which aims at the reduction of heat loss of the whole system are taken into consideration. Hydrocarbon fuels are suitable to act as heat sources of the power generation system for their high energy density[5]. The catalytic combustion, which can provide sustainable temperature input, is commonly used as the heat source of power generation system with TEG[6]. The Pt nanoparticles shows great ability on the fuel percent conversion and ignition behavior. Methanol was chosen as the reactant fuel for its property of self-ignition at room temperature which proved by McNally et al[7], who explored catalytic combustion with commercial hydrocarbon fuels include methane, propane, butane and ethanol, only methanol showed the property of room-temperature ignition.

Though methanol can be accessed easily from sustainable source, the toxicity and environmental hazards make it be abandoned by many major fuel consumers. Ethanol is acceptable for its less toxic than methanol, broadly applicable and more user friendly. It was taken into consideration as the reactant fuel of the catalytic combustion based on Pt nanoparticles. The combination of fuel, catalyst and TEG accepts a lot of focus to optimize output performance of the coupled device [7-9].

In this paper, a coupled device which composed of ATTEG and catalytic combustor based on Pt nanoparticles was proposed for a higher output performance. Both methanol and ethanol were taken into consideration as the fuel of the catalytic combustion for their unique characteristics. To make a choice of the fuel for different application scenarios with the best output performance, the working condition and the corresponding performance output characteristics of the device were both taken into consideration. Single cycle test was applied to the tf-MTC device with methanol and ethanol, and multicycle test was only applied to methanol for the self-ignition property at room-temperature.

2. EXPERIMENT METHOD

2.1 The fabrication of tf-MTC device

The tf-MTC device which composed of the ATTEG and catalytic combustor based on Pt nanoparticles was proposed, as shown in Fig. 1, to avoid the heat loss between the heat source and TE generator for the mechanical connection.

The ATTEG is fabricated by RF magnetron sputtering (Kurt J.Lesker PVD 75) with stainless steel masks, the fabrication method is the same as our previous study[10]. The substrate used in ATTEG is polyimide substrate (DuPont Trading (Shanghai) Co.,Ltd, 125 μm in

Fig. 1. The photograph of the ATTEG and the tf-MTC device

thickness, 30mm\times30mm in area). The thickness of Ti-layer, Cu-layer, Ni-layer of the electrode and p-type BST layer and n-type BTS layer of the TE legs is 50 nm, 200 nm, 100 nm, 1000 nm, 1000 nm, respectively. The catalyst combustor is composed of high thermal conductivity paste (OMEGATHERM"201") and Pt-black nanoparticles (Alfa Aesar, CAS:12755), where the Pt-black nanoparticles are evenly covered on the paste which acts as the adhensive layer between the substrate and the catalyzer. The radius of the catalyzer is 4.5mm which is shorter than the central area of the ATTEG with radius of 5 mm to ensure the independence of ATTEG and catalytic combustor.

2.2 Reactor design and measure method

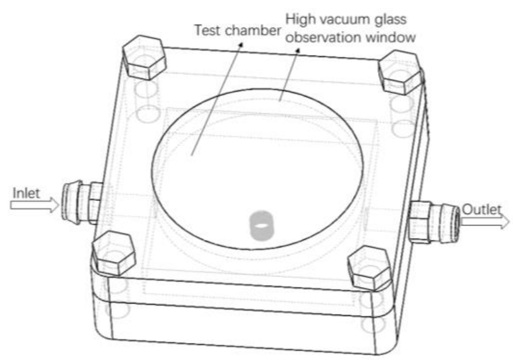

Fig. 2. The diagram of the test chmber

With the consideration of different characteristic of methanol and ethanol, no additional ignition equipment was required for methanol for its self-ignition at room temperature, and an electric soldering iron with accurate temperature control was chosen as the ignition equipment of ethanol-air. A self-made stainless steel test chambers (55mm\times55mm\times10mm) was designed, as shown in Fig. 2. The top area of the test chamber is covered by a high vacuum glass observation window to observe the tf-MTC device at the test chamber. There is a screw hole at the bottom of the test chamber to place the additional ignition equipment for the ethanol-air, when the catalytic combustion is stable, the hole is blocked by a screw to ensure the consistency of inlet and outlet conditions with methanol-air which was tested with the hole closed. Gas washing cylinder was used to bubble the fuel to form the methanol-air or ethanol-air for the catalytic combustion, the flow rate was controlled by flowmeter (ALICAT). Digit multimeter truevolt (34465A, KEYSIGHT) and multiplex temperature recorder (Shenzhen Shenhwa Technology Co.,Ltd.) with K-type thermocouples were applied to the test of typical temperature and output performance of the tf-MTC device.

3. RESULTS

3.1 The study of methanol for tf-MTC device

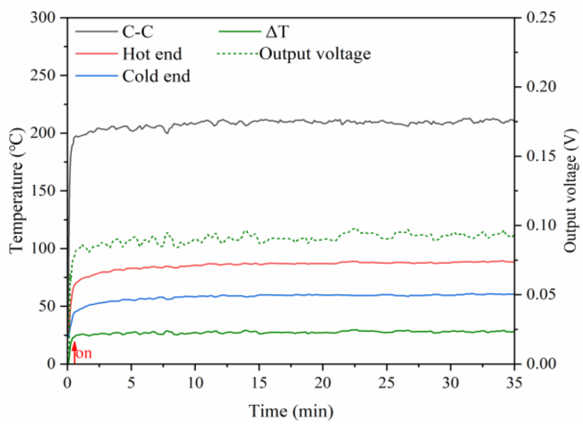

Fig. 3. The typical temperature and output voltage of single cycle test of the tf-MTC device based on methanol

The tf-MTC device was tested with a flow rate of 500 mL/min, the results of typical temperatures and the corresponding output voltage vs. time were shown in Fig. 3. The typical temperature of the catalyst combustor (C-C) can intuitively reflect the situation of catalytic combustion. Note that the temperature of C-C rapidly increase before the stable state of catalytic combustion as the methanol-air is turned on, which meet a great agreement with self-ignition at room-temperature based on Pt nanoparticles. The typical temperature change trend of hot end and cold end are both similar to the typical temperature of C-C, for the reason of the temperature input of the hot end and cold end both come from the heat transfer of substrate and TE legs to the hot source of C-C. The response time of cold end is more than hot end for its far distance from C-C, which results in a lower slope of curve at cold end than hot end. The curve of output voltage is similar to the curve of temperature difference, where the output voltage mainly depends on the temperature difference ($V = \alpha\Delta T$, where V, α and ΔT is the output voltage, Seebeck coefficient and temperature difference, respectively).

The mean value of the typical temperature is 208.5 ℃, 86.3 ℃, 58.7 ℃, 27.6 ℃ for C-C, hot end, cold end and the corresponding temperature difference. The mean output voltage reaches 0.091V. During the stable state of the catalytic combustion, the mean resistance of the tf-MTC device during stable state of catalytic combustion is 4.2 kΩ, which results in a maximum power output of 0.49 μW, which was calculated using $P_{max} = V^2/(4 \times R)$, where P_{max}, V and R represents the maximum output power, output voltage and internal resistance, respectively.

Multicycle test was proposed with the consideration of saving fuel in some application scenario where the device need to work at intervals and the characteristic of the methanol. The methanol-air flow was turned on for 180 seconds and then off for 120 seconds as a cycle.

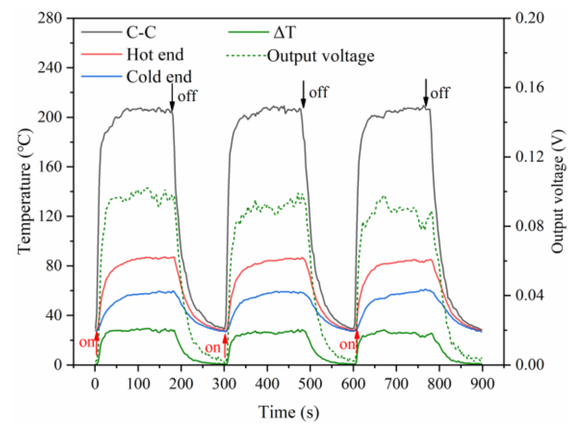

Fig. 4. The typical temperature and output voltage of multicycle test of the tf-MTC device based on methanol

The results are shown in Fig. 4. Note that there is a rapidly response of the typical temperature at C-C with the on and off of the methanol air. Similar trend shows in other typical temperature and the corresponding output voltage.

Table 1. The mean values of typical temperature and output voltage during stable state of catalytic combustion at each cycle

	C-C (℃)	Hot end (℃)	Cold end (℃)	ΔT (℃)	Voltage (V)
Cycle 1	206.0	85.7	57.5	28.2	0.098
Cycle 2	206.0	84.2	57.9	26.3	0.091
Cycle 3	205.6	83.3	57.7	25.5	0.089

The mean value of the typical temperature of C-C, hot end, cold end, the corresponding temperature difference, and the output voltage during the stable state of catalytic combustion at each cycle are illustrated in Table 1.

The mean value of parameters at each cycle are almost the same, the repeat close results show the output performance stability of the tf-MTC device with methanol-air at multicycle working condition. The stability is benefit to provide stable power output for portable device as power source. The typical temperature at C-C of each cycle is slightly lower than the single cycle with long time use, which can be attributed to the limited time of the relative stable state of catalytic combustion.

3.2 The study of ethanol for tf-MTC device

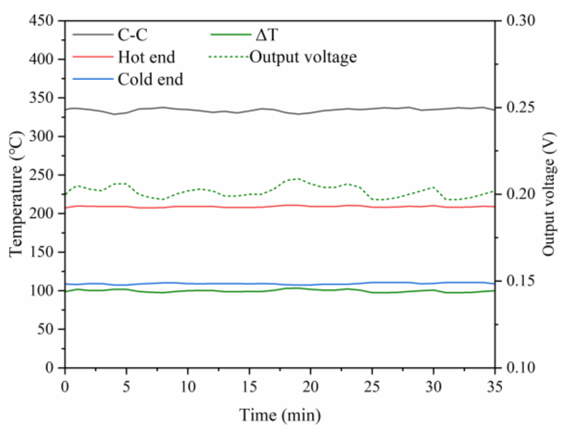

Fig. 5. The typical temperature and output voltage of single cycle test of the tf-MTC device based on ethanol

The same flow rate of 500 mL/min was applied to the test of the output performance of tf-MTC device with fuel of ethanol. The temperature of 380 ℃ of the ignition equipment shows stable performance to ignite the catalytic combustion without preheating the ethanol-air.

Fig. 5. illustrates the typical temperatures and the output voltage of the tf-MTC device during the steady stable of catalytic combustion. The mean parameters of catalytic combustor, hot end, cold end, the corresponding temperature difference and output voltage is 334.3 ℃, 209.0 ℃, 109.2 ℃, 99.8 ℃ and 0.202 V, respectively. There are small fluctuations of the typical temperature at catalytic combustor as well as the other parameters, which indicates the stability of the catalytic combustion and the output performance of the tf-MTC device. During steady catalytic combustion, the value of internal resistance is nearly 4.3KΩ. The maximum output power which corresponding to the mean value of output voltage during stable state of catalytic combustion can reach 2.38μW.

3.3 Discussion

Both methanol and ethanol were taken into consideration as the reactant fuels of the catalytic combustion with Pt nanoparticles, and the typical temperature and output performance of tf-MTC device were measured. Table 2 illustrates the mean parameters during stable state of catalytic combustion with long time use for a clear comparison of the performance of the tf-MTC device with ethanol and methanol.

Note that the temperature difference of ethanol is obviously higher than methanol, which results in a high output voltage and the corresponding maximum output power. The output voltage of the tf-MTC device with ethanol reach as high as 0.202 V which can supply power for some low power sensor without any booster device. Though the output performance of the tf-MTC device is lower than ethanol. the self-ignition at room temperature characteristic of methanol makes it portable and easy to miniaturization for the whole system of power generation. There are specific advantages and disadvantages for both methanol and ethanol which makes it possible to choose proper reactant fuel according to the working condition.

Table 2. The comparison of the output performance of tf-MTC device with ethanol and methanol.

	ΔT (℃)	output voltage (V)	the maximum output power (μW)
Ethanol	99.8	0.202	2.38
Methanol	27.6	0.091	0.49

4. CONCLUSION

Flexible tf-MTC device with in situ heat source was proposed in this paper and two fuels were taken into consideration for their special characteristics to paly a guide role of the choice of the fuel at different scenarios. Pt nanoparticles was chosen as catalyzer for its high percent conversion and ignition behavior. Methanol and ethanol were tested as the fuels of the tf-MTC device. The output performance with ethanol is higher than methanol, which reaches 0.201 V of output voltage and 2.38 μW of the maximum output power. The results show that, when output performance is the primary consideration, ethanol is the great choice; When portability is the primary consideration, methanol is a great choice for the power generation system, and when the device need to work at intervals, methanol is benefit to save fuel without effecting the power output performance of the tf-MTC device.

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