



Zirconia and alumina based catalysts for steam reforming of naphthalene

Francesco Ferella^{a,*}, Joachim Stoehr^a, Ida De Michelis^b, Andreas Hornung^a

^a Institute of Technical Chemistry-Thermal Waste Treatment, ITC-TAB, Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

^b Department of Chemistry, Chemical Engineering and Materials, University of L'Aquila, Monteluco di Roio, 67040 L'Aquila, Italy

H I G H L I G H T S

- ▶ ZrO₂ and Al₂O₃ catalysts with Ni, Cr, Fe, Ce, Co and Pt were tested.
- ▶ Al₂O₃ catalysts had 90–100% C₁₀H₈ conversion, 30–40% CO, 300–350% H₂ yield at 550 °C.
- ▶ Al₂O₃-supported catalysts were more active on average than the zirconia ones.
- ▶ It was possible to regenerate catalysts by O₂ and steam.
- ▶ Activation of catalyst by H₂ increased performance even at 450 °C.

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The present paper deals with experimentation of ZrO₂ and Al₂O₃-supported catalysts for conversion of naphthalene, chosen as tar model compound of pyrolysis or gasification syngas. In particular, the reforming capacity of active metals and promoters such as Co, Ni, Fe, Cr, Ce and Pt was tested in a fixed bed reactor at temperature from 400 to 900 °C. As regards ZrO₂-supported catalysts, the best results were achieved by the Ni/Fe/Pt catalyst with 96% naphthalene conversion, 78% and 280% as CO and H₂ production yield at 800 °C. Regarding Al₂O₃-supported catalysts, they were more active on average than the zirconia ones, achieving a very good performance even at 500 °C (90–100% naphthalene conversion, 30–40% CO yield and 300–350% H₂ yield at 550 °C). Influence of different amounts of alumina, montmorillonite and carbon on carrier composition as well as pellets' size were also studied. Both zirconia and alumina catalysts showed deactivation at higher temperatures due to coke deposition, resulting in a strong H₂ production drop. Regeneration of catalysts by O₂ and steam as well as activation by H₂ were also studied. The activated catalyst was able to convert more than 99% naphthalene at 450 °C with a CO and H₂ production yield of 26% and 420%, respectively.

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1. Introduction

Hydrocarbons derived from petroleum, natural gas or coal are essential in many ways to modern life and its quality. An ever-increasing world population and energy consumption, compared with non-renewable fossil fuel resources, which will be increasingly depleted, are clearly on a collision course. According to the most reliable projections, the demand for energy is expected to grow from 120 PWh to around 189 PWh in 2025 [1]. With regard to electric energy generation, coal still represents the 41% of the total world energy: in some countries such as China, India, Australia, South Africa, Poland and Kazakhstan more than 69% of the total electricity comes from coal. The importance of coal to electricity generation worldwide is set to continue, with coal fuelling 44% of global electricity in 2030: this will be due to coal availability and

rising prices of other fossil fuels [2]. On the other hand, the plentiful coal reserves will be available at a higher socio-economic and environmental cost because of CO₂ emission and subsequent global warming. One possible substitute as a fuel or raw material for chemical industry is represented by pyrolysis and gasification of organic materials from agriculture and forestry. Depending on subsequent processes the gas requires additional cleaning steps which at least includes removal of particulate matter and conversion of tar. Thermal and catalytic processes are widely used in industrial applications. If the flue-gas is directly burnt in a furnace, for example in a oil- or coal-fired power plant, there are no problems in terms of tar content. The only restriction is heat insulation of pipelines since gas must be kept at temperature greater than the dew point of tar compounds. Nevertheless, in internal combustion engines (ICE) for power generation the feeding gas has to be at cold temperature, so tar is indeed a problem and must be removed. If the syngas feeds gas turbines or fuel cells, tar concentration must be very low, since they condense on cold surfaces giving mechanical problems [3].

* Corresponding author. Tel.: +39 0862 434221; fax: +39 0862 434203.

E-mail address: francesco.ferella@univaq.it (F. Ferella).

Table 1
Summary of main experimental studies on tar cracking and reforming catalysts.

Reactor	Catalyst/Active metals	Biomass	Representative tar compound	Catalyst temperature (°C)	Cracking yield	Reference
External catalytic fixed bed	Ni-based Calcined dolomite	Almond shells	–	770	2.5 g tar kg ⁻¹ biomass 1.9 m ³ kg ⁻¹ biomass	[3]
Fixed bed	Y-zeolite Ni–Mo Silica Alumina Lime	–	1-Methylnaphthalene	550	98% (9 h) 100% (9 h) 73% (9 h) 80% (9 h) 78% (9 h)	[4]
Single and dual in-bed	Rh/CeO ₂ /SiO ₂ (in-bed, single and dual-bed reactor, FBR)	Cellulose Cedar wood Jute stick Baggase Rice straw	–	550–650	94–99% (20 min) 78–81% 71–84% 68–82% 65–80%	[5]
External catalytic fixed bed	Fe-based Dolomite	Birch	–	700–900	16–2.2 g tar kg ⁻¹ biomass 16–2.4 g tar kg ⁻¹ biomass 0.63–0.97 m ³ kg ⁻¹ biomass 0.83–1.12 m ³ kg ⁻¹ biomass	[6]
Fixed bed	Calcined olivine	–	Naphthalene	900	80%	[7]
Fixed bed	Ni/dolomite	–	Naphthalene	500–700	45–95% (1 h)	[8]
External catalytic fixed bed	Dolomite Silica	Miscanthus pellets Birch Straw pellets Olive kernels	–	700–900	1.3 mg C ₁₀ H ₈ /g feed (900 °C)	[9]
Fluidized in-bed gasifier	Olivine Calcined dolomite	Almond shells	–	700–820	6–1 g tar kg ⁻¹ biomass 0.7 g tar kg ⁻¹ biomass (770 °C)	[10]
Fixed bed	Ni/calcined natural olivine	–	Methane	650–850	95% CH ₄ conversion (750 °C) 80% H ₂ yield (750 °C)	[11]
Fluidized in-bed gasifier	Co/MgO	Radiata pine	–	600	83% cellulose tar conversion (Co 36% w/w)	[12]
External catalytic fixed bed (two-stages non catalytic steam gasification)	Fe ₂ O ₃ Fe ₂ O ₃ /Al ₂ O ₃	Cedar wood	–	600	40–90% (yield of carbon)	[13]
External catalytic fixed bed (two-stages non catalytic steam gasification)	CaO	(1) Coal tar (2) Petroleum distillation residue (3) Coke oven tar	–	720	23–38 mol H ₂ g ⁻¹ tar 4.5–7.3 mol CO g ⁻¹ tar 1–5.7 mol CH ₄ g ⁻¹ tar	[14]
Fixed bed	Ni/MgO	–	Naphthalene Benzene Methane	400–950	100% 100% 100%	[15]
Fixed bed	Ni/Al ₂ O ₃	–	Toluene Benzene Naphthalene Anthracene Pyrene	700–875	80% 87% 90% 98% 86%	[16]

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