

Accelerating adoption rates of chemical recycling

Evaluation of collaborative efforts towards the development of technological solutions for facilitating commercial-scale pyrolysis oil upgrading through an FCC unit

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Global plastics waste volumes are expected to increase from 260 million tons per year (mmtpy) in 2016 to 460 mmtpy per year by 2030, possibly resulting in important environmental concerns.¹ Only 16% of plastics waste is collected for recycling, while the majority is sent to incineration (25%) and landfills (40%) or lost as unmanaged dumps (19%). Plastics waste going to landfill and incineration is ultimately lost as a resource and cannot be reused or recycled. Reusing plastics can influence cost savings and reduce carbon emissions for plastics production.¹ Mechanical recycling is often considered an optimal solution for recycling plastics. However, its reach is constrained, especially when dealing with plastics that are mixed or contaminated with unwanted substances like metals, paper, other types of polymers or fillers.

Chemical recycling has been shown to be a valuable alternative to transform waste plastic into its monomeric forms or other valuable products when mechanical recycling is not an option.² Chemical recycling via pyrolysis uses thermochemical conversion of recycled plastics in the absence of oxygen – often referred to as plastic pyrolysis.³ The to-be recycled plastics are introduced into a pyrolysis reactor (which can be purely thermal or catalytic) to produce a range of products from monomers to intermediate products (oils).

The pyrolysis oil market has been growing in recent years with great interest from refiners and chemical manufacturers. This growth is further accelerated when different organisations, companies, and research organisations collaborate as technology allies to further develop and share knowledge. In this collaboration, Quantafuel worked with BASF to select an optimal catalyst for its catalytic pyrolysis process, providing the flexibility to tailor the oil boiling point range of the resulting product, depending on end-user needs. In this case, BASF and Quantafuel made a decision to evaluate a full boiling point range pyrolysis oil with little to no pre-distillation. BASF then conducted in-depth analyses and further crackability studies.

The in-depth study was divided into two parts with distinct objectives. The first part evaluated the upgrading of pyrolysis oil in a fluid catalytic cracking (FCC) context at a 10% co-processing level. This study was designed to demonstrate what a commercial FCC might do in terms

of a co-processing trial. Since refiners often do not elect to go for 100% recycled oil right away, they often do step tests from 1-20% before increasing to higher levels. Thus, 10% pyrolysis oil mixed with 90% vacuum gas oil (VGO) is representative of an FCC trial. The resulting products from such an FCC trial include sustainable transportation fuels (for example, naphtha) and LPG olefins.

In the second part of this study, the pyroil was processed at 100%, while using different catalyst designs to produce more sustainable chemicals, including BTX (benzene, toluene, and xylenes), propylene, and butylenes. This study was designed to be more applicable to a chemicals producer.

Experimental

Catalytic measurements were made using a laboratory-scale fluid bed cracking reactor. Cracking severity was varied to target different products by adjusting both reaction temperature (550°C and 600°C) and catalyst-to-oil ratio (4-to-6). For product evaluations, the naphtha product was

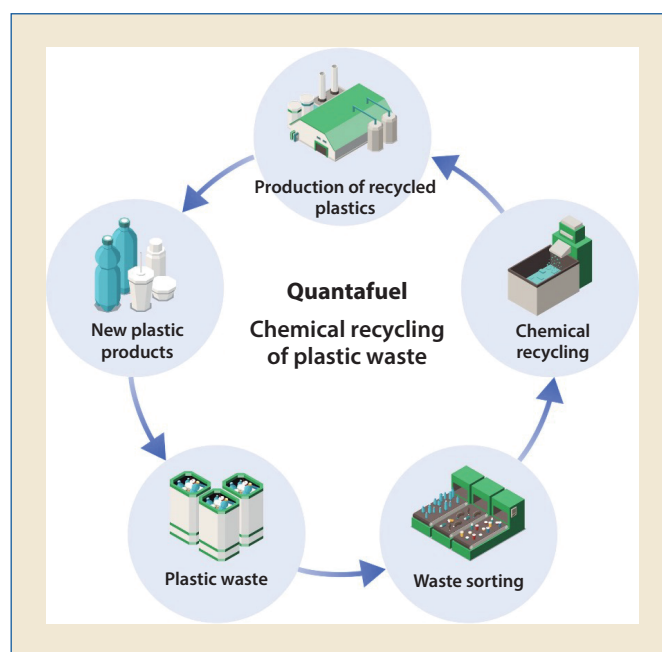


Figure 1 Quantafuel's chemical recycling process of waste plastics

Physical and chemical properties of plastic pyoil and VGO feedstocks		
	Plastic pyoil	VGO
Physical properties		
API (S.G.)	42.7 (0.812)	24.5 (0.907)
K factor	12.7	11.8
Refractive index	1.4527	1.5048
Pour point, °C	46	37
Flash point, °C	38	149
Conradson carbon, wt%	0.32	0.26
Total nitrogen, ppmw	447	998
Total sulphur, wt%	<0.01	0.76
Elemental analyses		
Al, ppmw	0.5	n.a.
Br, ppmw	4.79	n.a.
Cl, ppmw	45.2	n.a.
Ca, ppmw	<0.1	n.a.
Cu, ppmw	0.1	0.1
Fe, ppmw	0.29	4.5
K, ppmw	<0.1	n.a.
Mg, ppmw	<0.1	n.a.
Na, ppmw	<0.1	2.8
Ni, ppmw	<0.1	0.1
P, ppmw	5	n.a.
Si, ppmw	10	n.a.
Ti, ppmw	<0.1	n.a.
V, ppmw	<0.1	0.3
Distillation (SimDis)		
IBP, °C	78.3	130.6
5%, °C	120.0	304.4
10%, °C	147.8	328.9
20%, °C	199.4	357.2
30%, °C	240.6	378.9
50%, °C	333.3	418.9
75%, °C	444.4	472.2
90%, °C	526.7	513.9
95%, °C	566.7	537.8
FBP, °C	648.9	600.0

Table 1

defined as C₅ to 232°C, light cycle oil (LCO) as 232°C–360°C, and heavy cycle oil (HCO or bottoms) as 360°C and higher. Lumped product yields were measured using a combination of gas chromatography and liquid chromatography. Additionally, the speciation of the liquid products was determined using a mixture of liquid chromatography and mass spectrometry.

Pyrolysis oil made from polyolefins (~85% LDPE and ~15% PP) was obtained from the industrial plant of Quantafuel in Skive (Denmark) and used without further pre-processing. In addition to this pyrolysis oil, a standard vacuum gasoil (VGO) feed was also used in the first part of this study. The physical and chemical properties of both feeds are shown in **Table 1**.

Generally, plastic pyrolysis oil is lighter than VGO feed. The contaminants profile is also quite different: the pyrolysis oil contains noticeable amounts of halides, phosphorous, and silica, but less iron and sodium. This pyoil appears rather clean despite a significant level of chlorides. Its effect

in an FCCU, especially nickel reactivation, cannot be evaluated from this study because no metals (nickel) deactivation was applied, and lab testing does not leave enough time for chlorides to reactivate nickel even if it were present.

Refining applications

Two catalyst designs were tested for upgrading two different feeds: (1) 100% VGO and (2) 10% pyoil blended with 90% VGO:

- FCC(a) catalyst was oriented towards transportation fuels maximisation.
- FCC(b) catalyst was oriented towards LPG olefins maximisation.

FCC(a) catalyst was designed with high rare earth content on zeolite for maximising conversion and naphtha yield. FCC(b) catalyst was designed with low rare earth on zeolite to minimise hydrogen transfer reactions for maximising olefins and additional functionality to convert naphtha-range olefins to LPG olefins. FCC(a) and FCC(b) catalysts were steam-deactivated at 788°C for 24 hours before being tested:

- Steam-deactivated FCC(a) catalyst: TSA = 224 m²/g and rare earth oxide (REO) = 5.5 wt%.
- Steam-deactivated FCC(b) catalyst: TSA = 173 m²/g and REO = 0.7 wt%.

To examine the impact of the feed, the cracking evaluations were examined at iso-conversion (72 wt% chosen to eliminate the need for extrapolation), obtained by regression of raw data for both catalysts. In the refinery industry, the conversion to naphtha and lighter products is typically expressed as 100 – LCO – bottoms. Unsurprisingly, FCC(a) with higher rare earth on zeolite induced higher activity compared to FCC(b) for both feeds tested. Co-processing 10% of this pyoil helps to further increase valuable products due to already converted material (for example, naphtha range molecules) and better K-factor entering as co-feed.

As a reminder, K-factor is a correlation using density and distillation to determine the paraffinicity or aromaticity character of a feed and thus, the overall crackability in the FCC. It is often stated that a feed with a K-factor of 13.0 is purely paraffinic, while a feed with a K-factor of 10.0 is purely aromatic. This pyoil, with a K-factor of 12.7, suggests paraffinic behaviour and higher crackability compared to the VGO used in this study. Hence, this pyoil can also contribute as a hydrogen donor to the VGO to further increase its crackability and produce more valuable products, such as naphtha and LPG. FCC(a), promoting hydrogen transfer reactions (hydrogen consuming reactions), leads to lower hydrogen selectivity compared to FCC(b).

Referring to **Table 2**, blending 10% pyoil helps produce lower coke selectivity for both catalysts due to a higher H/C ratio and, hence, lower coke precursors present in the blend. Unsurprisingly, FCC(b) based on low REO on zeolite with an additional olefins-generating functionality is more selective to light olefins (such as C₂=, C₃=, and C₄=) and LPG compared to FCC(a), which is more oriented towards naphtha selectivity. Furthermore, blending 10% pyoil further improves the propylene, C₄ olefins, and LPG selectivities for both catalysts compared to 100% VGO. At iso-conversion,

**Catalytic cracking evaluation of plastic pyoil and VGO at 550°C with catalyst
FCC(a) and FCC(b) at iso-conversion (72 wt%)**

	FCC(a)		FCC(b)	
	100% VGO	10% Pyoil	100% VGO	10% Pyoil
Cat-to-oil, wt/wt	4.0	3.5	4.9	4.4
Hydrocarbon yields				
H ₂ , wt%	0.06	0.05	0.13	0.11
Methane, wt%	0.98	0.79	0.86	0.67
Ethane, wt%	0.62	0.53	0.71	0.58
Ethylene, wt%	0.84	0.73	4.19	5.01
Propane, wt%	1.11	0.99	2.62	2.95
Propylene, wt%	5.40	5.52	14.28	15.70
n-Butane, wt%	0.88	0.83	1.08	1.22
i-Butane, wt%	3.59	3.50	2.97	2.93
n-Butenes, wt%	4.95	5.26	6.75	7.35
i-Butylene, wt%	2.20	2.35	4.40	4.84
Naphtha, wt%	49.15	49.70	31.99	29.03
LCO, wt%	16.32	16.22	16.67	15.95
Bottoms, wt%	11.68	11.78	11.33	12.05
Coke, wt%	2.21	1.75	2.03	1.63
Calculated values				
Total valuable liquids*, wt%	83.60	84.36	80.75	79.95
Total dry gas, wt%	2.51	2.10	5.89	6.37
Total LPG, wt%	18.13	18.45	32.10	34.97
LPG olefinicity, wt/wt	0.69	0.71	0.79	0.80
Total C ₄ =, wt%	7.14	7.61	11.15	12.18
C ₃ olefinicity, wt/wt	0.83	0.85	0.84	0.84
C ₄ olefinicity, wt/wt	0.62	0.64	0.73	0.75

*Total valuable liquids defined as LPG + naphtha + LCO

Table 2

FCC(b) with 10% pyoil produced fewer valuable liquids (LPG + naphtha + LCO) compared to 100% VGO.

In fact, the olefin-inducing characteristics of FCC(b) converted some naphtha-range olefins, already present or formed after cracking reactions, to ethylene. For FCC(a), the difference in terms of LCO and bottoms yields is rather marginal between both feeds, while the difference is more

noticeable for FCC(b). In fact, this might be explained by the presence of bottom precursors in this pyoil that cannot be easily cracked in the range of 72 wt% conversion, but it can be improved at higher conversion levels with FCC(b). More valuable products, including naphtha and LPG, can be further produced as the ratio of pyoil increases because of a higher H/C ratio and better crackability.

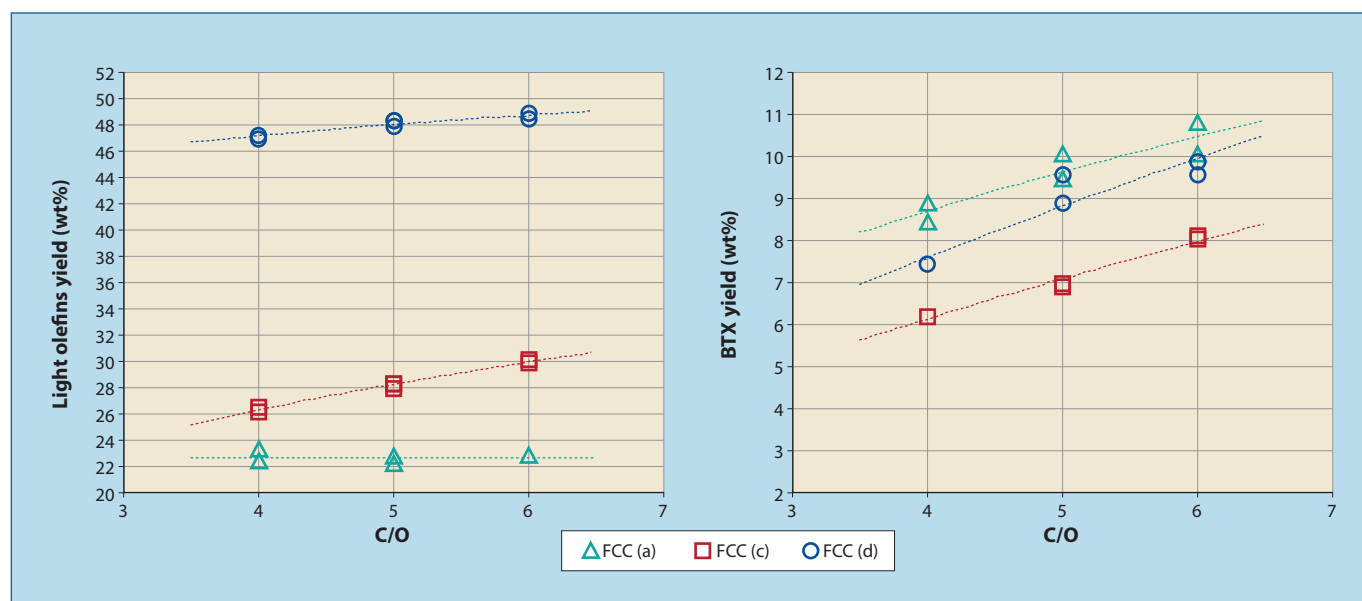


Figure 2 Impact of catalysts on light olefins and BTX yields by upgrading 100% of plastic pyoil at 550°C

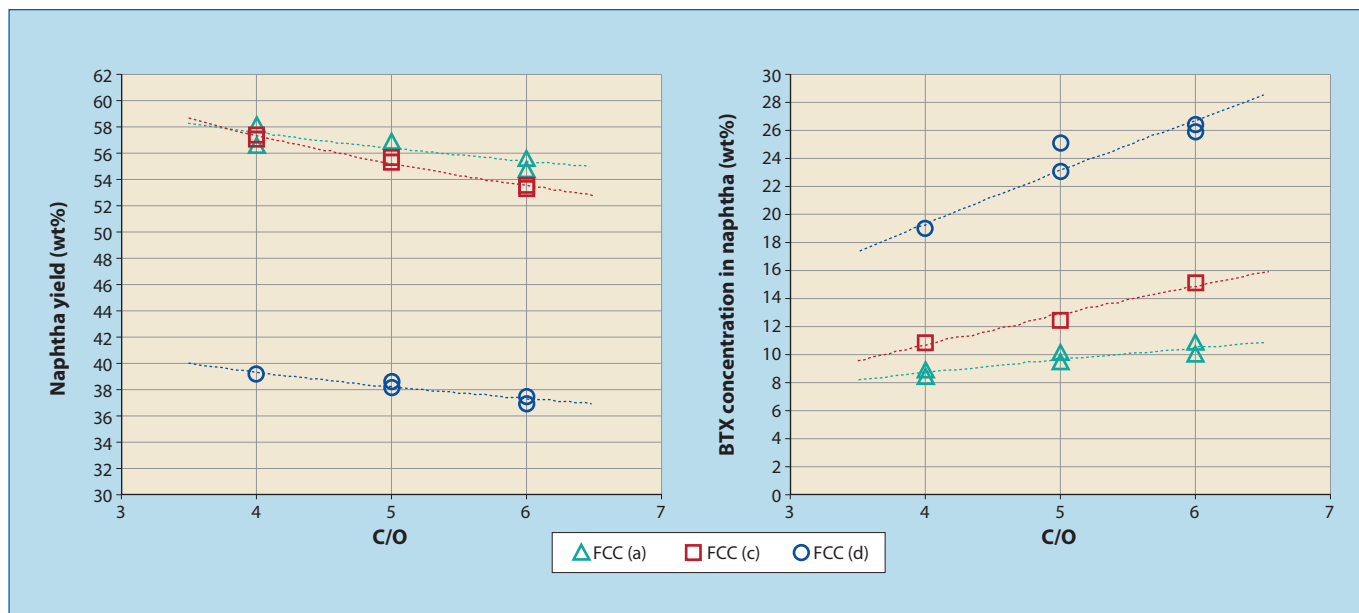


Figure 3 Impact of catalysts on naphtha yield and BTX concentration in naphtha by upgrading 100% of plastic pyoil at 550°C

Chemical applications

Three catalyst designs were tested for upgrading 100% of plastic pyoil to understand the maximisation of chemicals, including both light olefins and BTX (see **Figure 2**):

- FCC(a) catalyst was designed with high REO on zeolite – the same one used for maximising naphtha in the first part of the study.
- FCC(c) catalyst was designed with low REO on zeolite.
- FCC(d) catalyst designed with low REO on zeolite and an additional functionality to convert naphtha-range olefins to LPG olefins.

The purpose of this second part of the study was to understand how hydrogen transfer reactions and operating conditions (for example, C/O and temperature) can influence the promotion of chemicals, including BTX and light olefins while incorporating an olefins functionality into the

catalyst. FCC(a), FCC(c), and FCC(d) catalysts were steam deactivated at 788°C for 24 hours before being tested at 550°C and 600°C:

- Steam-deactivated FCC(a) catalyst: TSA = 224 m²/g and REO = 5.5 wt%.
- Steam-deactivated FCC(b) catalyst: TSA = 187 m²/g and REO = 0.9 wt%.
- Steam-deactivated FCC(d) catalyst: TSA = 172 m²/g and REO = 0.4 wt%.

The cracking evaluations were examined at iso-conversion, namely 93 wt%, and at 550°C (see **Table 3**). High conversion levels were observed with all the catalysts while upgrading this pyoil without any blending with fossil feedstock. This can be explained by the presence of 27% as naphtha in this pyoil as well as its paraffinic behaviour resulting in high crackability. FCC(c), with low REO on

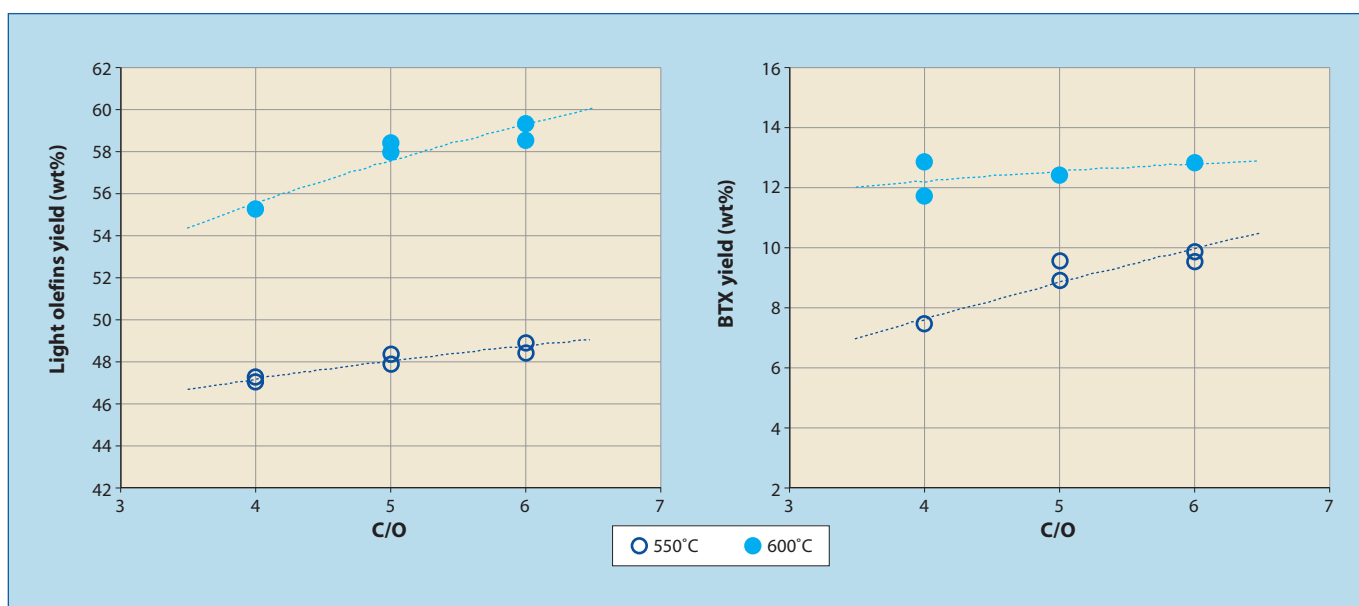


Figure 4 Impact of operating conditions (C/O and operating temperature) on light olefins and BTX by upgrading 100% of plastic pyoil over catalyst FCC(d)

zeolite, allows for achieving higher LPG olefinicity due to lower hydrogen transfer reactions and consequently higher olefins preservation compared to FCC(a).

FCC(d), with its olefins functionality, allows a further boost to the selectivity of light olefins. FCC(a) promotes hydrogen transfer reactions resulting in higher activity and naphtha and coke yields compared to FCC(c). The olefin functionality in FCC(d) can help to directly convert precursors already present in this pyoil to light olefins (such as C₂=, C₃=, and C₄=), resulting in a lower coke yield compared to FCC(a) and FCC(c). FCC(d) shows fewer valuable liquids due to its higher selectivity to ethylene compared to FCC(c). FCC(d) is fully oriented towards light olefins production and exhibits lower naphtha selectivity compared to FCC(a) and FCC(c) due to its combination of low REO on zeolite and olefins functionality. The selectivity towards light olefins could be further boosted while operating FCC(d) at higher severity, such as higher cat-to-oil and/or higher temperature.

BTX yields were also assessed with all three catalysts. The BTX yields were calculated according to the naphtha yield multiplied by the concentration of BTX in naphtha measured by detailed hydrocarbon analysis (DHA) illustrated in **Figure 3**. This shows how the yield of light olefins follows the expected trend, where the combination of low REO on zeolite and additional functionality (FCC-d) maximises light olefins, while high REO on zeolite (FCC-a) favours H-transfer reactions, minimising light olefins.

Regarding the yield of BTX, **Figure 3** highlights different pathways to aromatic formation. In the case of FCC(a), the higher hydrogen transfer rate promoting the formation of naphtha paraffins also increases the rate of alkane dehydrocyclisation, resulting in the highest overall yield of aromatics. In the case of FCC(d), promoting higher yields of light olefins also promotes aromatic formation through oligomerisation of light olefins. The formation of olefins is at the expense of naphtha, resulting in the highest concentration of aromatics in naphtha, as illustrated in **Figure 4**.

The FCC(d) formulation, combining both low REO on zeolite and an olefins functionality, allows for maximising light olefins and BTX concentration in naphtha. The impact of the operating conditions, including cat-to-oil and operating temperature, on the light olefins and BTX yields was also investigated by focusing on FCC(d). As per **Figure 4**, both light olefins and BTX yields can be further enhanced as the cat-to-oil and/or the operating temperature increases. The light olefins yield at 600°C, and a cat-to-oil C/O of 6.0 is rather significant, namely about 59 wt% as C₂=, C₃=, and C₄=, and can be further boosted by using higher severities, such as higher cat-to-oil (for example, >6.0) and/or higher operating temperature (for example, >600°C).

Conclusion

Plastic recycling has gained attention in the circular economy due to its potential for making the plastics lifecycle more sustainable. As there are challenges to the mechanical recycling of all plastics waste, chemical recycling has been considered a promising alternative as a means to increase the rate of global recycling.

Quantafuel produces high-quality pyrolysis from

Catalytic cracking evaluation of plastic pyoil at 550°C with FCC(a), FCC(c), and FCC(d) at iso-conversion (93 wt%)

	FCC(a)	FCC(c)	FCC(d)
Cat-to-oil, wt/wt	5.4	6.3	4.3
Hydrocarbon yields			
H ₂ , wt%	0.04	0.06	0.03
Methane, wt%	0.89	0.75	0.28
Ethane, wt%	0.52	0.51	0.27
Ethylene, wt%	0.97	1.17	4.34
Propane, wt%	1.94	1.26	2.04
Propylene, wt%	9.45	13.04	22.28
n-Butane, wt%	1.53	0.92	1.01
i-Butane, wt%	6.45	4.59	2.18
n-Butenes, wt%	8.42	10.42	12.53
i-Butylene, wt%	3.84	5.89	8.27
Naphtha, wt%	56.12	52.87	39.09
LCO, wt%	5.73	5.93	5.84
Bottoms, wt%	1.27	1.07	1.16
Coke, wt%	2.85	1.52	0.69
Calculated values			
Total valuable liquids*, wt%	93.46	94.91	93.24
Total dry gas, wt%	2.42	2.49	4.91
Total LPG, wt%	31.62	36.12	48.31
LPG olefinicity, wt/wt	0.69	0.81	0.89
Total C ₄ =, wt%	12.26	16.31	20.80
C ₃ olefinicity, wt/wt	0.83	0.91	0.92
C ₄ olefinicity, wt/wt	0.61	0.75	0.87
Total light olefins, wt%	22.68	30.53	47.42

*Total valuable liquids defined as LPG + gasoline + LCO

Table 3

polyolefins plastics waste, which can be considered in lieu of fossil-based feedstocks, either as co-feeding or standalone alternative feeds to be further cracked in FCC units to produce lower carbon intensity finished products.

It has been demonstrated that the FCC catalyst can be fine-tuned to produce more sustainable transportation fuels or chemicals via typical co-processing applications for refiners. The upgrading of this type of pyoil also appears to be a very efficient alternative to produce sustainable chemicals, such as light olefins and aromatics, for units operating at high severities to produce petrochemical feedstocks.

References

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Olav Stadaas joined Quantafuel in 2018 to support the plastic-to-liquid initiatives from pilot plants to full-scale operational plants, followed by close cooperation with BASF since 2019. Stadaas earned an MSC in mechanical engineering and has broad experience as a manager in small and larger companies within technology and business development processes.

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Niklas Jakobsson has advanced through several positions from R&D program manager through business development and business segment management at Haldor Topsoe A/S to become partner and co-founder of ChemAcc AB, Kagerod, Sweden. He is the author of several scientific papers within catalysis and absorption technology. He has been granted numerous patents within the fields of catalysis and process technology. Jakobsson holds a doctorate in chemical engineering from Lund University.

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