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Microwave-assisted methane decomposition over pyrolysis residue of sewage sludge for hydrogen production

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ABSTRACT

The microwave-assisted methane decomposition over a pyrolysis residue of sewage sludge (PRSS), which acted as a microwave receptor and a low-cost catalyst without further activation, was investigated in a multimode microwave reactor. For comparison purpose, methane conversion (MC) over an activated carbon (AC) was also studied. The results indicate that PRSS is a better microwave receptor than AC. Under the same microwave power (MWP), MC over PRSS is markedly higher than that over AC, due to the remarkably higher Microwave heating (MWH) performance of PRSS. MWH of PRSS and AC is heavily influenced by atmosphere. Under the same MWP, the stable temperatures of the catalysts in hydrogen, nitrogen and methane atmosphere follow the sequence: $T_{nitrogen} > T_{hydrogen} > T_{methane}$. On the other hand, it was observed that nitrogen showed different effect on MC over PRSS and AC under MWH. Specifically, under the microwave-assisted methane decomposition reactions, the effect of nitrogen on MC over PRSS is not obvious, but it has remarkable effect on MC over AC. Additionally, a large number of molten beads were formed on the surface of the used PRSS by microwave irradiation. The composition and formation mechanism of the molten beads were also reported.

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Introduction

Hydrogen is regarded as the sustainable and clean energy of the future. Recently, steam reforming of natural gas (SMR) is the most widely used technology for large scale hydrogen production. However, SMR produces large amount of CO_2 emissions. One alternative to this process is thermo-catalytic decomposition of natural gas (reaction (1)), which is potentially able to produce hydrogen in a single-step process without CO/CO_2 emission [1,2].

$$CH_4 \leftrightarrow C + 2H_2 \quad \Delta H_{298} = 75.6\ kJ\ mol^{-1} \tag{1}$$

Noncatalytic thermal decomposition of methane requires quite a high temperature (1500–2000 K) in order to obtain a reasonable hydrogen yield. Therefore, catalysts are used in order to reduce the temperature of methane decomposition. Many references can be found in the literature related the methane decomposition using metal catalysts [3–6]. More recently, Muradov et al. [7] have proposed the use of carbon catalysts instead of metal ones, owing to some advantages of carbon catalysts: (i) high temperature resistance; (ii) lower

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Table 1 – Proximate and ultimate analysis of dried sewage sludge.

Proximate analysis, wt.%				Ultimate analysis, wt.%				
Moisture	Ash	Volatile matter	Fixed carbon	C	H	O	N	S
2.5	30.6	61.0	5.9	35.9	5.9	19.2	4.7	1.2

price; (iii) tolerance to sulfur and other potentially harmful impurities in the feedstock; (iv) no need for the regeneration of the catalyst by burning carbon off the catalyst surface; (v) production of a marketable byproduct carbon; and (vi) mitigation of CO_2 emission.

It was reported that the active centers of carbon catalysts for methane decomposition are high energy sites (HES) including surface defects, dislocations, vacancies and other energetic abnormalities, and that lower degree of crystallinity gave a larger number of HES, and therefore, a higher catalytic activity [8,9]. Thus activated carbon (AC) and carbon black (CB), which are typical amorphous carbon catalysts, show relatively higher catalytic activity than other more ordered carbon materials, such as graphite, diamond, carbon fibers and carbon nanotubes [7–9]. More recently, Bai et al. [10] explored a kind of coal char as a low-cost carbonaceous material, with a similar activity for methane decomposition to improve the economics of the process. Zhang et al. [11–13] reported that mesoporous carbon could be prepared from coal char by KOH activation, and it showed high catalytic activity and stability for methane decomposition.

On the other hand, the operation of endothermic catalytic reactors is limited by the transfer of radial heat through the packed bed, which will result in a pronounced temperature profile and finally affecting the rate of reaction [14]. The microwave heating (MWH) of dielectric materials offers a number of advantages over conventional heating (CVH), such as volumetric heating, selective material heating, quick start-up and stopping, heating from the interior of the material body, and higher level of safety and automation [15,16]. Cooney et al. [17] compared the MWH and the CVH applied to the production of hydrogen by means of the decomposition of pure methane over a coal char catalyst. MWH was found to require temperatures 30–50 °C lower to give the same methane conversion (MC) obtained with CVH. Domínguez et al. [18] reported that MWH gives rise to higher MC than CVH, probably due to the formation of hot spots (micro-plasmas) inside the catalyst bed. Nitrogen was found to be favorable for the generation of more energetic microplasmas, i.e., hot spots of higher temperature that may favor methane decomposition [19].

Since many of the studies focused on relatively expensive carbon such as AC and CB, it is necessary to explore a low-cost carbonaceous material with high catalytic activity. In China, sewage sludge production has been increasing significantly in recent years due to more stringent regulation of wastewater treatment [20]. Pyrolysis of sewage sludge is being extensively studied as an interesting energy alternative, and pyrolysis residue of sewage sludge (PRSS) is one of the main pyrolysis products [21]. Since PRSS is usually characterized as high ash content and low heating value, it was considered landfill material rather than an alternative fuel [22]. Therefore, it would be a promising technology to use PRSS as a catalyst for microwave-assisted methane decomposition, given that PRSS showed a high catalytic activity under MWH. However, to the best of our knowledge, there is no public report on microwave-assisted MC over PRSS.

In this study, methane decomposition over PRSS was investigated under MWH, and it was compared with methane decomposition over AC. Special attentions were paid to the MWH characteristics of PRSS and AC in methane, nitrogen and hydrogen atmosphere, and to the effect of nitrogen on MC over PRSS and AC. The difference between MWH and CVH was also evaluated.

Experimental

Preparation of catalysts

A kind of mechanically dewatered sewage sludge with moisture content of 80 wt.% was sampled from a municipal wastewater treatment plant in Shanghai, China. The wet sludge samples were dried in an electric drying cabinet at 105 °C for 12 h. Proximate and ultimate analysis of the dried sludge (as shown in Table 1) were carried out in a SDLA618 thermo-balance and a Vario EL III apparatus, respectively. It should be noted that the content of C, H, N, S in the dried sludge were measured by the Vario EL III apparatus directly, and the content of O was calculated by subtraction. The dried sludge samples were pyrolyzed in nitrogen atmosphere in an electric furnace at 800 °C for 30 min. PRSS was produced and collected as a catalyst for methane decomposition experiments. For comparison purpose, a commercial AC, made from coconut shell and activated with steam, was also used. The particles of PRSS and AC were sized within 0.45–1.0 mm. Table 2 shows the elemental analysis of PRSS and AC. The metal elements (K, Al, Fe, Ca, Na, Mg, Ni, Co) in PRSS and AC were analyzed in an inductively coupled plasma emission spectrometer (ICP-AES, Leeman Prodigy).

Table 2 – Elemental analysis of PRSS and AC.

	C	H	O	N	K	Al	Fe	Ca	Na	Mg	Ni	Co
	wt.%				mg g^{-1}							
AC	91.8	2.1	4.0	0.8	0.3	0.6	0.9	2.9	5.5	0.6	<0.1	<0.1
PRSS	18.3	0.7	36.0	1.4	7.4	90.0	57.1	49.2	3.6	8.7	0.5	<0.1

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