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Supplementary Information to

Propane to Olefins Tandem Catalysis: a Selective Route Towards Light Olefins Production

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1. Socio-economical Context

1.1 Olefin Markets details

Compound Annual Growth Rate (CAGR) is defined as the rate of return of an investment growing from its beginning balance to its ending balance, assuming the profits are reinvested at the end of each year of the investment's lifespan. It is according to the formula:

$$CAGR\ (\%) = 100\left[\left(\frac{balance_f}{balance_i}\right)^{\frac{1}{n}} - 1\right]$$

Where $balance_f$ and $balance_i$ are the final and initial balance over n years.

Table S1. Global ethylene demand by application, as of 2019 [1].

Application	**Demand (%)**
Polyethylene	60.9
Ethylene Oxide	15.0
Ethylene Dichloride	9.5
Others	14.6

Table S2. Global propylene demand by application, as of 2019 [2].

Application	**Demand (%)**
Polypropylene	68
Propylene Oxide	9
Acrylonitrile	7
Others	16

Table S3. Global alpha-olefin market by type, as of 2019 [3].

Type	**Production (wt. %)**
1-butene	19
1-hexene	29
1-octene	25
1-decene	11
1-dodecene	5
Others	10

Table S4. Market specifications for ethylene, propylene and alpha-olefins [1,2,4]

Olefin	Worldwide production in 2019 (Mt)	Market size (billion $)	CAGR 2015-2019 (%)	Projected CAGR 2019-2023 (%)[a]
Ethylene	207.6	222.1	5.25	4.77

Propylene	110	83.7	2.95	-1.81
α-Olefins	4.6	9.5	3.9	4.2

[a] The disruption caused by the coronavirus outbreak is not accounted for here, but it is set to negatively impact the markets.

1.2 Propylene/propane spread

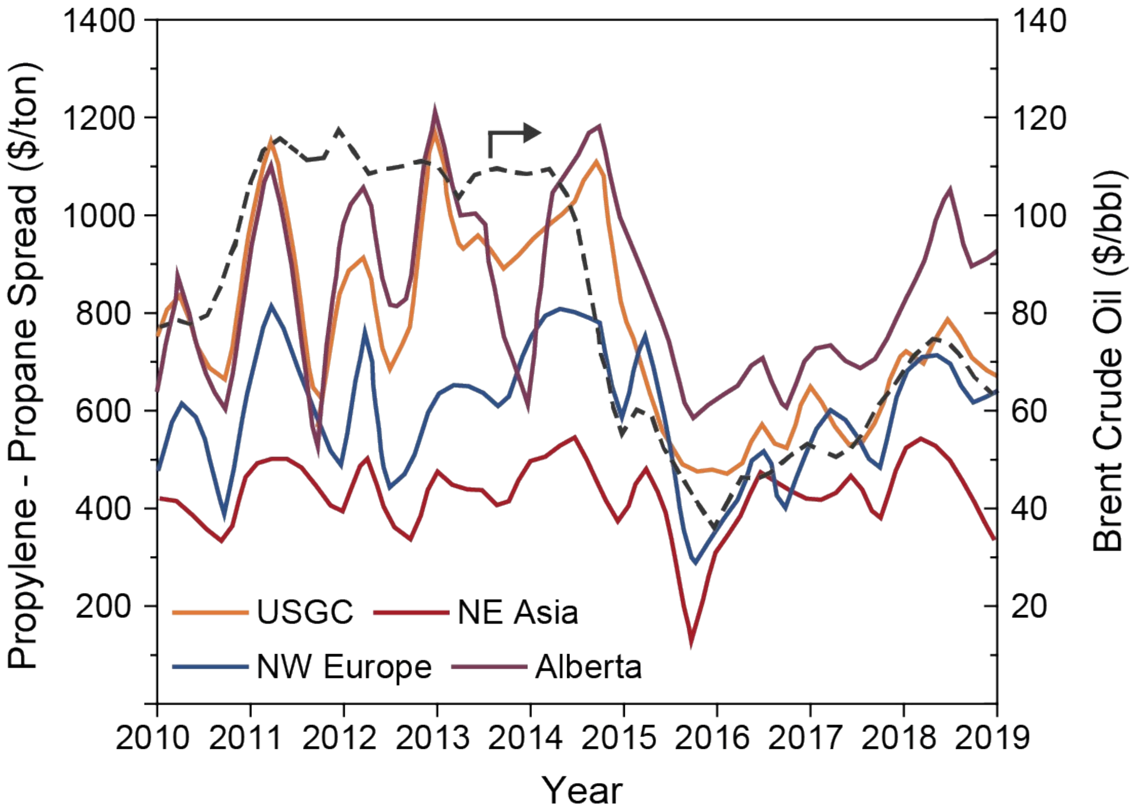

Figure S1. Propylene-propane spread in $ per ton, by region, over the years 2010-2019. Brent crude oil price is reported as a dashed line for reference. Adapted from [5].

1.3 List of PDH, MTO and OCT installations

Table S5. PDH installations worldwide, in alphabetical order, divided by Technology. The capacity of the plant is reported in thousands of metric tons per year, followed by the investment (if known) and the year of (expected) beginning of operation in brackets. For multiple installations on the same locations, each plan details are given separated by commas.

Title	Description	Technology	Longitude	Latitude
Arzew, Algeria	640 km.t./yr, 1.3 bn $ (2022)	Oleflex	-0.31368	35.85635
Ba Ria City, Ba Ria-Vung Tau Province,	306 km.t./yr, 1.2 bn $ (-)	Oleflex	107.1688	10.49627

Vietnam				
Ceyhan, Turkey	457 km.t./yr, 1.4 bn $ (2023)	Oleflex	35.87123	37.05564
Freeport, Texas, United States of America	750 km.t./yr, - bn $ (2016)	Oleflex	-95.3597	28.95414
Fujian, China,	600 km.t./yr, - bn $ (2021)	Oleflex	117.8428	26.545
Houston, Texas, United States of America	658 km.t./yr, 2.1 bn $ (2010)	Oleflex	-95.3633	29.76328
Jubail, Eastern Region, Saudi Arabia - Al-Waha Plant	450 km.t./yr, - bn $ (2006)	Oleflex	49.66012	25.40929
Kuantan, Pahang, Malaysia	300 km.t./yr, - bn $ (2000)	Oleflex	103.322	3.798564
Map Ta Phut, Rayong Province, Thailand	300 km.t./yr, - bn $ (2007)	Oleflex	101.1679	12.71365
Mont Belvieu, Texas, United States of America - Enterprise PDH 2	750 km.t./yr, - bn $ (2023)	Oleflex	-94.9408	29.83879
Police, Police County, Poland	400 km.t./yr, 1.5 bn EUR (2023)	Oleflex	14.56574	53.54864
Qinzhou, Guangxi, China	450 km.t./yr, - bn $ (2021)	Oleflex	108.6208	21.96478
Strathcona County, Canada	525 km.t./yr, 2.7 bn $ (2021)	Oleflex	-113.126	53.57627

Takreer, Abu Dhabi, Abu Dhabi Emirate, United Arab Emirates	500 km.t./yr, - bn $ (2018)	Oleflex	54.53518	24.32352
Tarragona, Catalonia, Spain	350 km.t./yr, - bn $ (2002)	Oleflex	1.254606	41.11724
The Palisades, Edmonton, AB, Canada	550 km.t./yr, 5 bn $ (2023-delayed)	Oleflex	-113.55	53.60174
Tobolsk, Russia	510 km.t./yr, - bn $ (2013)	Oleflex	68.25129	58.19981
Yanbu`, Al Madinah Region, Saudi Arabia - NATPET Plant	420 km.t./yr, - bn $ (2009)	Oleflex	38.06668	24.0889
Zhangjiagang, Jiangsu, China,	600 km.t./yr, - bn $ (2014)	Oleflex	120.5515	31.8783
Zhoushan, Zhejiang, China - Phase 1 and 2	400, 200 km.t./yr, - bn $ (2019, 2020)	Oleflex	122.203	29.98733
Al Jubayl, Eastern Region, Saudi Arabia - SPC, APC Plant and future APC plant	455, 455, 843 km.t./yr, - bn $ (2004, 2007, 2024)	Catofin	49.65322	27.0007
Antwerp, Belgium - INEOS	750 km.t./yr, - bn $ (2023)	Catofin	4.399708	51.22111
Atyrau, Atyrau Region, Kazakhstan	630 km.t./yr, - bn $ (2021)	Catofin	51.92445	47.10534

Beveren, East Flanders, Belgium - Borealis Plant	740 km.t./yr, 1.1 bn $ (2022)	Catofin	4.27803	51.25195
Dalian, Liaoning, China	1000 km.t./yr, - bn $ (2019)	Catofin	121.6283	38.91817
Dongguan, Guangdong Province, China	600 km.t./yr, - bn $ (2021)	Catofin	113.7466	23.04447
Mont Belvieu, Texas, United States of America - Enterprise PDH1	750 km.t./yr, - bn $ (2017)	Catofin	-94.8856	29.84478
Munich, Bavaria, Germany	750 km.t./yr, - bn $ (2023)	Catofin	11.57538	48.13711
Seoul, South Korea - SK Advanced	600 km.t./yr, - bn $ (2016)	Catofin	126.9783	37.56668
Asalouyeh, Bushehr Province, Iran	650 km.t./yr, - bn $ (2020)	STAR Uhde	52.60704	27.476
Bandar-e-Mahshahr County, Iran	450 km.t./yr, - bn $ (2020)	STAR Uhde	49.26043	30.35308
Kachchh, Mundra (CT), Gujarat, India	- km.t./yr, 4 bn $ (2024)	STAR Uhde	69.72541	22.83737
Ludwigshafen am Rhein, Rhineland-Palatinate, Germany - test facility	300 km.t./yr, - bn $ (2022)	STAR Uhde	8.438157	49.47041

Point Comfort, Calhoun County, Texas, United States of America	545 km.t./yr, - bn $ (2016)	STAR Uhde	-96.5603	28.67972
Port Said, Egypt	400 km.t./yr, 0.4 bn $ (2015)	STAR Uhde	32.30551	31.26324
Bahia Blanca, Partido de Bahia Blanca, Argentina	- km.t./yr, - bn $ (-)	Unknown	-62.2655	-38.7177
Binzhou City, Shandong, China	600 km.t./yr, - bn $ (planning)	Unknown	117.9647	37.38212
Caofeidian, Hebei, China	600 km.t./yr, - bn $ (planning)	Unknown	118.454	39.2725
Chaozhou, Guangdong Province, China	600*3 km.t./yr, - bn $ (planning)	Unknown	116.6205	23.65662
Hengshui City, Hebei, China	500 km.t./yr, - bn $ (planning)	Unknown	115.6677	37.73643
Humen, Guangdong Province, China	600*2 km.t./yr, - bn $ (planning)	Unknown	113.6539	22.80413
Jiaxing, Zhejiang, China	450 km.t./yr, - bn $ (planning)	Unknown	120.7511	30.74744
Karachi, Sindh, Pakistan	450 km.t./yr, 1.75 bn $ (2026)	Unknown	67.29624	24.82734
Lianyungang City, Jiangsu, China	1500, 600 km.t./yr, - bn $ (planning)	Unknown	119.2158	34.59786
Luoyang City, Henan, China	200 km.t./yr, - bn $ (planning)	Unknown	112.421	34.65569

Ningbo, Zhejiang, China	660, 600, 600 km.t./yr, - bn $ (2020, planning, planning)	Unknown	121.6204	29.86222
Pinghu, Zhejiang, China	450 km.t./yr, - bn $ (2018)	Unknown	121.0116	30.67824
Qingdao City, Shandong, China	900 km.t./yr, - bn $ (planning)	Unknown	120.3781	36.0638
Raigad, Usar, Maharashtra, India	500 km.t./yr, - bn $ (2022)	Unknown	73.07777	18.42193
Rizhao City, Shandong, China	600*2 km.t./yr, - bn $ (planning)	Unknown	119.5215	35.41507
Taixing City, Jiangsu, China	450*2, 600*2 km.t./yr, - bn $ (planning)	Unknown	120.0161	32.16961
Tatneft, Tatarstan, Russia	390 km.t./yr, - bn $ (2025)	Unknown	49.07583	55.80523
Ulsan, South Korea - Hyosung Corp	300 km.t./yr, - bn $ (2019)	Unknown	129.3119	35.53917
Yangpu, Danzhou City, Hainan Province, China	300 km.t./yr, - bn $ (planning)	Unknown	109.1859	19.73524
Zhangjiagang, Jiangsu, China	600 km.t./yr, - bn $ (planning)	Unknown	120.5515	31.8783
Zhangzhou City, Fujian, China -SABIC	600 km.t./yr, 3.9 bn $ (2021)	Unknown	117.6496	24.50819
Zibo City, Shandong, China	250 km.t./yr, - bn $ (planning)	Unknown	118.0444	36.80478

Altamira, Mexico	400 km.t./yr, - bn $ (-)	Proposed but abandoned	-98.1165	22.56302
Cartagena, Colombia	300 km.t./yr, - bn $ (-)	Proposed but abandoned	-75.5271	10.41958
Ras Lafan Industrial Services Area, Al Khor and Al Thakhira, Qatar	750 km.t./yr, - bn $ (-)	Proposed but abandoned	51.41663	25.90836
Tarija, Bolivia	250 km.t./yr, - bn $ (-)	Proposed but abandoned	-63.8333	-21.5833

1.4 List of Patents

Table S6. List of most relevant dehydrogenation (DH), metathesis (Met) and DH-Met patents.

Dehydrogenation					
Title	**Year**	**Inventor**	**Assignee**	**Number**	**Reference**
Group viii noble metal, tin and solid inorganic refractory metal oxide catalyst composites and their use in hydrocarbon dehydrogenations	1968	Everett Clippinger, Bernard F Mulaskey	Chevron Research and Technology Co	US3531543A	[6]
Dehydrogenation with a catalytic composite containing platinum, rhenium and tin	1970	Richard E Rausch	Universal Oil Products Co	US3584060	[7]
Multicomponent dehydrogenation	1973	John C Hayes	Honeywell UOP LLC	US3878131	[8]

catalyst					
Reduction of metal surface-initiated cracking in dehydrogenation reactors	1979	William M. Castor, Barbara S. Taylor	Dow Chemical Co	US4132743	[9]
Dehydrogenation of hydrocarbons with a halogen contacting step	1983	Tamotsu Imai, Chi-Wen Hung	Honeywell UOP LLC	US4438288	[10]
Dehydrogenation catalyst compositions and method of their preparation	1984	Tamotsu Imai, Hayim Abrevaya	Honeywell UOP LLC	US4595673	[11]
Dehydrogenation catalyst composition	1987	Tamotsu Imai	Honeywell UOP LLC	US4716143	[12]
Dehydrogenation catalyst composition and hydrocarbon dehydrogenation process	1988	Tamotsu Imai, Hayim Abrevaya, Jeffery C. Bricker, Deng-Yang Jan	Honeywell UOP LLC	US4827072	[13]
Dehydrogenation of isobutane over a zeolitic catalyst	1988	Stephen J. Miller	Chevron Research and Technology Co	US4727216	[14]
Dehydrogenation process	1989	Dwight L. McKay, Michael E. Olbrich	Phillips Petroleum Company, now ThyssenKrupp Industrial Solutions AG	US4902849	[15]
Dehydrogenation process	1989	Michael E. Olbrich, Dwight L.	Phillips Petroleum Company	US4926005	[16]

		McKay, B. Jean			
Dehydrogenation process, equipment and catalyst loads therefor	1994	John W. Heyse, Paul G. Johnson, Bernard F. Mulaskey	Chevron Chemical Company	USOO5723707A	[17]
Paraffin dehydrogenation catalyst	2003	Saeed Alerasool, Harold Manning	SABIC Global Technologies BV	US20030232720A1	[18]
New Catalyst for Higher Production Rates in Hydrocarbon Dehydrogenation	2010	Mark G. Riley, Bipin V. Vora	Honeywell UOP LLC	US20100240941A1	[19]
Alkane dehydrogenation catalysts	2010	Wolfgang Ruettinger, Michael J. Breen, Richard Jacubinas, Saeed Alerasool	SABIC Global Technologies BV	US20100312035A1	[20]
Dehydrogenation process with heat generating material	2014	Vladimir Fridman, Michael A. Urbancic	Clariant Corporation	US 2015/0259265 A1	[21]
Metathesis					
Title	**Year**	**Inventor**	**Assignee**	**Number**	**Reference**
Conversion of Olefins	1965	Louis F. Heckelsberg	Phillips Petroleum Company	US3365513	[22]
Olefin oligomerization	1968	Lawrence G. Cannel	Shell Oil Company	US3592869	[23]
Isoamylenes from butenes	1976	Roland E. Dixon, Douglas P.	Phillips Petroleum	US4085158	[24]

		Hann	Company		
DH-Met					
Title	**Year**	**Inventor**	**Assignee**	**Number**	**Reference**
Disproportionation of saturated hydro carbons employing a catalyst that comprises platinum and tungsten	1973	Robert L. Burnett	Chevron Research Company	US3856876	[25]
Combined dehydrogenation and disproportionation	1965	Louis F. Heckelsberg and Robert L. Banks	Phillips Petroleum Company	US3445541	[26]
Dual catalyst system for alkane metathesis	2005	Alan Goldman, Maurice Brookhart, Amy Roy, Ritu Ahuja, Zheng Huang	University of North Carolina at Chapel Hill Rutgers State University of New Jersey	US20070060781A1	[27]
A process for conversion of a hydrocarbon feed	2017	Kongkiat Suriye, Amnart Jantharasuk, Wuttithep Jareewatchara	SMH Co., Ltd.	WO/2017/001446	[28]
A hydrocarbon conversion catalyst for producing less saturated hydrocarbon product	2017	Kongkiat Suriye, Amnart Jantharasuk, Wuttithep Jareewatchara	SMH Co., Ltd.	WO/2017/001445	[29]
Catalyst system and process utilizing the catalyst system	2018	Kongkiat Suriye, Amnart Jantharasuk	SMH Co., Ltd.	WO/2018/108443	[30]

1.5 List of previous reviews

Table S7. List of previous reviews on propane dehydrogenation and olefin metathesis.

Dehydrogenation		
Title	**Year**	**Reference**
Alkane dehydrogenation over supported chromium oxide catalysts	1999	[31]
Advanced Catalytic Dehydrogenation Technologies for Production of Olefins	2012	[32]
Development of Dehydrogenation Catalysts and Processes	2012	[33]
Catalytic Dehydrogenation of Light Alkanes on Metals and Metal Oxides	2014	[34]
Molecular understandings on the activation of light hydrocarbons over heterogeneous catalysts	2015	[35]
Light alkane dehydrogenation to light olefin technologies: a comprehensive review	2015	[36]
Lower alkanes dehydrogenation: Strategies and reaction routes to corresponding alkenes	2016	[37]
State-of-the-art catalysts for direct dehydrogenation of propane to propylene	2019	[38]
Recent Progress in Commercial and Novel Catalysts for Catalytic Dehydrogenation of	2020	[39]

Light Alkanes		
The role of heat transfer on the feasibility of a packed-bed membrane reactor for propane dehydrogenation	2020	[40]
Propane dehydrogenation: catalyst development, new chemistry, and emerging technologies	2021	[41]
C–H bond activation in light alkanes: a theoretical perspective	2021	[42]
Metathesis		
Title	**Year**	**Reference**
Industrial applications of olefin metathesis	2004	[43]
From olefin to alkane metathesis: A historical point of view	2006	[44]
Olefin Metathesis by Supported Metal Oxide Catalysts	2014	[45]
Investigation of the preparation and catalytic activity of supported Mo, W, and Re oxides as heterogeneous catalysts in olefin metathesis	2016	[46]
Heterogeneous catalysts for gas-phase conversion of ethylene to higher olefins	2017	[47]
Olefin metathesis: what have we learned about homogeneous and heterogeneous catalysts from surface organometallic chemistry?	2021	[48]

2. Thermodynamics

Thermodynamics calculations have been performed using the software HSC 9.6.1, in the *Gem equilibrium composition* module for Gibbs free energy minimization and in the *Rea reaction equations* module for the calculation of ΔH, ΔS, ΔG and K.

2.1 Overall Thermodynamic Boundaries

The PTO reaction is affected by side reactions leading to formation of unwanted products such as paraffins and coke. When including all possible species in the thermodynamic calculations, one can gather some important information about trends in byproduct stability. Figure S2a reports the equilibrium amount when including all possible species in the PTO reaction (i.e. H_2, graphitic carbon, methane, ethane, ethylene, propane, propylene, butanes, butenes, benzene, toluene). Unsurprisingly, the dominating species are methane and graphitic carbon, whose formation is coupled with H_2 evolution. Notably, methane is the dominant species at temperatures lower than approx. 450 °C, while coke is favored at higher temperature. When coke is not considered in the calculation, aromatics take over as the second-most stable species (Figure S2b). These are indeed observed as a side product in the DH reactions, and are precursors to the formation of coke. Removing coke and methane from the picture, ethane formation boundaries can be observed (Figure S2c). Similarly to methane, ethane dominates at lower temperatures, giving yield to unsaturated products and hydrogen at higher temperatures. Altogether, these calculations show how kinetic control is pivotal in the PTO reaction, since the targeted olefins are less thermodynamically stable than unwanted byproducts, and they give an indication on the proper choice of reaction temperature in order to hinder the production of a certain byproduct.

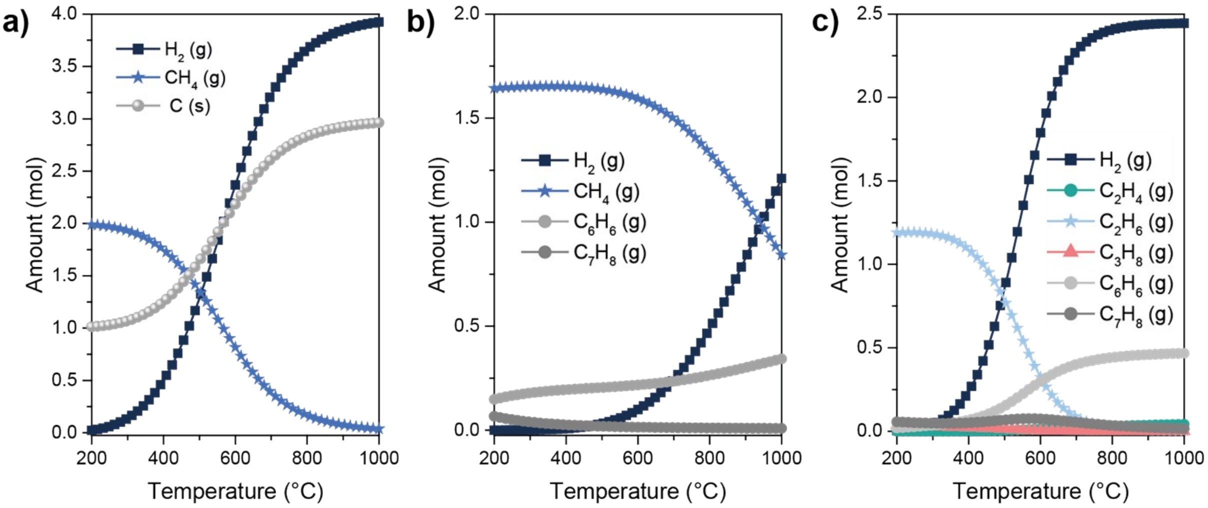

Figure S2: Equilibrium amount of species which can be produced during the PTO reaction, including: **a)** all main species (i.e. H_2, graphitic carbon, methane, ethane, ethylene, propane, propylene, butanes, butenes, benzene, toluene), **b)** all species but coke, and **c)** all species except for methane and coke. Equilibrium calculations were performed through HSC Chemistry 9.1 software.

2.2 PTO Stoichiometry-Restricted Calculations

The HSC 9 software provides powerful tools to calculate thermodynamics of complex systems using the Gibbs free energy minimization algorithm [49]. This method finds the free energy minimum of a given mixture of chemical species based on a very detailed and extensive database of thermodynamic properties. Nonetheless, the algorithm doesn't allow to consider just a certain reaction or subset of reactions which would entangle some products in a certain stoichiometric ratio. This can be observed in Figure 7b in the main text for the metathesis reaction: the Gibbs energy minimization differs from the stoichiometric equilibrium because the metathesis stoichiometry is disregarded. The stoichiometric amounts of ethylene, propylene and butenes at equilibrium can be calculated using the reaction equation module of HSC 9, which gives a value of K, the equilibrium constant, for each reaction temperature.

This is a trivial task when calculating single reactions equilibria, but in order to find the equilibrium conversion in the case of the sequential PTO reaction, one must solve a system of two equations, as follows:

Reaction 1		C_3H_8 (g)	↔	C_3H_6 (g)	+	H_2 (g)
	Initial	1		-		-
	Equilibrium	1-x		x-2y		x
Reaction 2		$2C_3H_6$ (g)	↔	C_2H_4 (g)	+	C_4H_8 (g)
	Initial	x		-		-
	Equilibrium	x-2y		y		y

The two reactions are subjected to the equilibrium constant K_1 and K_2, as follows:

$$K_1 = \frac{(x-2y)x}{1-x} \tag{1}$$

$$K_2 = \frac{y^2}{(x-2y)^2} \tag{2}$$

We can now solve the system of Equations 1 and 2 for each temperature (i.e. each set of K_1, K_2), with the boundaries $0 < x < 1$ and $0 < y < 0.5$, to finally be able to calculate the equilibrium amount of specie, selectivity and yield at the stoichiometric restricted equilibrium (Figure S3).

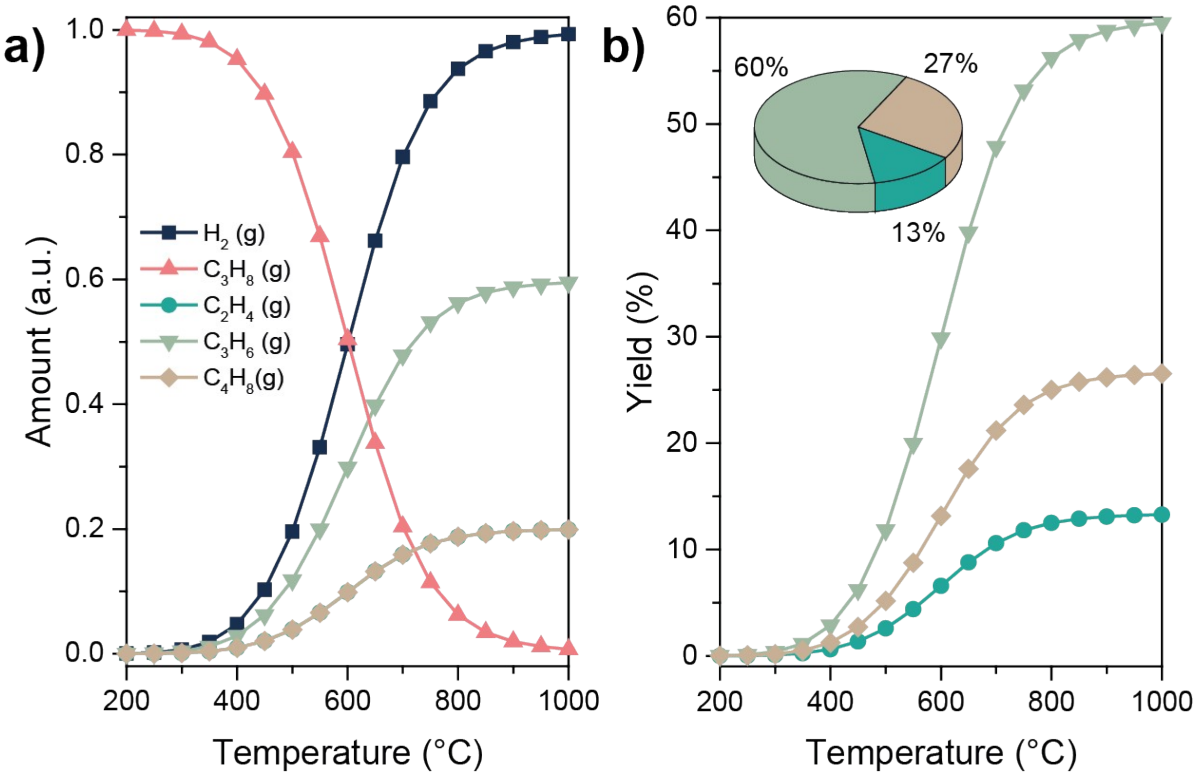

Figure S3: **a)** equilibrium amount of species in the PTO reaction, restricted to the stoichiometric amounts of the dehydrogenation and metathesis reactions as a function of temperature; **b)** Respective products yield vs. temperature. The inset shows the selectivity at 600°C. Selectivity only varies of ±1% in the considered temperature range. Equilibrium constants were calculated in the reaction equilibrium module of the HSC Chemistry 9.1 software.

2.3 Temperature Dependency of Reaction Enthalpy, Entropy, Free Energy and Equilibrium Constant for the Dehydrogenation, Metathesis and Side Reactions

Table S8. Summary of parameters at 600 °C for the main and side reactions involved in PTO. Note that all side reactions are thermodynamically favored (ΔG values) with respect to DH and Met.

Reaction	**ΔH (kJ mol^{-1})**	**ΔS (J K^{-1} mol^{-1})**	**ΔG (kJ mol^{-1})**	**K**	**Log K**
Propane Dehydrogenation	130.582	139.380	8.882	2.942E-001	-0.531
Propylene Metathesis (trans-2-butene)	0.704	-17.604	16.075	1.092E-001	-0.962
Propylene Metathesis (cis-2-butene)	2.198	-17.855	17.787	8.626E-002	-1.064
Propane Cracking	80.268	132.458	-35.388	1.310E+002	2.117
Propylene Hydrogenolysis	-50.314	-6.922	-44.270	4.453E+002	2.649
Butene Cracking	111.042	134.575	-6.462	2.436E+000	0.387

Butene Hydrogenolysis	-31.493	0.642	-32.054	8.274E+001	1.918
Propylene Coupling	59.715	162.540	-82.207	8.285E+004	4.918
Trans-2-butene Isomerization to 1-butene	8.515	5.883	3.378	6.279E-001	-0.202

2.3.1 Propane Dehydrogenation

Table S9. Temperature dependency of reaction parameters for the propane dehydrogenation reaction, calculated using the reaction equations module of HSC 9.1.

Reaction Equation

C3H8(g)=C3H6(PPYg) + H2(g)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
0.000	124.581	125.411	90.325	5.316E-018	-17.274
50.000	125.568	128.730	83.969	2.667E-014	-13.574
100.000	126.506	131.432	77.462	1.431E-011	-10.844
150.000	127.350	133.558	70.835	1.800E-009	-8.745
200.000	128.078	135.186	64.115	8.342E-008	-7.079
250.000	128.686	136.409	57.324	1.888E-006	-5.724
300.000	129.180	137.312	50.479	2.507E-005	-4.601
350.000	129.573	137.971	43.596	2.215E-004	-3.655
400.000	129.882	138.449	36.685	1.423E-003	-2.847
450.000	130.131	138.806	29.754	7.090E-003	-2.149
500.000	130.324	139.065	22.806	2.878E-002	-1.541
550.000	130.470	139.248	15.848	9.868E-002	-1.006
600.000	130.582	139.380	8.882	2.942E-001	-0.531
650.000	130.669	139.477	1.911	7.796E-001	-0.108
700.000	130.739	139.550	-5.065	1.870E+000	0.272
750.000	130.793	139.605	-12.044	4.120E+000	0.615
800.000	130.834	139.644	-19.025	8.435E+000	0.926
850.000	130.861	139.669	-26.008	1.621E+001	1.210
900.000	130.874	139.680	-32.992	2.945E+001	1.469
950.000	130.870	139.677	-39.976	5.097E+001	1.707
1000.000	130.847	139.658	-46.959	8.449E+001	1.927

Species Data

Formula	M g/mol	Conc. wt-%	Input Amounts mol	 g	 Volume	 Unit	Extrapolated From T(K)
C3H8(g)	44.096	100.000	1.000	44.096	22.414	l	-
C3H6(PPYg)	42.080	95.428	1.000	42.080	22.414	l	-
H2(g)	2.016	4.572	1.000	2.016	22.414	l	-

2.3.2 Metathesis to trans-2-butene

Table S10. Temperature dependency of reaction parameters for the propylene metathesis reaction to the thermodynamically favored trans-2-butene, calculated using the reaction equations module of HSC 9.1.

Reaction Equation

2C3H6(PPYg)= C2H4(g) + C4H8(T2Bg)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
0.000	0.587	-17.681	5.416	9.208E-002	-1.036
50.000	0.676	-17.377	6.291	9.616E-002	-1.017
100.000	0.634	-17.495	7.162	9.940E-002	-1.003
150.000	0.531	-17.753	8.043	1.016E-001	-0.993
200.000	0.441	-17.955	8.936	1.031E-001	-0.987
250.000	0.403	-18.032	9.836	1.042E-001	-0.982
300.000	0.408	-18.022	10.738	1.050E-001	-0.979
350.000	0.448	-17.957	11.638	1.058E-001	-0.976
400.000	0.506	-17.867	12.533	1.065E-001	-0.973
450.000	0.569	-17.777	13.424	1.072E-001	-0.970
500.000	0.627	-17.699	14.311	1.079E-001	-0.967
550.000	0.674	-17.640	15.194	1.086E-001	-0.964
600.000	0.704	-17.604	16.075	1.092E-001	-0.962
650.000	0.715	-17.592	16.955	1.098E-001	-0.959
700.000	0.704	-17.604	17.835	1.103E-001	-0.957
750.000	0.669	-17.638	18.716	1.108E-001	-0.956
800.000	0.611	-17.694	19.599	1.112E-001	-0.954
850.000	0.529	-17.768	20.486	1.115E-001	-0.953
900.000	0.423	-17.861	21.376	1.117E-001	-0.952
950.000	0.287	-17.974	22.272	1.119E-001	-0.951
1000.000	0.147	-18.087	23.174	1.120E-001	-0.951

Species Data

Formula	M	Conc.	Input Amounts				Extrapolated From T(K)
	g/mol	wt-%	mol	g	Volume	Unit	
C3H6(PPYg)	42.080	100.000	2.000	84.159	44.827	l	-
C2H4(g)	28.053	33.333	1.000	28.053	22.414	l	-
C4H8(T2Bg)	56.106	66.667	1.000	56.106	22.414	l	-

2.3.3 Metathesis to 2-cis-butene

Table S11. Temperature dependency of reaction parameters for the propylene metathesis reaction to the thermodynamically unfavored cis-2-butene, calculated using the reaction equations module of HSC 9.1.

Reaction Equation
2C3H6(PPYg)=C2H4(g)+C4H8(C2Bg)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
0.000	4.756	-11.923	8.013	2.935E-002	-1.532
50.000	4.444	-12.968	8.634	4.020E-002	-1.396
100.000	4.000	-14.241	9.314	4.966E-002	-1.304
150.000	3.534	-15.415	10.057	5.734E-002	-1.242
200.000	3.155	-16.265	10.850	6.339E-002	-1.198
250.000	2.905	-16.768	11.677	6.824E-002	-1.166
300.000	2.719	-17.108	12.524	7.219E-002	-1.142
350.000	2.592	-17.321	13.385	7.549E-002	-1.122
400.000	2.501	-17.462	14.255	7.830E-002	-1.106
450.000	2.427	-17.568	15.131	8.072E-002	-1.093
500.000	2.357	-17.661	16.012	8.282E-002	-1.082
550.000	2.283	-17.755	16.897	8.466E-002	-1.072
600.000	2.198	-17.855	17.787	8.626E-002	-1.064
650.000	2.098	-17.965	18.683	8.765E-002	-1.057
700.000	1.982	-18.088	19.584	8.886E-002	-1.051
750.000	1.848	-18.223	20.492	8.990E-002	-1.046
800.000	1.695	-18.368	21.407	9.077E-002	-1.042
850.000	1.525	-18.523	22.329	9.151E-002	-1.039
900.000	1.338	-18.686	23.259	9.211E-002	-1.036
950.000	1.127	-18.862	24.198	9.259E-002	-1.033
1000.000	0.913	-19.033	25.145	9.295E-002	-1.032

Species Data

Formula	M	Conc.	Input Amounts				Extrapolated From T(K)
	g/mol	wt-%	mol	g	Volume	Unit	
C3H6(PPYg)	42.080	100.000	2.000	84.159	44.827	l	-
C2H4(g)	28.053	33.333	1.000	28.053	22.414	l	-
C4H8(C2Bg)	56.106	66.667	1.000	56.106	22.414	l	-

2.3.4 Propane Cracking

Table S12. Temperature dependency of reaction parameters for propane cracking side reaction (propane giving methane and ethylene), calculated using the reaction equations module of HSC 9.1.

Reaction Equation

C3H8(g) = CH4(g) + C2H4(g)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
0.000	82.359	134.910	45.509	1.980E-009	-8.703
50.000	82.608	135.754	38.740	5.464E-007	-6.262
100.000	82.735	136.122	31.941	3.376E-005	-4.472
150.000	82.752	136.168	25.133	7.894E-004	-3.103
200.000	82.674	135.995	18.328	9.473E-003	-2.024
250.000	82.514	135.674	11.536	7.049E-002	-1.152
300.000	82.286	135.260	4.762	3.681E-001	-0.434
350.000	82.005	134.791	-1.989	1.468E+000	0.167
400.000	81.687	134.299	-8.717	4.747E+000	0.676
450.000	81.351	133.817	-15.419	1.300E+001	1.114
500.000	80.997	133.345	-22.099	3.113E+001	1.493
550.000	80.632	132.888	-28.754	6.681E+001	1.825
600.000	80.268	132.458	-35.388	1.310E+002	2.117
650.000	79.911	132.061	-42.001	2.381E+002	2.377
700.000	79.567	131.698	-48.594	4.060E+002	2.609
750.000	79.239	131.369	-55.171	6.559E+002	2.817
800.000	78.930	131.074	-61.732	1.012E+003	3.005
850.000	78.638	130.808	-68.279	1.499E+003	3.176
900.000	78.362	130.567	-74.813	2.145E+003	3.331
950.000	78.099	130.348	-81.336	2.977E+003	3.474
1000.000	77.848	130.147	-87.848	4.023E+003	3.605

Species Data

Formula	M g/mol	Conc. wt-%	Input Amounts mol	 g	 Volume	 Unit	Extrapolated From T(K)
C3H8(g)	44.096	100.000	1.000	44.096	22.414	l	-
CH4(g)	16.042	36.381	1.000	16.042	22.414	l	-
C2H4(g)	28.053	63.619	1.000	28.053	22.414	l	-

2.3.5 Propylene Hydrogenolysis

Table S13. Temperature dependency of reaction parameters for the propylene hydrogenolysis side reaction (propylene and hydrogen giving methane and ethylene), calculated using the reaction equations module of HSC 9.1.

Reaction Equation

C3H6(PPYg)+H2(g)=CH4(g)+C2H4(g)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K

0.000	-42.222	9.499	-44.817	3.724E+008	8.571
50.000	-42.960	7.024	-45.229	2.049E+007	7.312
100.000	-43.771	4.690	-45.521	2.359E+006	6.373
150.000	-44.598	2.610	-45.703	4.387E+005	5.642
200.000	-45.404	0.809	-45.787	1.136E+005	5.055
250.000	-46.172	-0.735	-45.788	3.734E+004	4.572
300.000	-46.894	-2.053	-45.717	1.468E+004	4.167
350.000	-47.568	-3.180	-45.586	6.630E+003	3.821
400.000	-48.195	-4.149	-45.402	3.337E+003	3.523
450.000	-48.780	-4.988	-45.173	1.833E+003	3.263
500.000	-49.327	-5.720	-44.905	1.082E+003	3.034
550.000	-49.838	-6.360	-44.603	6.770E+002	2.831
600.000	-50.314	-6.922	-44.270	4.453E+002	2.649
650.000	-50.758	-7.417	-43.912	3.054E+002	2.485
700.000	-51.171	-7.852	-43.530	2.171E+002	2.337
750.000	-51.554	-8.236	-43.127	1.592E+002	2.202
800.000	-51.904	-8.570	-42.707	1.199E+002	2.079
850.000	-52.224	-8.861	-42.271	9.248E+001	1.966
900.000	-52.513	-9.113	-41.821	7.282E+001	1.862
950.000	-52.771	-9.329	-41.360	5.840E+001	1.766
1000.000	-52.998	-9.511	-40.889	4.761E+001	1.678

Species Data

Formula	M g/mol	Conc. wt-%	Input Amounts mol	g	Volume	Unit	Extrapolated From T(K)
C3H6(PPYg)	42.080	95.428	1.000	42.080	22.414	l	-
H2(g)	2.016	4.572	1.000	2.016	22.414	l	-
CH4(g)	16.042	36.381	1.000	16.042	22.414	l	-
C2H4(g)	28.053	63.619	1.000	28.053	22.414	l	-

2.3.6 Butene Cracking

Table S14. Temperature dependency of reaction parameters for the butene cracking side reaction (butene to ethylene), calculated using the reaction equations module of HSC 9.1.

Reaction Equation

C4H8(2BTg) = 2C2H4(g)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
0.000	113.789	138.934	75.839	3.133E-015	-14.504
50.000	113.794	138.952	68.891	7.300E-012	-11.137
100.000	113.742	138.805	61.947	2.127E-009	-8.672
150.000	113.639	138.549	55.013	1.616E-007	-6.791
200.000	113.483	138.200	48.094	4.899E-006	-5.310

250.000	113.271	137.776	41.194	7.702E-005	-4.113
300.000	113.021	137.318	34.316	7.452E-004	-3.128
350.000	112.739	136.847	27.462	4.987E-003	-2.302
400.000	112.436	136.380	20.632	2.506E-002	-1.601
450.000	112.113	135.918	13.824	1.003E-001	-0.999
500.000	111.773	135.463	7.040	3.345E-001	-0.476
550.000	111.415	135.015	0.278	9.602E-001	-0.018
600.000	111.042	134.575	-6.462	2.436E+000	0.387
650.000	110.655	134.144	-13.180	5.569E+000	0.746
700.000	110.256	133.723	-19.876	1.167E+001	1.067
750.000	109.847	133.313	-26.552	2.268E+001	1.356
800.000	109.418	132.904	-33.208	4.135E+001	1.616
850.000	108.972	132.498	-39.843	7.131E+001	1.853
900.000	108.516	132.101	-46.458	1.171E+002	2.069
950.000	108.056	131.716	-53.053	1.844E+002	2.266
1000.000	107.594	131.347	-59.629	2.797E+002	2.447

Species Data

Formula	M g/mol	Conc. wt-%	Input Amounts mol	g	Volume	Unit	Extrapolated From T(K)
C4H8(2BTg)	56.106	100.000	1.000	56.106	22.414	l	-
C2H4(g)	28.053	100.000	2.000	56.106	44.827	l	-

2.3.7 Butene Hydrogenolysis

Table S15. Temperature dependency of reaction parameters for the butene hydrogenolysis side reaction (butene and hydrogen giving ethane and ethylene), calculated using the reaction equations module of HSC 9.1.

Reaction Equation

C4H8(2BTg) +H2(g)= C2H6(g) + C2H4(g)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
0.000	-21.805	20.242	-27.334	1.689E+005	5.228
50.000	-22.762	17.025	-28.264	3.707E+004	4.569
100.000	-23.705	14.311	-29.045	1.165E+004	4.066
150.000	-24.623	12.002	-29.702	4.642E+003	3.667
200.000	-25.522	9.994	-30.250	2.187E+003	3.340
250.000	-26.405	8.219	-30.705	1.164E+003	3.066
300.000	-27.258	6.662	-31.076	6.798E+002	2.832
350.000	-28.074	5.296	-31.374	4.267E+002	2.630
400.000	-28.845	4.105	-31.609	2.838E+002	2.453
450.000	-29.571	3.064	-31.787	1.978E+002	2.296
500.000	-30.252	2.154	-31.917	1.434E+002	2.157

550.000	-30.891	1.352	-32.004	1.074E+002	2.031
600.000	-31.493	0.642	-32.054	8.274E+001	1.918
650.000	-32.061	0.010	-32.070	6.528E+001	1.815
700.000	-32.596	-0.555	-32.056	5.257E+001	1.721
750.000	-33.101	-1.061	-32.015	4.311E+001	1.635
800.000	-33.586	-1.524	-31.951	3.592E+001	1.555
850.000	-34.053	-1.950	-31.864	3.034E+001	1.482
900.000	-34.498	-2.337	-31.756	2.595E+001	1.414
950.000	-34.916	-2.686	-31.631	2.243E+001	1.351
1000.000	-35.307	-2.999	-31.488	1.959E+001	1.292

Species Data

Formula	M	Conc.	Input Amounts				Extrapolated From T(K)
	g/mol	wt-%	mol	g	Volume	Unit	
C4H8(2BTg)	56.106	96.532	1.000	56.106	22.414	l	-
H2(g)	2.016	3.468	1.000	2.016	22.414	l	-
C2H6(g)	30.069	51.734	1.000	30.069	22.414	l	-
C2H4(g)	28.053	48.266	1.000	28.053	22.414	l	-

2.3.8 Propylene dimerization to benzene (coke initiation)

Table S16. Temperature dependency of reaction parameters for the propylene to benzene side reaction, calculated using the reaction equations module of HSC 9.1.

Reaction Equation

2C3H6(PPYg)=C6H6(BZEg)+3H2(g)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
0.000	41.179	124.367	7.208	4.183E-002	-1.379
50.000	43.170	131.057	0.819	7.372E-001	-0.132
100.000	45.106	136.630	-5.878	6.650E+000	0.823
150.000	46.957	141.286	-12.829	3.835E+001	1.584
200.000	48.727	145.242	-19.995	1.613E+002	2.208
250.000	50.416	148.638	-27.344	5.375E+002	2.730
300.000	52.024	151.573	-34.851	1.501E+003	3.176
350.000	53.545	154.120	-42.495	3.650E+003	3.562
400.000	54.978	156.332	-50.257	7.946E+003	3.900
450.000	56.316	158.250	-58.123	1.580E+004	4.199
500.000	57.555	159.908	-66.078	2.915E+004	4.465
550.000	58.689	161.330	-74.110	5.049E+004	4.703
600.000	59.715	162.540	-82.207	8.285E+004	4.918
650.000	60.624	163.554	-90.360	1.298E+005	5.113
700.000	61.413	164.387	-98.560	1.953E+005	5.291
750.000	62.076	165.051	-106.796	2.836E+005	5.453

800.000	62.606	165.558	-115.062	3.990E+005	5.601
850.000	63.015	165.930	-123.350	5.459E+005	5.737
900.000	63.351	166.223	-131.654	7.284E+005	5.862
950.000	63.616	166.444	-139.971	9.505E+005	5.978
1000.000	63.816	166.605	-148.297	1.216E+006	6.085

Species Data

Formula	M g/mol	Conc. wt-%	Input Amounts mol	g	Volume	Unit	Extrapolated From T(K)
C3H6(PPYg)	42.080	100.000	2.000	84.159	44.827	l	-
C6H6(BZEg)	78.112	92.814	1.000	78.112	22.414	l	-
H2(g)	2.016	7.186	3.000	6.048	67.241	l	-

2.3.9 Trans-2-butene isomerization to 1-butene

Table S17. Temperature dependency of reaction parameters for the isomerization of trans-2-butene to 1-butene, calculated using the reaction equations module of HSC 9.1.

Reaction Equation

C4H8(2BTg) = C4H8(1BTg)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
0.000	8.497	5.935	6.875	4.843E-002	-1.315
50.000	8.495	5.932	6.578	8.641E-002	-1.063
100.000	8.482	5.894	6.283	1.320E-001	-0.880
150.000	8.465	5.851	5.989	1.822E-001	-0.739
200.000	8.445	5.806	5.698	2.349E-001	-0.629
250.000	8.421	5.758	5.409	2.883E-001	-0.540
300.000	8.408	5.734	5.121	3.414E-001	-0.467
350.000	8.411	5.739	4.835	3.933E-001	-0.405
400.000	8.432	5.771	4.547	4.437E-001	-0.353
450.000	8.461	5.813	4.257	4.925E-001	-0.308
500.000	8.491	5.853	3.966	5.396E-001	-0.268
550.000	8.511	5.879	3.672	5.847E-001	-0.233
600.000	8.515	5.883	3.378	6.279E-001	-0.202
650.000	8.496	5.861	3.085	6.690E-001	-0.175
700.000	8.460	5.824	2.792	7.081E-001	-0.150
750.000	8.423	5.787	2.502	7.452E-001	-0.128
800.000	8.381	5.747	2.214	7.803E-001	-0.108
850.000	8.338	5.707	1.927	8.135E-001	-0.090
900.000	8.295	5.670	1.643	8.450E-001	-0.073
950.000	8.260	5.641	1.360	8.748E-001	-0.058
1000.000	8.234	5.620	1.079	9.031E-001	-0.044

Species Data

Formula	M g/mol	Conc. wt-%	Input Amounts mol	g	Volume	Unit	Extrapolated From T(K)
C4H8(2BTg)	56.106	100.000	1.000	56.106	22.414	l	-
C4H8(1BTg)	56.106	100.000	1.000	56.106	22.414	l	-

2.3.10 Copper oxide reduction

Copper oxide (supported on alumina) can be used as a heat-generating component in the endothermic dehydrogenation reaction, according to US patent 2015/0259265 A1 [21]. We here report some of the possible reactions producing Cu under reducing conditions typical of propane dehydrogenation, all showing negative Gibbs free energy.

Table S18. Temperature dependency of reaction parameters for the reduction of CuO(s) by H_2 or propane, calculated using the reaction equations module of HSC 9.1.

Reaction Equation

CuO(s)+H2(g) =Cu(s) +H2O(g)

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
200.000	-87.210	41.489	-106.841	6.251E+011	11.796
250.000	-88.048	39.805	-108.872	7.437E+010	10.871
300.000	-88.896	38.257	-110.823	1.261E+010	10.101
350.000	-89.748	36.833	-112.700	2.804E+009	9.448
400.000	-90.599	35.519	-114.508	7.697E+008	8.886
450.000	-91.446	34.305	-116.254	2.500E+008	8.398
500.000	-92.287	33.180	-117.940	9.307E+007	7.969
550.000	-93.120	32.137	-119.573	3.876E+007	7.588
600.000	-93.941	31.168	-121.155	1.772E+007	7.249
650.000	-94.748	30.269	-122.691	8.766E+006	6.943
700.000	-95.540	29.434	-124.183	4.637E+006	6.666
750.000	-96.313	28.659	-125.635	2.598E+006	6.415
800.000	-97.064	27.942	-127.050	1.530E+006	6.185
850.000	-97.791	27.280	-128.431	9.407E+005	5.973
900.000	-98.490	26.671	-129.779	6.011E+005	5.779
950.000	-99.157	26.114	-131.098	3.972E+005	5.599
1000.000	-99.792	25.605	-132.391	2.705E+005	5.432

Species Data

Formula	M g/mol	Conc. wt-%	Input Amounts mol	g	Volume	Unit	Extrapolated From T(K)
CuO(s)	79.545	97.528	1.000	79.545	12.606	ml	-
H2(g)	2.016	2.472	1.000	2.016	22.414	l	-
Cu(s)	63.546	77.912	1.000	63.546	7.092	ml	-

H2O(g)	18.015	22.088	1.000	18.015	22.414	l	-

Reaction Equation

$10CuO(s)+C_3H_8(g) = 10Cu(s) +3CO_2(g) +4H_2O(g)$

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
200.000	-476.946	1012.972	-956.233	3.757E+105	105.575
250.000	-480.410	1006.019	-1006.709	3.348E+100	100.525
300.000	-484.302	998.920	-1056.833	2.107E+096	96.324
350.000	-488.547	991.822	-1106.601	5.848E+092	92.767
400.000	-493.084	984.821	-1156.017	5.145E+089	89.711
450.000	-497.855	977.987	-1205.086	1.130E+087	87.053
500.000	-502.826	971.341	-1253.818	5.201E+084	84.716
550.000	-507.966	964.900	-1302.223	4.387E+082	82.642
600.000	-513.233	958.689	-1350.312	6.121E+080	80.787
650.000	-518.594	952.719	-1398.096	1.304E+079	79.115
700.000	-524.014	947.002	-1445.588	3.978E+077	77.600
750.000	-529.462	941.543	-1492.801	1.652E+076	76.218
800.000	-534.906	936.347	-1539.747	8.957E+074	74.952
850.000	-540.312	931.423	-1586.440	6.127E+073	73.787
900.000	-545.643	926.779	-1632.894	5.140E+072	72.711
950.000	-550.865	922.420	-1679.123	5.164E+071	71.713
1000.000	-555.947	918.347	-1725.141	6.093E+070	70.785

Species Data

Formula	M	Conc.	Input Amounts				Extrapolated From T(K)
	g/mol	wt-%	mol	g	Volume	Unit	
CuO(s)	79.545	94.748	10.000	795.454	126.062	ml	-
C3H8(g)	44.096	5.252	1.000	44.096	22.414	l	-
Cu(s)	63.546	75.691	10.000	635.460	70.922	ml	-
CO2(g)	44.010	15.726	3.000	132.029	67.241	l	-
H2O(g)	18.015	8.583	4.000	72.061	89.654	l	-

Reaction Equation

$3CuO(s)+C_3H_8(g) = 3Cu(s) +3CO(g) +4H_2(g)$

Reaction Data

T °C	ΔH kJ	ΔS J/K	ΔG kJ	K	Log K
200.000	253.826	840.702	-143.953	7.823E+015	15.893
250.000	254.997	843.064	-186.052	3.786E+018	18.578
300.000	255.704	844.361	-228.241	6.350E+020	20.803
350.000	256.002	844.864	-270.475	4.721E+022	22.674
400.000	255.943	844.776	-312.718	1.854E+024	24.268
450.000	255.580	844.258	-354.945	4.371E+025	25.641

500.000	254.944	843.410	-397.138	6.811E+026	26.833
550.000	254.061	842.304	-439.282	7.549E+027	27.878
600.000	252.960	841.008	-481.365	6.299E+028	28.799
650.000	251.673	839.575	-523.380	4.140E+029	29.617
700.000	250.222	838.044	-565.321	2.222E+030	30.347
750.000	248.625	836.445	-607.184	1.002E+031	31.001
800.000	246.904	834.804	-648.965	3.895E+031	31.590
850.000	245.077	833.140	-690.664	1.329E+032	32.124
900.000	243.162	831.472	-732.279	4.051E+032	32.608
950.000	241.177	829.815	-773.811	1.118E+033	33.048
1000.000	239.139	828.182	-815.261	2.826E+033	33.451

Species Data

Formula	M	Conc.	Input Amounts				Extrapolated From T(K)
	g/mol	wt-%	mol	g	Volume	Unit	
CuO(s)	79.545	84.404	3.000	238.636	37.819	ml	-
C3H8(g)	44.096	15.596	1.000	44.096	22.414	l	-
Cu(s)	63.546	67.427	3.000	190.638	21.277	ml	-
CO(g)	28.010	29.721	3.000	84.030	67.241	l	-
H2(g)	2.016	2.852	4.000	8.064	89.654	l	-

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