

Electronic Supplementary Information

Sustainable processing of waste plastics to produce high yield hydrogen-rich synthesis gas and high quality carbon nanotubes

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Table 1 Reaction conditions and yield of products for the gasification of polypropylene.

Gas yield: weight of produced gases (CO, H₂, CO₂, CH₄, C₂H₄, C₂H₆, C₃H₈, C₃H₈, C₄H₈ and C₄H₁₀) divided by the weight of the waste polypropylene (WPP). Yield of CO and CO₂: weight of the CO and CO₂ divided by the weight of WPP.

Catalyst	Ni/Ca-Al	Ni/Zn-Al
Pyrolysis temperature (°C)	500	500
Gasification temperature (°C)	800	800
Polypropylene weight (g)	0.5	0.5
Carrier gas N ₂ flow (ml min ⁻¹)	80	80
Water injection rate (g h ⁻¹)	4.74	4.74
Catalyst weight (g)	0.25	0.25
Gas yield	137.8	186.1
Yield of CO and CO ₂ (wt.%)	111.9	144.3

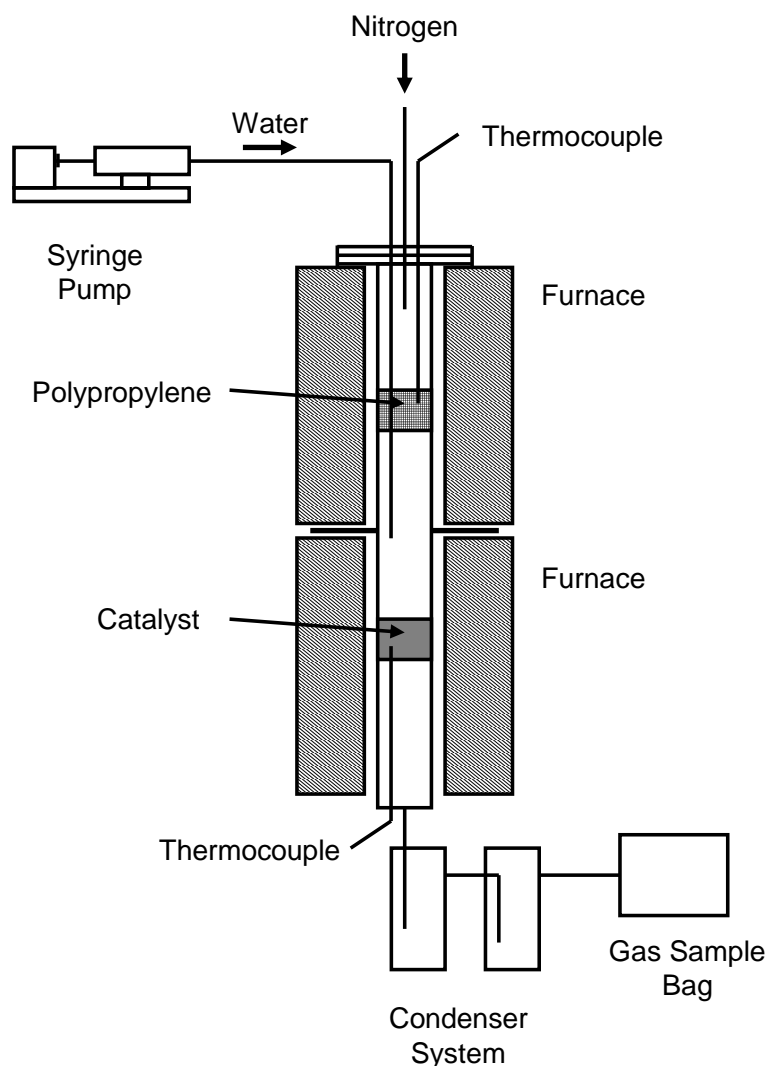


Fig. 1 Diagram of the reaction system. N_2 was used as carrier gas during all the reaction period. The temperature of the second stage was initially heated to the setting point (gasification temperature). Then the first reactor was started to be heated to the pyrolysis temperature at a heating rate of $40\text{ }^\circ\text{C min}^{-1}$. At the mean time, water was injected to the place between the two reactors. Chemical compounds derived from the first pyrolysis stage were reformed in the presence of steam and catalyst at the second stage. The products after gasification were cooled to collect the condensed liquid. The non-condensed gases were collected using a Tedlar™ gas bag, and further analyzed with gas chromatograph. The process conditions were shown in Table 1.

Experimental method

Gasification of waste polypropylene was conducted with a fixed-bed two-stage reaction system. Waste plastic was pyrolysed in the first stage and the derived compounds were gasified in the presence of steam and catalyst in the second reactor. The generated non-condensed gas was analyzed with gas chromatography. A high resolution scanning electron microscope (SEM, LEO 1530) and transmission electron microscope (TEM) (Philips CM200) were used to characterize the CNTs deposited on the reacted catalyst. The temperature-programmed oxidation (TPO) of reacted catalysts was carried out using a Stanton-Redcroft thermogravimetric analyser (TGA and DTG) to determine the properties of the CNTs deposited on the reacted catalysts. Raman spectra of CNTs were obtained using a Renishaw Invia equipment at a wavelength of 514 nm.

The catalysts were prepared by co-precipitation method with the initial Ni-loading mole ratio was 20%. $\text{Ni}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ($\geq 97.0\%$), $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ ($\geq 99\%$), $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ($\geq 99\%$), $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ($\geq 99\%$), Na_2CO_3 ($\geq 99\%$), and $(\text{NH}_4)_2\text{CO}_3$ ($\geq 98\%$) were purchased from Sigma-Aldrich. Precursors with desired Ni/Ca-Al (Ca/Al molar ratio is 1:1, Ni content is 20 mol.%) or Ni/Zn-Al (Zn/Al molar ratio is 1:4, Ni content is 20 mol.%) ratio were prepared by dissolving the certain amount of metal salts in water. The precursor mixture was precipitated with the basic solution of Na_2CO_3 or $(\text{NH}_4)_2\text{CO}_3$ drop by drop in order to adjust the pH of the suspension between 6 and 9. After precipitation, the suspension was aged under agitation for an hour and then filtered under vacuum. The filter cake obtained was rinsed with deionized water for several times and followed by drying at 80 °C overnight. At the end, the solids were calcined at 800 °C for 4h with a heating rate of 1 °C min⁻¹ in static air.