Supplementary: Conceptual Design of Pyrolytic Oil Upgrading Process Enhanced by Membrane-Integrated Hydrogen Production System

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The space velocity of the packed-bed reactor determined the performance directly. The impacts of the reactor size on space velocity are shown in Figure S1. As the figure shows, greater length and diameter of the reactor led to lower space velocity, which would increase the residence time of the fluid (oil and hydrogen), and hence increase the conversion. Figure S1 indicates that when the reactor diameter was 3-5 m, and the bed height was higher than 30 m, the space velocity was 0.1-2 h⁻¹, which could satisfy the requirements of a normal HT process.

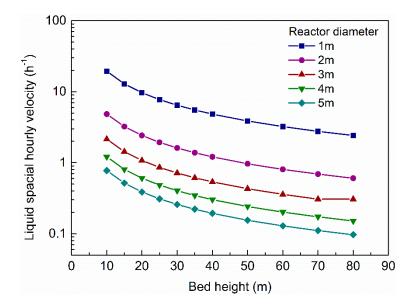


Figure S1. Liquid special hourly velocity of pyrolytic oil hydroprocessing reactor.

The influences of membrane area (membrane-203) on the conversion and hydrogen consumption are shown in Figure S2. Because of the small capacity of membrane-203, doubling the membrane area provided little impact on the performance, and hence during the investigations the membrane area of membrane-203 was set at a constant 500 m². Note that membrane-203 was still a indispensable unit for hydrogen recovery, since the gases recovered by it could compensate the cost of the membrane and module.

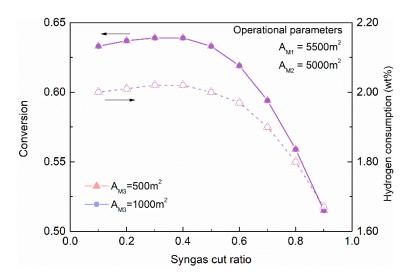


Figure S2. Impacts of membrane area (Membrane-203) on conversion and hydrogen consumption. The mass balance of the proposed SMR-HT process was shown in Table S1.

| Name | Natural gas | Steam | 109 | Sweet Syngas | Pyrolytic Oil | Hydro Product | Hydrogen | 218 | 223 | Upgrade d Oil | Air | Flue gas | Vent |
|-----------------------|----------------|----------------|----------------|-----------------|------------------|------------------|----------|-----------|------------|------------------|----------------|----------------|------------|
| | | | | | | | | | | | | | |
| Temperature, °C | 20 | 500 | 90 | 30 | 370 | 370 | 40 | 150 | 90 | 151 | 60 | 1059 | 90 |
| Pressure, kPa | 3100 | 3200 | 2591 | 11,000 | 10000 | 10,000 | 2450 | 9900 | 2500 | 300 | 300 | 78 | 2500 |
| Molar Flow, kmol/h | 1522.80 | 6651.91 | 10,427.16 | 4866.48 | 420.95 | 4447.83 | 2551.14 | 3623.87 | 66.99 | 675.09 | 5753. 14 | 6676. 76 | 12.26 |
| Mass Flow, kg/h | 24,536.27 | 119,834. 78 | 144,376.8 7 | 23,944.23 | 119,010.00 | 142,951.41 | 5167.10 | 21,660.73 | 471.4 4 | 111,824.6 2 | 165,9 79.65 | 196,0 85.14 | 267.8 5 |
| Component | Flow | | | | | | | | | | | | |
| H2, kmol/h | 7.61 | 0.00 | 3922.92 | 4207.59 | 0.00 | 3019.23 | 2549.46 | 2955.90 | 50.50 | 12.41 | 0.00 | 0.00 | 0.00 |
| N2, kmol/h | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 4544. 98 | 4545. 03 | 0.00 |
| O2, kmol/h | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 1208. 16 | 57.53 | 0.00 |
| CH4, kmol/h | 1507.57 | 0.00 | 381.67 | 241.49 | 0.00 | 252.95 | 1.63 | 243.01 | 7.40 | 4.05 | 0.00 | 0.00 | 6.32 |
| CO, kmol/h | 0.00 | 0.00 | 589.10 | 406.99 | 0.00 | 425.84 | 0.04 | 414.31 | 8.69 | 2.76 | 0.00 | 0.00 | 5.94 |
| CO2, kmol/h | 7.61 | 0.00 | 544.65 | 0.00 | 0.00 | 0.25 | 0.00 | 0.23 | 0.01 | 0.01 | 0.00 | 1137. 05 | 0.00 |
| H2O, kmol/h | 0.00 | 6651.91 | 4988.81 | 10.41 | 0.56 | 11.24 | 0.00 | 10.41 | 0.40 | 0.42 | 0.00 | 937.1 5 | 0.00 |
| HNV, kg/h | 0.00 | 0.00 | 0.00 | 0.00 | 58,690.12 | 22,314.48 | 0.00 | 0.00 | 0.00 | 21,306.16 | 0.00 | 0.00 | 0.00 |
| LNV, kg/h | 0.00 | 0.00 | 0.00 | 0.00 | 43,910.49 | 48,302.67 | 0.00 | 0.00 | 0.00 | 48,225.50 | 0.00 | 0.00 | 0.00 |
| Phenol, kg/h | 0.00 | 0.00 | 0.00 | 0.00 | 14,660.63 | 8509.23 | 0.00 | 0.00 | 0.00 | 8052.19 | 0.00 | 0.00 | 0.00 |
| Aromatics, kg/h | 0.00 | 0.00 | 0.00 | 0.00 | 1737.38 | 41,448.56 | 0.00 | 0.00 | 0.00 | 34,036.06 | 0.00 | 0.00 | 0.00 |
| Gases, kg/h | 0.00 | 0.00 | 0.00 | 0.00 | 1.39 | 89.93 | 0.00 | 0.00 | 0.00 | 29.55 | 0.00 | 0.00 | 0.00 |

Table S1. Mass balance of SMR-HT process.