# Introduction

This document provides supplemental information for ARPA-E's exploratory research program, "Recycle Underutilized Solids to Energy" (REUSE). The goal is to provide additional technical information to prospective Applicants. Further information is available in a blog interview and webinar at https://arpa-e.energy.gov/?q=news-item/trash-treasure-reuse-creates-feedstock-plastic-waste

Award information, submission requirements, evaluation criteria, and other applicable information is provided in Funding Opportunity Announcements DE-FOA-0001953 and DE-FOA-0001954 (SBIR/STTR). REUSE-specific requirements are provided in Topic K of those FOAs., and is available at https://arpa-e-foa.energy.gov/Default.aspx?Search=0001953&SearchType=#FoaIde8647d89-1cac-4b58-8622-1b04de8958c4.

Under the REUSE Topic, ARPA-E seeks to fund the development of technologies to convert high-energy materials currently going to landfills to a high-energy content liquid product capable of displacing energy imports used for fuel or chemical production. The high-energy materials include plastics (#1-7 polymers, rubber, and composites) and paper. As discussed below, we estimate 30-34 MM ton plastic, 2-6 MM ton rubber, 18 MM ton of paper, and up to 0.1 MM ton composites are potentially available annually for this purpose. These numbers may be conservative based on changes in the plastic and paper export markets.

ARPA-E anticipates deployment of multiple low-cost, simple, flexible, small-scale (100-500 ton per day) regional facilities using modular plants. This scale is consistent with the sources for high-energy materials, which include ~300 Material Recovery Facilities and industrial waste sources. The assumption is that such facilities can be more economical than the paradigm of large-scale facilities making purity products, due to cost for transporting and aggregating waste and the high operating costs (OPEX) and capital cost (CAPEX) for product purification. This document gives a brief technical review for multiple potential process technologies. The review is not intended to be comprehensive or limiting, only to provide an introduction to potential Applicants.

# Plastic Feeds

Plastics (#1-7 polymers, rubber, and composites) are attractive due to their volume, high energy content, and an existing infrastructure for collecting, sorting, grading, and marketing them. The amount of plastics sent to landfills has recently increased because supply exceeds demand; quality of the materials is either too low or variable; logistics make it uneconomical to transport waste; and/or technology to recycle material is too expensive or does not exist.

Plastics sent to landfills come from diverse sources, and estimates for the amounts vary. There are two categories – pre-consumer and post-consumer. There is relatively little data on US "pre-consumer" industrial plastic waste. European experience shows that production of pre-consumer plastic is about 30% of post-consumer plastic.[1] Pre-consumer plastic recycling rates are significantly higher than post-consumer waste, but 5-20% is still landfilled,[4,2] suggesting that approximately 1-2 MM ton/yr pre-consumer plastic is landfilled in the US.

The largest source of post-consumer plastic waste is municipal solid waste (MSW), which is collected from residential and commercial sites. The Environmental Protection Agency (EPA) estimated 34.8 MM tons of plastic in MSW, of which 26.3 MM tons were landfilled, in 2017.[3] Table 1 provides the EPA's summary of plastic waste generation and fate by type. Note that the "Recycled" category includes domestically recycled materials and exported materials.

**Table 1 Plastic Waste Generation and Fate, 2017 (EPA)**[3]

| Product Category | Generation | Recycled | | Combusted with energy Recovery | Landfilled |
|---|---|---|---|---|---|
| | (Thousand tons) | (Thousand tons) | (Percent of generation) | (Thousand tons) | (Thousand tons) |
| **Total Plastics in MSW, by resin** | | | | | |
| PET | 5,020 | 930 | 18.5% | | |
| HDPE | 6,010 | 620 | 10.3% | | |
| PVC | 930 | Neg. | | | |
| LDPE/LLDPE | 7,930 | 480 | 6.1% | | |
| PLA | 90 | Neg. | | | |
| PP | 7,800 | 70 | 1.0% | | |
| PS | 2,350 | 30 | 1.3% | | |
| Other resins | 4,740 | 1,110 | 23.4% | | |
| ***Total Plastics in MSW*** | ***34,870*** | ***3,240*** | ***9.3%*** | ***5,340*** | ***26,290*** |

‡ Nondurable goods other than containers and packaging.

§ Due to source data aggregation, PET cups are included in "Other Plastic Packaging".

* All other nondurables include plastics in disposable diapers, clothing, footwear, etc.

** Injection stretch blow molded PET containers as identified in *Report on Postconsumer PET Container Recycling Activity in 2016.* National Association for PET Container Resources. Recycling includes caps, lids and other material collected with PET bottles and jars.

† White translucent homopolymer bottles as defined in the *2016 United States National Postconsumer Plastics Bottles Recycling Report*. American Chemistry Council and the Association of Postconsumer Plastic Recyclers.

Neg. = negligible, less than 5,000 tons

HDPE = High density polyethylene | PET = Polyethylene terephthalate | PS = Polystyrene

LDPE = Low density polyethylene | PP = Polypropylene | PVC = Polyvinyl chloride

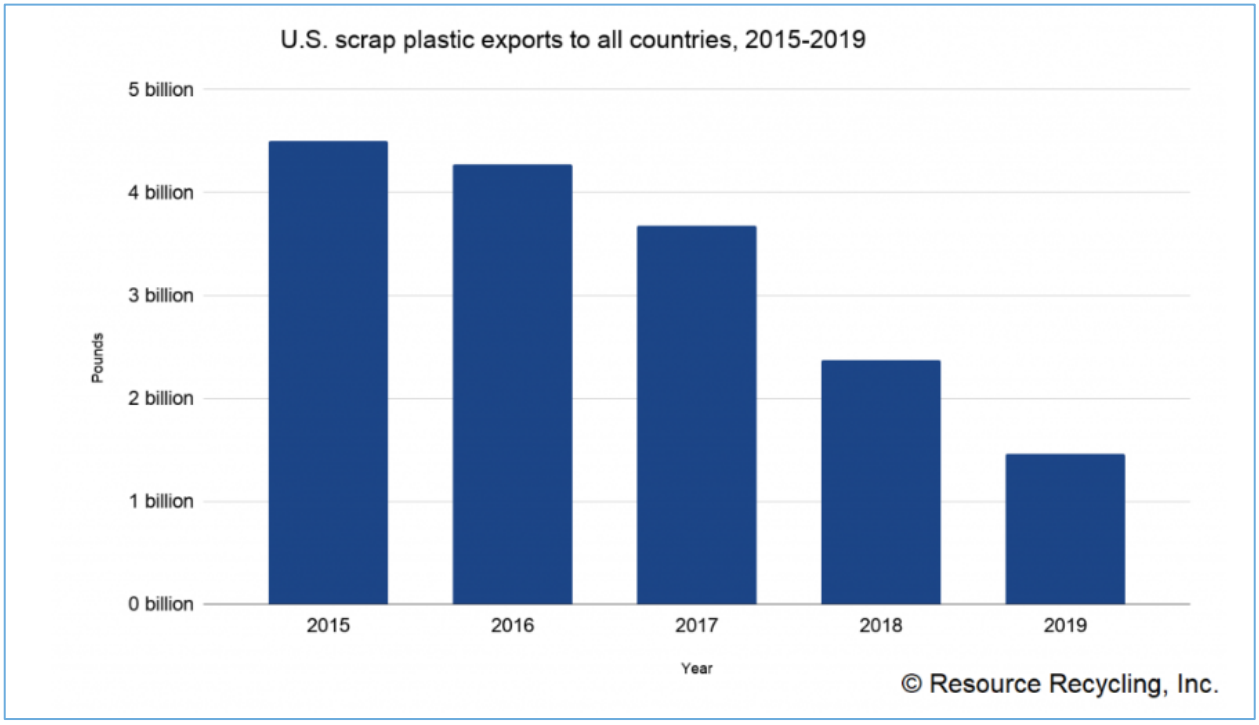
LLDPE = Linear low density polyethylene | PLA = Polylactide

‡ Other plastic packaging includes coatings, closures, lids, caps, clamshells, egg cartons, produce baskets, trays, shapes, loose fill, etc. PP and HDPE caps and lids recycled with PET bottles and jars are included in the recycling estimate for PET bottles and jars. Other resins include commingled/undefined plastic packaging recycling. Some detail of recycling by resin omitted due to lack of data.

Exports historically were a major component of EPA's "Recycled" category. For example, the 2017 EPA data shows 3.24 MM ton plastics were recycled. Per Figure 1,[4] exports accounted for more than half of this total, suggesting the domestic plastic recycling rate is less than 5%.

Plastic exports have been in decline for several years, and the amount of plastic going to landfills has increased.[5] China, formerly the largest destination for US recycled plastic, began restricting imports in 2013 and effectively banned imports in 2018, followed by several other Asian countries.[6] Plastic exports have dropped about 70% from 2015 to 2019, Figure 1 shows annual plastic exports dropped 1.5 MM tons from 2015-2019.

**Figure 1 US Scrap Plastic Exports**[4]




The EPA numbers may underestimate the amount of plastic entering landfills. For 2011 Themelis and Mussche estimated 39.3 MM tons of plastic in MSW, of which 32.5 MM ton were landfilled.[7] Their numbers are 20% higher than the EPA numbers, which they attribute to EPA's narrower definition of MSW. They also updated the heating value for plastics, compared to EPA. The combination of higher tonnage and heating values resulted in their estimate that landfilled plastics contained 1.0 quads, compared to the EPA estimate of 0.65 quads. Their estimated MSW plastic composition is shown in Table 2.

**Table 2. Estimated #3-7 Plastics in US MSW** (Themelis and Mussche)

| Materials | T&M Non-recycled plastics ( million tons) | MJ/kg |
|---|---|---|
| #1 Polyethylene terephthalate (PET) | 4.874 | 24 |
| #2 High-density polyethylene (HDPE) | 6.977 | 44 |
| #3 Polyvinyl chloride (PVC) | 2.162 | 19 |
| #4 Low density polyethylene (LDPE) | 7.705 | 44 |
| #5 Polypropylene (PP) | 5.464 | 44 |

| | | |
|---|---|---|
| #6 Polystyrene (PS) | 3.420 | 41 |
| #7 Other | 8.648 | 25 |
| **Total plastic in MSW** | 39.310 | |

Today more than 90% of the US population has access to curbside recycling, and/or drop-off centers,[8] which send recyclables to ~ 300 Material Recovery Facilities (MRFs). MRFs separate and bale multiple recyclables, and sell products to domestic and internationals recyclers. As noted above, over 3 MM tons of recycled plastics were exported in 2017. Due to restrictions by many countries MRFs are now sending some of the sorted and baled plastics to landfills due to lack of markets. Sorting single-source recyclables at MRF costs about $70/ton[9]. Machine sorting is imperfect, requiring downstream manual sorting. Consequently sorted bales that end up in landfills represent an economic and environmental burden.

Two plastics have maintained attractive prices from MRFs: PET and HDPE bottles. PET bottles have historically been recycled. PET thermoforms were previously discouraged in many recycling programs. Consequently only 40,000 tons of PET thermoform were recycled in 2017.[10] However, newer recycling technologies can process PET bottles and thermoforms.[11] HDPE natural and colored bottles are also financially attractive for recycling. Consequently REUSE does not encourage processes focused solely on PET or HDPE bottles.

The situation for most other plastics is not positive. The market for "#3-7" plastics (PVC, LDPE, PP, PS, and other, respectively) has effectively collapsed due to bans by waste importing countries. Recently the price of mixed plastic dropped to zero.[12] Many MRFs do not accept #3-7 plastics, and the low price has led some communities to suspend collection of plastics #3-7 plastics.[13]

Films (bags, sheets, packaging) are currently discouraged from most curbside recycling programs because they cause problems in sorting equipment and lead to contamination.[14] Films overlap the #3-7 category because films include #2 HDPE, #4 LDPE, and #5 PP. The 2020 Curbside Recycling Report estimates 3.6 MM tons of film material is available. The most recent industry report shows recycling rates for films dropped 24% to 0.5 MM tons in 2017 because bales could not meet Chinese quality standards.[15] They observed that it landfilling was more economical than recycling by the end of 2017. Most of the 0.4 MM ton/yr agricultural plastic use is films, and also has a low recycle rate.

Similar to films, bulk rigids are discouraged from many curbside programs. The primary components are injection molded HDPE and PP. Bales are likely to contain smaller amounts of other #3-7 polymers, depending on the MRF processing system.[16]

As noted above, the amount of plastic from MRF going to landfills has increased. One category is MRF bales that fall below buyers' specifications. China's ban on recycled paper or plastics with more than 0.5% contamination has led to a collapse of prices for bales that do not meet this requirement. Another category is residual bales from MRF. Typically 15-30% of the MRF incoming material is rejected as residuals.[17] Residual bales are often landfilled or sent to waste to energy facilities, but

contain enough plastic and paper to support "secondary" MRF processing. Consequently they could be attractive feeds for REUSE processes with sufficient flexibility.

There are several other post-consumer sources of plastics sent to landfills, including construction and demolition waste; auto shredder residue; agricultural waste; tires; and a latent but rapidly growing amount of composite wastes. Themelis and Mussche estimated 0.5 MM ton/yr plastic from construction and demolition waste are landfilled, along with 1.9 MM ton/yr plastics and 1.1 MM ton/yr rubber from auto shredder residue.[18] Agriculture used 0.4 MM ton plastic in 2018 for irrigation tubing, silage, greenhouses, and mulching, with very little recycled.[19] The US Tire Manufacturing Association reported 0.6 MM ton tires were landfilled in 2017,[20] much lower than the Environmental Protection Agency estimate of 4.95 MM ton for 2017.[21]

There is very little information regarding the amount of composite waste generated and/or sent to landfill. Global production of glass fiber composites was 5.8 MM ton in 2019; US production was 1.3 MM, which is expected to grow to 1.5 MM ton by 2025.[22] Natural fiber composites use cellulosic materials for fibers and petrochemical or bio-derived matrix. They are used in primarily in automobiles and construction applications, with an estimated global production of 0.5 MM ton in 2019 and 10% annual growth rate.[23] Carbon-based composite production is 0.1 MM ton/yr, and uses include aviation and transportation. All composites are found in items that are imported, exported, and typically have long lives, such as vehicles, construction, pipes and tanks. Further, recycling options are limited. Consequently these materials are likely to enter landfills as construction and demolition waste or auto shredder residue, although with a considerable lag time. For example, Gopalraj and Kärki have estimated future landfill quantities for composite turbine blades and circuit boards.[24]

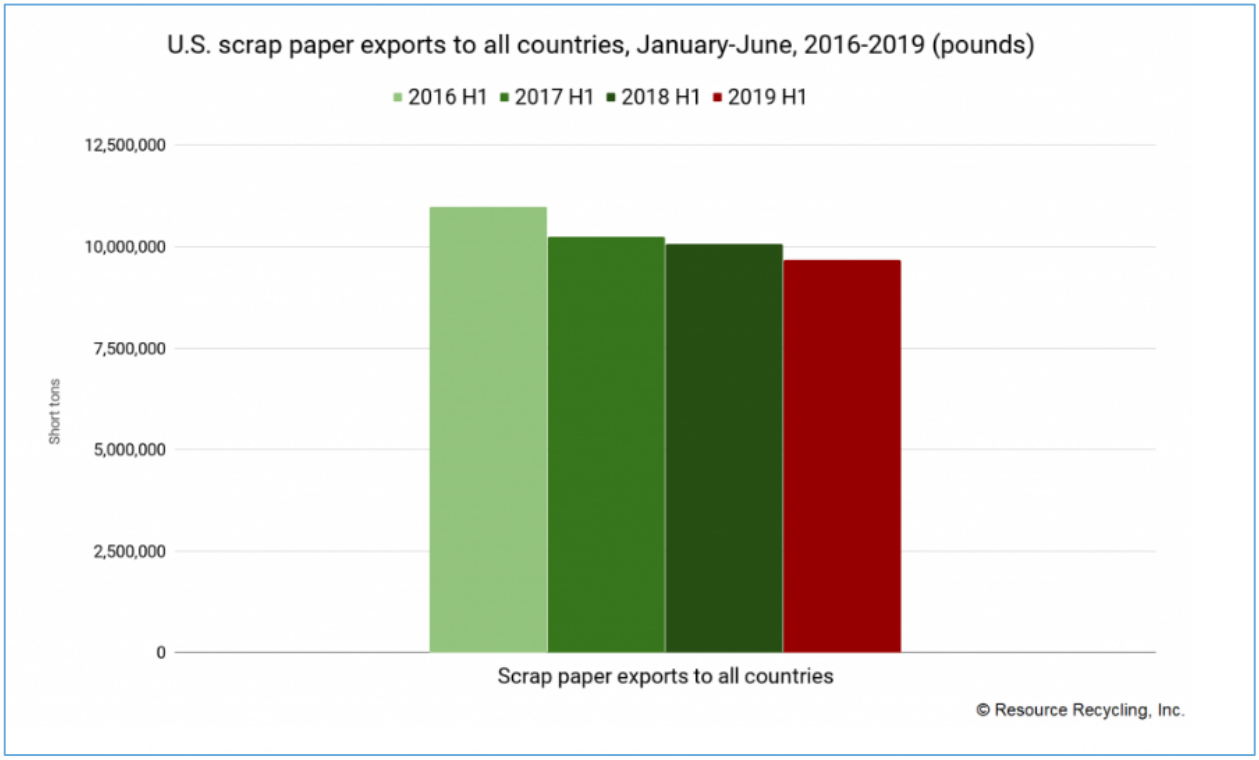
## Paper Feeds

Similar to plastics, paper is also an attractive feed. In 2017, landfills received 18.4 million tons of paper and paperboard.[25] However, compared to plastics, paper has a higher oxygen content, except PET, and potentially higher ash content. Oxygen and ash have unique impacts on the target processes. Consequently the maximum amount of paper that can be fed or co-fed with plastics will vary with processes and process conditions.

The primary target for waste paper feeds is paper containers and packaging (excluding corrugated boxes) which have a recycling rate of 15%. Other paper products have relatively high recycling rates and therefore out of scope for REUSE, including corrugated boxes (88%), newspaper (77%), and nondurable goods made of paper, excluding newspapers (48%).

The US is the largest exporter of waste paper in the world. Consequently waste import bans have impacted paper recycling and MRF economics. Figure 2 shows recent trends for exports.[26]

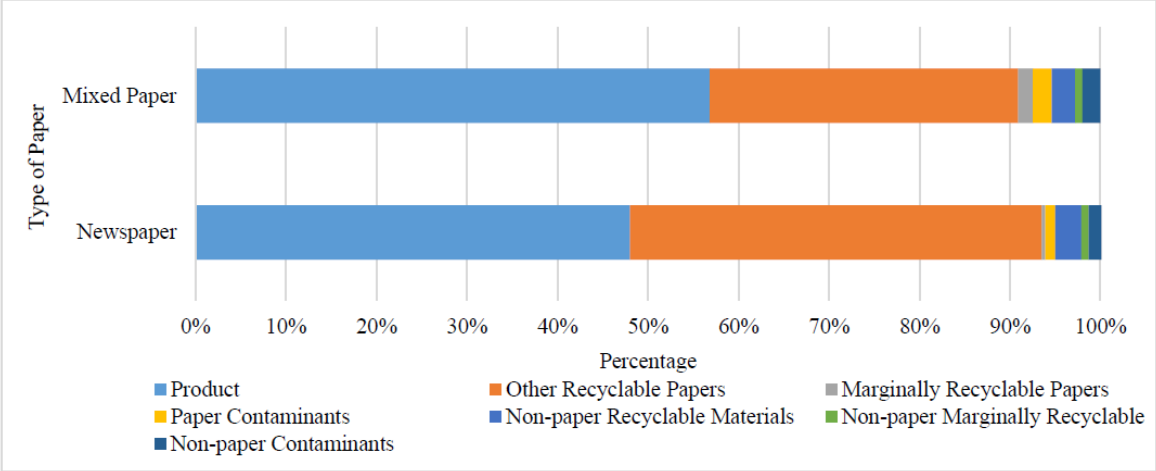
**Figure 2 US Scrap Paper Exports January-June, 2016-2019[26]**




Paper exports experienced a record 3 MM ton decrease in 2019, dropping from 21 MM tons in 2018 to 18 MM tons in 2019,[27] and the amount of paper going to landfills has increased.[28] The market for lower grades of paper from MRFs has collapsed. In August 2019 the national average price of mixed paper (PS 54) was $1.56/ton; currently it is -$2/ton.[29] Mixed paper is essentially paper that does not meet specifications for corrugated cardboard or newspaper. It includes phone books, magazines, junk mail, and office paper. It also includes paper containers and packaging, which have a low 15% recycling rate, as noted above. There is significant upside supply potential for mixed paper, since the Curbside Recycling Report notes the capture rate is only 46%.

Paper bales are often below the standards, and hence get sent to landfills. Machine sorting is imperfect, and even with downstream manual sorting plastics often get mixed into paper bales. For example, a recent study showed 5-29% of various types of plastics were lost to the paper stream in MRFs.[30] Yasar, et al reviewed the Institute of Scrap Recycling Industries, Inc. (ISRI) specifications for recycled paper categories.[31] Mixed paper can contain up to 2% prohibited materials (glass, metals, etc), and up to 5% prohibited materials plus outthrows (fiber grades inconsistent with mixed paper). The actual composition is variable, and depends on the MRF equipment and inbound materials. Cascadia's study showed that most of the MRF contaminants are found in the newsprint and mixed paper bales.[32] Figure 3 shows the distribution of prohibited materials and outthrows, from the Cascadia study, as reported by Yasar.

**Figure 3 Components of Mixed Paper and Newsprint from Cascadia Study[31]**




## Potential REUSE Technologies

ARPA-E is interested in processes that can process plastics, paper, and/or co-process plastics with paper. There are multiple process options for converting solid plastic or paper to a liquid product. Given the variable quality and quantity of high-energy waste streams, ARPA-E is interested in processes that are flexible and robust. The price and quality of feed is inversely related; the amounts of materials available depends on domestic and international markets. Consequently processes should have wide operating ranges, since feed quantity, quality, and price will vary over the life of a plant.

Slurrying the solid wastes may improve material handling and enhance reactivity. Several hydrocarbon refinery streams, including crude oil, vacuum gas oil (VGO), heavy cycle oil (HCO), light cycle oil (LCO), benzene/toluene/xylene (BTX), naphtha, and middle distillates, are potential slurrying agents.

Table 3 below provides an overview of process technologies, starting with processes that operate under reducing conditions, and progressing to processes that use increasing amounts of oxidation agents. The common step is cracking carbon-carbon bonds, which requires some combination of heat, chemical reactants, and/or catalysts. Table 3 is not meant to be comprehensive or limiting. It is used only to organize the following discussion of process technologies.

**Table 3 Overview of Process Options**

| Process | Slurry media | Heat | Reactant | Catalyst |
|---|---|---|---|---|
| Hydrotreating/hydrocracking/ hydrogenolysis | optional | From reaction | $H_2$ | Yes |
| Thermal cracking | Refinery | External | $H_2$ optional | |

| | | | | |
|---|---|---|---|---|
| | intermediate stream | | | |
| Torrefaction/Pyrolysis/Solvolysis /Hydrothermal liquefaction | varies | External | | optional |
| Autothermal pyrolysis/gasification | | From reaction | Air/oxygen | optional |
| Fluid catalytic cracking | optional | From catalyst regeneration | Air | Yes |
| Oxidative processes | | From reaction | various | |

**Hydrotreating/hydrocracking/hydrogenolysis**

Hydrotreating has been demonstrated with plastics slurried in hydrocarbon liquids. For example, German researchers in the 1990's catalytically hydrogenated slurries of various plastics and slurry media to produce aliphatic and aromatic compounds.[33] Thermal cracking followed by mild hydrotreating was piloted by RWE[22] and Veba[24] in the 1990's. More recently Palos et al showed HDPE slurried in light cycle oil (LCO) under hydroprocessing conditions gave good yields for middle distillates.[34] Proctor and Gamble patented a recycled plastics process using hydrotreating and kerosene as the solvent.[35]

Researchers at Manchester developed the Nova-Crack™ process, using hydrocracking catalysts to produce light alkanes. [36] Munir et al, have reviewed this area, including the concept of "co-processing", or co-feeding plastics with crudes and/or refinery intermediate streams. [37] They discuss results with various acidic micro/mesoporous catalysts.[38] Pakhmanova slurried LDPE, PP, and PET in VGO and found high conversion to light olefins and gasoline-range liquids.[39] Ucar, et al also used VGO in cracking tests for several plastics and plastic mixtures.[40] Karayildirima et al has demonstrated that mixtures of low density polystyrene and PVC in heavy vacuum oil have the ability to be converted to fuel with a catalytic cracking step using commercially available catalyst, DHC-8 in sulfide form of powder.[41]

An alternative approach is to use catalysts to "reverse" polymerization reactions to "deconstruct" polymers. For example, recent work on hydrogenolysis of HDPE using $Pt/SrTiO_3$ showed optimal conversion to distillates at 300 C and 170 psi $H_2$ with no slurry media/solvent [42].

There is considerable literature on hydrotreating biomass and biomass-derived liquids, but less work on hydrotreating paper. The previously mentioned BioCRACK process uses a hydrotreater to process the non-polar hydrocarbon liquid stream into distillate products. CRI Catalyst Company has demonstrated co-feeding refuse-derived fuel (RDF) and paper waste with biomass in the slurry-based hydrotreating $IH^2$ process.[43]

**Thermal cracking**

Thermal cracking in this context covers processes that slurry the plastic and/or paper feed in a refinery intermediate stream and use an indirectly heated reactor operating under reducing

conditions. From the 1970's through about 2000 there were numerous studies to convert plastics to fuel, often starting with a thermal cracking step. Coenen and Hagen discuss dissolution and thermal cracking of a wide range of plastics and tires in hydrocarbon slurry media and water from 150-500C.[44] Miller discusses dissolving and thermally cracking polyolefin wastes in Fischer-Tropsch liquids to produce lubricants.[45] RWE,[46,47] Rheinbraun[48] and Veba[49]conducted pilot studies on several types of plastics using thermal cracking at the front end of the overall process. Kohli et al used waste plastics as hydrogen donors when thermally cracking heavy crudes and vacuum resid to improve yields.[50] Dement'ev, et al obtained high yields by thermally depolymerizing polystyrene in HCO and LCO at 500 C.[51] Marcilla et al thermally cracked LDPE in vacuum gas oil (VGO).[52] The Western Research Institute thermally cracked several types of plastics in a modification of their ROPE™ process for heavy oil conversion.[53, 54] More recently Baraniec-Mazurek and Mianowski have investigated thermal cracking of polyolefins in tetralin, with and without a catalyst.[55]

Starting in the 1970's researchers also investigated thermal cracking of cellulosic materials. Wood flour, paper, and plastics was investigated in lab-based pilot units using donor solvents as an extension of coal-to-liquid processes.[56,57] Bouvier et al summarized earlier work on wood liquefaction.[58] Recently researchers at Iowa State investigated biomass with tetralin and lignin in a phenolic solvent[59]. Lange review's outlines the techno-economic challenges associated with solvent selection, and the advantages of aromatic-rich refinery streams in the gasol boiling range.[60] His group liquefied pinewood and model lignocellulose compounds in 1-methylnaphthalene.[61] Treusch et al also reported success with this approach, thermally cracking several biomass feeds mixed with VGO using the BioCRACK™ process.[62]

**Torrefaction/Pyrolysis/Solvolysis/Hydrothermal liquefaction**

Torrefaction/Pyrolysis in this context covers processes that use an indirectly heated reactor operating under reducing conditions (ie allothermal reactor). Solvolysis refers to in which the solvent is the primary reactant with the substrate. Hydrothermal liquefaction refers to sub- or supercritical reactions with water. There is extensive literature on pyrolysis, solvolysis, and hydrothermal liquefaction of composites,[63] plastics, cellulosic materials, and mixtures of plastics and cellulosic materials.

**Autothermal pyrolysis/gasification**

Autothermal pyrolysis/gasification in this context covers the continuum of processes that use substoichiomeric quantities of air/oxygen to produce liquid-range products. The equivalence ration, $\phi$ (moles oxidant fed/moles oxidant for complete combustion) ranges from about 0.5 for autothermal operations to about 0.4 for gasification. As $\phi$ increases, there is a general trend that the amount of liquids in the product will decrease and the amount of light gases will increase. For example, Arena et al show essentially no condensable liquids from gasification of residual plastics with a $\phi$ of 0.21-0.24.[64]

Several reactor configurations have been used to convert plastic wastes. Wey et al used an autothermal fluidized bed to convert tires[65] and polyethylene[66] to fuel streams. Sun et al used an autothermal fixed-bed catalytic reactor to decompose polystyrene.[67] PRTI has commercialized an updraft batch autothermal gasifier for producing liquid and gaseous fuels from tires.[68]

There is a long history of using gasifiers to produce condensable liquids from cellulosic materials. Reed and Das summarized early work, with a focus on downdraft gasifiers.[69] Milhe et al compared product slates for a continuous downflow reactor operating in pyrolysis (allothermal) versus autothermal modes, and found oxidation reactions in the autothermal mode produced less char and organic condensates, suggesting rapid oxidation of thermal decomposition products. Researchers at Iowa State have described a fluidized bed autothermal pyrolysis system that has been scaled up to 50 tons/day for biomass.[70] Their target products are oxygenated species, including sugars, acids, and phenolic oil.

**Fluid catalytic cracking**

There are many investigations of co-feeding plastics and refinery streams into catalytic cracking processes. Odjo et al reported on cracking LDPE slurried with VGO under FCC cracking conditions.[71] Arandes, et all found commercially available Octidyne 1160 BR on a REY zeolite gave complete conversion of polypropylene, polystyrene, and polystyrene–polybutadiene slurried in LCO.[72] They subsequently found good yields using mesoporous silica to crack polystyrene slurried with LCO , but low yields with polystyrene-butadiene mixtures slurried in LCO.[73]

There are fewer studies for co-processing paper or cellulosic materials under FCC conditions. Corma et al used sorbitol and glycerol as model compounds for biomass in FCC experiments. They found co-feeding glycerol with VGO was beneficial.[74] Similarly Swoboda et al tested 0-100% mixtures of glycerol and VGO.[75] Kumar et al found LCO and VGO could be used to co-feed lignocellulose to an FCC unit with attractive economics.[76]

Not all FCC processes require a slurry media. Anellotech's Bio-TCat™ process feeds biomass directly into the FCC riser[77]. They recently announced their related Plas-TCat™ technology can convert most waste plastics into commodity chemicals precursors used to make plastics for packaging and other products.[78]

**Oxidative processes**

Oxidation using reactants other than oxygen can produce liquids from hydrocarbons to highly-oxygenated products including ketones, aldehydes, and acids. Oluwoye, et al calculated the reactions of NO and $NO_2$ with polyethylene, suggesting NOx in the presence of oxygen could lead to oxidative depolymerization.[79] Yao et al describe production of valuable oxygenated compounds by oxidizing waste plastic with nitric acid.[80] Dang et al reported decomposition of glass fiber reinforced epoxy resin cured with amine using nitric acid, and the product could be recycled to produce new composites.[81] Sarkar et al report thermally cracking PET with $Ca(OH)_2$, although it is not clear if the $Ca(OH)_2$ is a catalyst or a reactant.[82] Jan et al report oxidative cracking of polyethylene with solid

sulfur, producing saturated alkanes and $H_2S$[83]. Adams et al report cracking polyethylene in ionic liquids.[84]

**Other Processes**

ARPA-E is interested in other novel and disruptive processes that can convert plastics and paper to a stable, easily transportable liquid product, ideally with high energy yield and minimal production of light gases and/or char. For example, in a series of papers, Saad and Williams describe a 2-stage process for dry reforming waste plastics, with the goal of producing syngas.[85] They were able to convert mixed plastics using $CO_2$ and steam, leaving open the potential to produce liquids instead of syngas.

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