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Article

Effect of Bed Hydrodynamics on Microwave-Assisted Steam-Carbon Gasification for Hydrogen Production: A Comparison of Packed and Fluidized Bed Reactors

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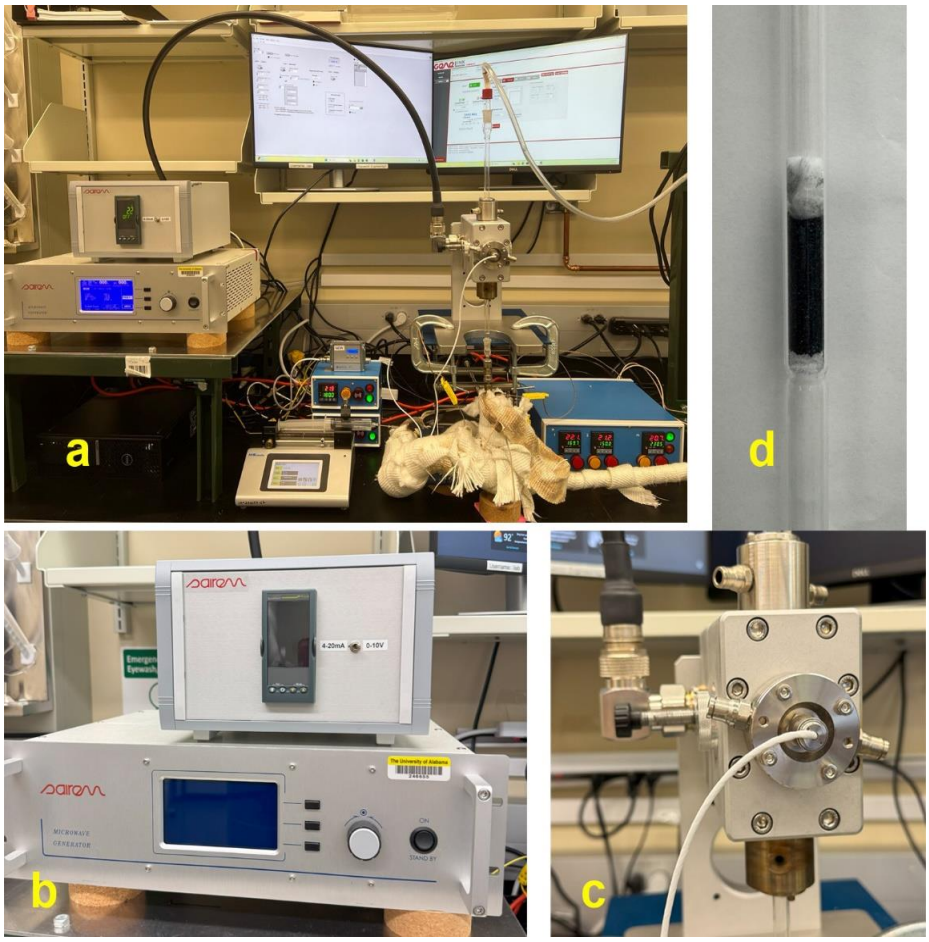


Figure S1. (a) Experimental setup, (b) microwave generator and controller, (c) microwave cavity, and (d) quartz reactor.

Table S1. Repeatability of experiments

Exp no	Yield	Uncertainty	Uncertainty 90%
P700-R2	24.9	4.493	7.369
P700-R2 repeat	25.2	3.889	6.379

Table S2. Thermochemical properties and lower heating values of key gaseous species (STP)

Gas	Lower Heating Value (LHV) (kJ/mol)	Chemical Exergy (exch) (kJ/mol)
H2	241.8	236
CO	283	275
CH4	802.3	831
CO2	0	19.87
C (graphite)	393.5	410
H2O (gas)	-	9.5
N2	-	0.72

Table S3. Fluidized bed parameters

Properties	Symbol	Value
Particle diameter (μm)	d_p	284
Particle bulk density (kg/m3)	ρ_b	-
Particle density (kg/m3)	ρ_d	450
Fluid density (kg/m3)	ρ_f	0.849279
Voidage	ε	0.5
Bed height (mm)	h_b	34

Table S4. Sensors' uncertainties

Item	Measuring Instruments	Specification	Uncertainty (%)
1	Mass Flow meter	FMA-1600A	0.08 (%RD) + 02 (%FS)
2	Mass Flow Controller	GE50A	±0.2 % (%FS)
3	Gas Analyzer	ETG MCA 100 syn	±2 % (FS)
4	Syringe Pump	KDS, legato 100,	0.35 %(RD)
5	IR Pyrometer	CT LT22	±1°C

Table S5. Exergy balance for all experiments (dead state: T_0 = 298.15 K, P_0=101.325 kPa).

Experiment Label	EX_{mw} kJ	EX_c kJ	EX_{steam} kJ	EX_{in} (°C) kJ	EX_{chem} kJ	EX_{mix} kJ	$EX_{products}$ kJ	EX_{loss} kJ	η_{ex} %
P600-R3	52.30	0.74	0.42	53.46	0.84	−0.05	0.79	52.67	1.48
P650-R3	120.69	3.97	0.76	125.42	4.53	−0.19	4.35	121.07	3.47
P700-R3	115.27	6.99	0.63	122.89	8.04	−0.27	7.77	115.12	6.32
P750-R3	92.03	5.18	0.50	97.71	5.95	−0.20	5.75	91.96	5.88

P700-R1	125.40	7.40	0.31	133.11	8.45	−0.25	8.2	124.91	6.16
P700-R2	128.87	3.42	0.26	132.55	3.78	−0.13	3.65	128.9	2.76
P700-R4	136.77	4.93	0.92	142.61	5.69	−0.20	5.5	137.12	3.85
F600-R3	136.29	0.68	0.48	137.45	0.72	−0.04	0.68	136.77	0.49
F650-R3	164.36	2.02	0.40	166.78	2.12	−0.11	2.01	164.77	1.21
F700-R3	211.79	4.01	0.55	216.35	4.6	−0.22	4.38	211.97	2.02
F750-R3	262.93	6.33	0.56	269.81	7.31	−0.31	7	262.81	2.59
F700-R1	259.30	4.98	0.29	264.56	5.67	−0.25	5.42	259.15	2.05
F700-R2	143.24	2.65	0.34	146.23	2.88	−0.15	2.74	143.5	1.87
F700-R4	149.61	2.59	0.85	153.04	2.94	−0.15	2.79	150.26	1.82

The energy and exergy analyses were performed with the system boundary at the inlet of the microwave cavity. The dead state was defined as $T_0 = 298.15$ K and $P_0 = 101.325$ kPa. The exergy input consists of the absorbed microwave energy, the chemical exergy of the converted carbon, and the chemical and physical exergy of the inlet steam:

$$Ex_{in} = Ex_{mw} + Ex_C + Ex_{steam} \qquad (1)$$

Because microwave energy is electromagnetic work, its exergy equals the absorbed microwave energy:

$$Ex_{mw} = W_{mw} = \sum P_{absorbed}\,\Delta t \qquad (2)$$

The exergy of the converted carbon is:

$$Ex_C = n_C\, ex_{ch,C}, \qquad n_C = n_{CO} + n_{CO_2} + n_{CH_4} \qquad (3)$$

The exergy of the inlet steam, supplied at $T_{in} = 413.15$ K, is:

$$Ex_{steam} = n_{H_2O}\left(ex_{ch,H_2O} + ex_{ph,H_2O}\right) \qquad (4)$$

$$ex_{ph,H_2O} = c_{p,H_2O}\left[(T_{in} - T_0) - T_0 \ln\frac{T_{in}}{T_0}\right] \qquad (5)$$

The exergy of the product gas is the sum of its chemical, physical, and mixing exergy:

$$Ex_{products} = Ex_{ch} + Ex_{ph} + Ex_{mix} \qquad (6)$$

$$Ex_{ch} = n_{H_2}\, ex_{ch,H_2} + n_{CO}\, ex_{ch,CO} + n_{CH_4}\, ex_{ch,CH_4} \qquad (7)$$

$$Ex_{mix} = R\,T_0 \sum_i n_i \ln y_i, \qquad y_i = \frac{n_i}{\sum_i n_i} \qquad (8)$$

where the summation in Eq. (8) includes H_2, CO, CO_2, CH_4 and N_2 in the dry product gas. Because the product gas leaves the cooling unit at the dead-state temperature, $Ex_{ph} = 0$, and its sensible heat is included in the exergy loss. The exergy loss and the exergy efficiency are:

$$Ex_{loss} = Ex_{in} - Ex_{products} \qquad (9)$$

$$\eta_{ex} = \frac{Ex_{products}}{Ex_{in}} \times 100 \qquad (10)$$

N_2 is inert, so its chemical exergy at the inlet and outlet cancels. Its physical exergy at the cavity inlet (140 °C) is less than 0.1% of the total exergy input and was neglected. N_2 is included in the mixing exergy (Eq. 8) through the mole fractions of the product gas.

Table S5. Electrical energy consumption and microwave energy

Experiment No	E_{cf} (J)	$\dot{W}_{mw}$ (kJ)
F600-R3	0.5	136.2
F650-R3	0.5	164.2
F700-R3	1.5	211.7
F750-R3	0.6	262.9
F700-R1	0.6	247.9
F700-R2	0.6	143.2
F700-R4	0.5	149.6

Uncertainty and error propagation analysis

The uncertainty analysis was conducted by following [1]. The experimental uncertainty is the combination of the sensor bias and the standard error, $\sigma_m = \sum\{(x_i - \langle x \rangle)^2 / [[n(n-1)]]\}^{0.5}$, where x_i are the instantaneous results, $\langle x \rangle$ are the results measurements, and n is the number of measurements. For indirect measurements of a variable V, we used the combined uncertainty, $u(V)$. The total uncertainty was calculated as $V = \langle V \rangle \pm tu(V)$, where t is Student's t factor calculated with a confidence interval of 90%.

The uncertainty in a function (δf) that depends on several independent variables (γ_i) can be estimated using the general uncertainty propagation formula when the final function is variables product or quotient of γ_i for independent random errors [2].

$$\delta f = \sqrt{\sum_{i=1}^{N} \left(\frac{\delta \gamma_i}{\gamma_i}\right)^2} \quad (1)$$

This approach allows for systematic assessment of how uncertainties in measured parameters propagate through the calculation to affect the final reported values. This method has been widely applied in various chemical engineering contexts to quantify experimental uncertainties [3, 4]. In total, 3 uncertainties in calculation for hydrogen yield are identified. These variables come from uncertainties related to the gas analyzer (q_g), mass flowmeter (q_f), and the balance used for weighing initial AC (m_{AC}).

Table S6 shows the most significant uncertainties of indirect measurements obtained concerning all the tests performed. Uncertainties of physical properties (e.g., density, molecular mass) were disregarded.

Table S6. – Indirect measurements uncertainty.

Parameter	Uncertainty propagation calculation
Hydrogen yield	$\delta y_{H2} = y_{H2} \sqrt{\left(\frac{\delta m_{AC}}{m_{AC}}\right)^2 + \left(\frac{\delta q_f}{q_f}\right)^2 + \left(\frac{\delta q_g}{q_g}\right)^2}$

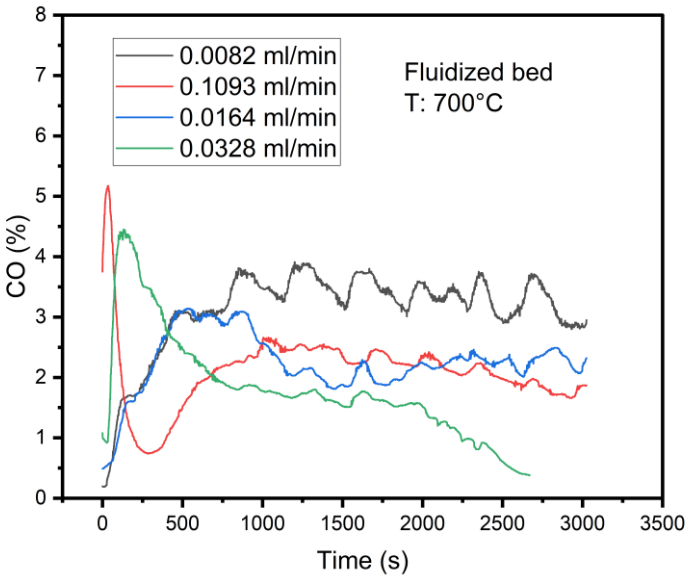


Figure S2. CO concentration in packed bed at constant temperature T=700 °C and different water flowrate

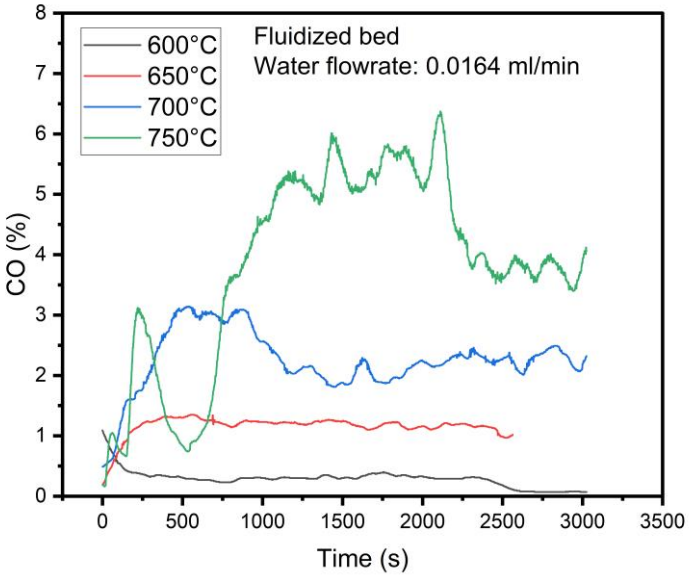


Figure S3. CO concentration in fluidized bed at constant water flowrate= 0.0164 ml/min and different temperature

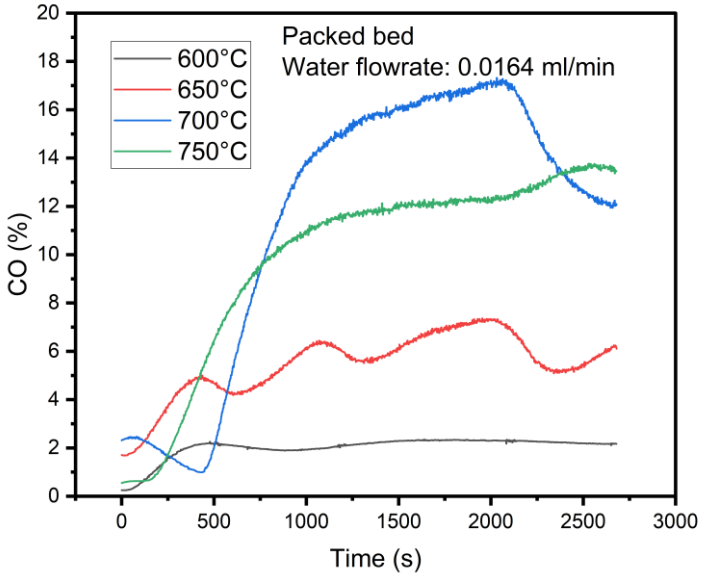


Figure S4. CO concentration in packed bed at constant water flowrate =0.0164 ml/min and different temperature

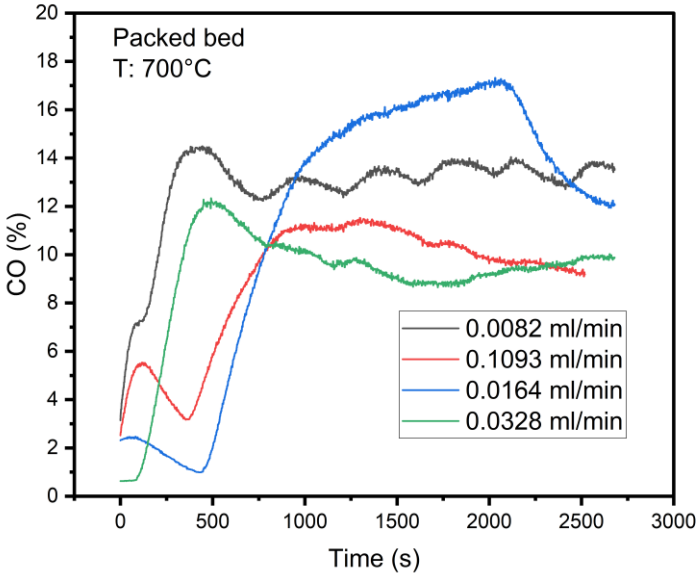


Figure S5. CO concentration in packed bed at constant temperature T=700 °C and different water flowrate

Table S8. Performance parameters and their measurement uncertainties (90% confidence level) for all experiments.

Experiment Label	H_2 production rate	Total H_2	Carbon conversion	Energy efficiency	Exergy efficiency	Hydrogen yield
P600-R3	0.00112± 0.00024	0.0031± 0.00067	2.9±0.4	2.47 ± 0.28	1.48 ± 0.26	
P650-R3	0.00409± 0.00128	0.02038± 0.00636	15.6±5.5	3.72 ± 1.06	3.47 ± 0.99	
P700-R3	0.00725± 0.0016	0.02959± 0.00653	27.6±7	12.52 ± 1.24	6.32 ± 1.16	
P750-R3	0.00696± 0.00152	0.02256± 0.00494	20.5±5	11.58 ± 1.23	5.88 ± 1.15	
P700-R1	0.00655± 0.00177	0.02671± 0.00721	29.2±7.2	13.36 ± 1.31	6.16 ± 1.23	
P700-R2	0.00469± 0.00139	0.01211± 0.00359	13.5±4	5.56 ± 0.70	2.76 ± 0.66	
P700-R3	0.00725± 0.0016	0.02959± 0.00653	19.4±5	12.52 ± 0.86	3.85 ± 0.80	
P700-R4	0.00733± 0.0018	0.02199± 0.0054	27.6±7	7.25 ± 1.24	6.32 ± 1.16	
F600-R3	2.65×10^{-4}±	8.26×10^{-4}±	2.7±0.1	0.56 ± 0.03	0.49 ± 0.03	09±0.04
F650-R3	0.00222± 0.00448	0.00585± 0.01609	8±0.3	1.80 ± 0.07	1.21 ± 0.06	7.8±0.3
F700-R3	0.00408± 0.00448	0.01467± 0.01609	15.8±13.5	3.7 ± 1.58	2.02 ± 1.46	14.8±16.2
F750-R3	0.00598± 0.00425	0.02189± 0.01556	33.8±15.4	5.2 ± 1.31	2.59 ± 1.22	21.9±15.6
F700-R1	0.00423± 0.00442	0.01575± 0.01646	19.7±16.6	4.1 ± 1.44	2.05 ± 1.34	31.6±33
F700-R2	0.00296± 0.00316	0.00996± 0.01063	24.2±8.1	3.74 ± 1.37	1.87 ± 1.26	16±17.1
F700-R3	0.00408 ± 0.00448	0.01467 ± 0.01609	15.8±13.5	3.7 ± 1.58	2.02 ± 1.46	14.8±16.2
F700-R4	0.00354± 0.00339	0.00978± 0.00936	10.2±3.4	3.3 ± 1.04	1.82 ± 0.96	6.2±5.9

References

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2. Yang, X., et al., *Uncertainty analysis of adsorption measurements using commercial gravimetric sorption analyzers with simultaneous density measurement based on a magnetic-suspension balance.* Adsorption (Boston), 2020. **26**.
3. Badalyan, A. and P. Pendleton, *Analysis of Uncertainties in Manometric Gas-Adsorption Measurements. I: Propagation of Uncertainties in BET Analyses.* Langmuir, 2003. **19**(19): p. 7919-7928.
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