PROCESS FOR CONVERTING MIXED WASTE PLASTIC INTO LIQUID FUELS AND WAXES BY CATALYTIC CRACKING

Abstract:
The present invention relates to a process for converting mixed waste plastic into liquid fuels and waxes by catalytic cracking. The process comprises the steps of introducing mixed waste plastic and a catalyst comprising a zeolite-type catalyst within a reactor; allowing at least a portion of the mixed waste plastic to be converted to liquid fuels and waxes within the reactor; and removing a product stream containing said liquid fuels and waxes from the reactor.

Title:
PROCESS FOR CONVERTING MIXED WASTE PLASTIC INTO LIQUID FUELS AND WAXES BY CATALYTIC CRACKING

Figure 1
Cumulative selectivity (%) of various product streams from the process, showing the distribution of gases, gasolines, kerosene, diesel, and HCO (hydrocarbons other than gasolines and diesel) at different reaction extents.

X = 88-93%

* 100HDPE
95HDPE-5PS
90HDPE-10PS
85HDPE-15PS
Process for converting mixed waste plastic into liquid fuels and waxes by catalytic cracking

This application claims priority to European application No. 15201 127.6 - filed on December 18, 2015-, the whole content of this application being incorporated herein by reference for all purposes.

The present invention relates to a process for converting mixed waste plastic into liquid fuels and waxes by catalytic cracking. The process comprises the steps of introducing mixed waste plastic and a catalyst comprising a zeolite-type catalyst within a reactor; allowing at least a portion of the mixed waste plastic to be converted to liquid fuels and waxes within the reactor; and removing a product stream containing said liquid fuels and waxes from the reactor.

In view of the increasing importance of polymers as substitutes for conventional materials of construction, such as glass, metal, paper, and wood, the perceived need to save non-renewable resources such as petroleum and dwindling amounts of landfill capacity available for the disposal of waste products, considerable attention has been devoted in recent years to the problem of recovering, reclaiming, recycling or in some way reusing waste plastic.

It has been proposed to pyrolyze or catalytically crack the waste plastic so as to convert high molecular weight polymers into volatile compounds having a much lower molecular weight. The volatile compounds, depending on the process employed, can be either relatively high boiling liquid hydrocarbons useful as fuel oils or fuel oil supplements or light to medium boiling hydrocarbons useful as gasoline-type fuels or as other chemicals.

Catalytic cracking of mixed waste plastic is a process well known to the person skilled in the art. For example, US 5,216,149 discloses a method for controlling the pyrolysis of a complex waste stream of plastics to convert such stream into useful high-value monomers or other chemicals, by identifying catalyst and temperature conditions that permit decomposition of a given polymer in the presence of others, without substantial decomposition of the other polymers.

polyethylene and polystyrene over spent FCC catalyst. The effect of the mixing proportions of polyethylene to polystyrene was studied and the authors found that an increase of polystyrene content in the reactants showed an increase of gasoline fraction and a decrease in kerosene, diesel and wax fractions in the obtained product. However, at the same time, the fraction of aromatic components in the liquid product dramatically increased to 70 % and more even at a polystyrene content of only about 40 %. This finding is confirmed in the publication of K.-H. Lee in Polymer Degradation and Stability 93 (2008) 1284-1289 where at a polystyrene content of 40 % even 90 % aromatics were obtained. The experiments reported by K.-H. Lee were conducted using 20 g of catalyst per 200 g of plastic.

While for certain applications increasing the fraction of gasoline obtained from the catalytic cracking of waste plastic can be of advantage, a high fraction of aromatic compounds can be undesired, for example due to the toxicity of such compounds. Furthermore, for other applications it is of advantage to increase the fractions of diesel and waxes, for example in case of an increasing demand for diesel in the market. It would therefore be desirable to provide a process for catalytic cracking of waste plastic wherein the ratio of gasoline to diesel fractions obtained can be tailored without undesirably increasing the fraction of aromatic compounds obtained.

The present inventors found that this and other problems as described below can surprisingly by solved by selecting a certain ratio of polystyrene to polyolefin in the mixed waste plastic and increasing the ratio of catalyst comprising a zeolite-type catalyst to waste plastic in the reactor.

The present invention therefore relates to a process for converting mixed waste plastic into liquid fuels and waxes by catalytic cracking, the process comprising:

- introducing mixed waste plastic and a catalyst comprising a zeolite-type catalyst within a reactor;
- allowing at least a portion of the mixed waste plastic to be converted to liquid fuels and waxes within the reactor; and
- removing a product stream containing said liquid fuels and waxes from the reactor,

characterized in that the mixed waste plastic contains from 2 to 50 % by weight of polystyrene and from 50 to 98 % by weight of polyolefin, each based on the
total weight of polystyrene and polyolefin in the mixed waste plastic, and in that the weight ratio of catalyst to mixed waste plastic in the reactor is above 1:10.

In the catalytic cracking of plastic several fractions of chemical compounds are obtained. Usually, there is a gas fraction containing light weight chemical compounds with less than 5 carbon atoms. The gasoline fraction contains compounds having a low boiling point of for example below 216°C. This fraction includes compounds having 5 to 11 carbon atoms. The kerosene and diesel fraction has a higher boiling point of for example 216°C to 359°C. This fraction generally contains compounds having 12 to 21 carbon atoms. The even higher boiling fraction is generally designated as wax (Heavy Cycle Oil or HCO). In all the fractions, the compounds are hydrocarbons which optionally comprise heteroatoms, such as N, O, etc. "Liquid fuels" in the sense of the present invention therefore are fuels like gasoline and diesel but may also be used as other valuable chemicals or solvents. "Waxes" designate such hydrocarbons which are solid at room temperature (23°C) and have a softening point of generally above 45°C.

A plastic is mostly constituted of a particular polymer and the plastic is generally named by this particular polymer. Preferably, a plastic contains more than 25 % by weight of its total weight of the particular polymer, preferably more than 40 % by weight and more preferably more than 50 % by weight. Other components in plastic are for example additives, such as fillers, reinforcing, processing aids, plasticizers, pigments, light stabilizers, lubricants, impact modifiers, antistatic agents, inks, antioxidants, etc. Generally, a plastic comprises more than one additive.

Plastics used in the process of the present invention are polyolefins and polystyrene, such as high density polyethylene (HDPE), low density polyethylene (LDPE), polypropylene (PP) and polystyrene (PS). Mixed plastics mostly constituted of polyolefin and polystyrene are preferred. In this context "mostly constituted" is to be understood such that the concentration of the polyolefin and the polystyrene in the mixed plastic is above 50 % by weight, more preferably above 75 % by weight, each based on the total weight of the dry mixed plastic. The mixed plastic may be constituted of polyolefin and polystyrene. Preferably, the mixed plastic contains less than 99.5 % by weight, more preferably less than 99 % by weight of polyolefin and polystyrene, based on the total weight of the dry mixed plastic.
Other plastics, such as polyvinylchloride, polyvinylidene chloride, polyethylene terephthalate, polyurethane (PU), acrylonitrile-butadiene-styrene (ABS), nylon and fluorinated polymers are less desirable. If present in the waste plastic, they are preferably present in a minor amount of less than 50 % by weight, preferably less than 30 % by weight, more preferably less than 20 % by weight, even more preferably less than 10 % by weight of the total weight of the dry waste plastic. Preferably, the individual content of any less desirable plastic is less than 5 % by weight, more preferably less than 2 % by weight based on the total weight of the dry waste plastic.

Preferably, the plastics waste starting material comprises one or more thermoplastic polymers and is essentially free of thermosetting polymers. Essentially free in this regard is intended to denote a content of thermosetting polymers of less than 15, preferably less than 10 and even more preferably less than 5 wt% of the composition.

Usually, waste plastic contains other non-desired components, namely foreign material, such as paper, glass, stone, metal, etc.

In the context of the present invention whenever it is referred to the weight of the waste plastic or the weight of the polystyrene and polyolefin in the mixed waste plastic, this weight relates to the weight of the dry plastic without any foreign material being admixed with the plastic. However, the weight includes any components in the plastic, such as the above described additives.

The present inventors found that when using a catalyst comprising a zeolite-type catalyst, at a weight ratio of catalyst to mixed waste plastic in the reactor of above 1:10, the addition of polystyrene to polyolefin plastic surprisingly has a significant impact on product distribution. Contrary to what was reported by K.-H. Lee (see above cited references), the inventors found that higher ratios of polystyrene in the mixed waste plastic lead to higher contents of diesel and waxes in the product stream obtained from the reactor. Thus, the process of the present invention allows modifying the selectivity of the product with respect to higher selectivity to diesel and waxes and lower selectivity to gases and gasoline. It was for example found that wax production even doubles when 15 % of polystyrene is present in the mixed waste plastic.

It was furthermore found that in the process of the present invention the diesel quality is improved with respect to an increased concentration of saturated compounds and a decreased concentration of unsaturated compounds, in particular aromatic compounds. This finding is particularly unexpected as the
addition of polystyrene (an aromatic based polymer) should lead to a higher content of unsaturated and in particular aromatic compounds. At the same time, the also obtained gasoline fraction maintains its high level of quality with respect to Research Octane Number (RON) and Motor Octane Number (MON).

It was found that the above advantages are achieved if the mixed waste plastic contains from 2 to 50% by weight of polystyrene, preferably from 2 to 40% by weight, more preferably from 2 to 30% by weight and even more preferably from 2 to 20% by weight of polystyrene, based on the total weight of polystyrene and polyolefin in the mixed waste plastic. Most preferably, the mixed waste plastic contains from 2 to 15% by weight, such as from 5 to 15% by weight of polystyrene based on the total weight of polystyrene and polyolefin in the mixed waste plastic.

The process of the present invention is also characterized in that the weight ratio of catalyst to mixed waste plastic in the reactor is above 1:10. Preferably, the weight ratio of catalyst to mixed waste plastic in the reactor is above 1:9, more preferably above 1:8, more preferably above 1:7, more preferably above 1:6, more preferably above 1:5, more preferably above 1:4 and even more preferably above 1:3, such as above 1:2. A particularly preferred weight ratio of catalyst to mixed waste plastic in the reactor is about 1:1.5.

The weight ratio of catalyst to mixed waste plastic in the reactor can be below 10:1, preferably below 7:1. Thus, the weight ratio of catalyst to mixed waste plastic in the reactor can be for example in the range of from 1:9 to 10:1, preferably from 1:8 to 10:1, preferably from 7:1 to 10:1, preferably from 1:6 to 10:1, preferably from 1:5 to 10:1, preferably from 1:4 to 10:1, preferably from 1:3 to 10:1 and even more preferably from 1:2 to 10:1 or from 1:2 to 7:1.

The catalyst used in the process of the present invention comprises a zeolite-type catalyst. In one embodiment, the catalyst predominantly is a zeolite-type catalyst. In a further embodiment, the catalyst consists of a zeolite-type catalyst. In a third embodiment, the catalyst additionally comprises a further catalyst, in particular an amorphous-type catalyst.

The embodiment in which the catalyst predominantly is a zeolite-type catalyst is preferred. In this context, the term "predominantly" defines a catalyst which is a mixture of a zeolite-type catalyst and a non-zeolite-type catalyst, such as an amorphous catalyst, but wherein the catalyst comprises more than 50% by weight of the zeolite-type catalyst based on the total weight of the catalyst. Preferably, the catalyst comprises more than 60%, more preferably more than
70 %, even more preferably more than 80 % and most preferably more than 90 % of the zeolite-type catalyst. The catalyst can comprise a single zeolite-type catalyst or a mixture of two or more zeolite-type catalysts.

As catalyst all types of FCC catalysts may be used. FCC catalysts are well known to the person skilled in the art. For example, the zeolite-type catalyst may be selected from crystalline microporous zeolites which are known to the person skilled in the art and which are commercially available. Preferred examples for zeolite-type catalysts are described in WO 2010/135273, the content of which is incorporated herein by reference. Specific examples for suitable zeolite-type catalysts include but are not limited to ZSM-5, ZSM-11, ZSM-22, ZSM-23, ZSM-35, ZSM-48, ZSM-50, TS-1, TS-2, SSZ-46, MCM-22, MCM-49, FU-9, PSH-3, ITQ-1, EU-1, NU-10, silicalite-1, silicalite-2, boralite-C, boralite-D, BCA, and mixtures thereof.

If the catalyst additionally comprises an amorphous-type catalyst, this may comprise silica, alumina, kaolin, or a mixture thereof. Silica, in particular in the form of sand, is a well known for FCC catalytic applications. Preferred amorphous-type catalysts comprise at least 60 % by weight, preferably at least 70 % by weight and even more preferably at least 80 % by weight of silica-equivalent of an oxodic compound based on silicon like silica (\( \text{SiO}_2 \)), kaolin, etc.

The catalyst can be fresh catalyst, equilibrated catalyst (such as spent catalyst), or a mixture thereof.

The mixed waste plastic and the catalyst comprising the zeolite-type catalyst can be introduced within the reactor simultaneously or subsequently. Furthermore, the mixed waste plastic and the catalyst comprising the zeolite-type catalyst can be introduced within the reactor batchwise or continuously.

The skilled person is aware of suitable apparatus and equipment for carrying out the process in accordance with the present invention and will select the suitable system based on his professional experience, so that no further extensive details need to be given here. However, without willing to be bound to theory, some examples of reactor technologies that can be effectively used to carry out the invention comprise the stirred reactor, the rotary kiln, the bubbling fluidized bed reactor and the circulating fluidized bed reactor riser or downer. The rotary kiln is a cylindrical vessel, inclined slightly to the horizontal, which is rotated slowly about its axis. The material to be processed is fed into the upper end of the cylinder. As the kiln rotates, material gradually moves down towards the lower end, and may undergo a certain amount of stirring and mixing. In a
bubbling fluidized bed reactor a fluid (gas or liquid) is passed through the
catalyst particles at high enough velocities to suspend the catalyst and cause it to
behave as though it were a fluid. In a circulating fluidized bed, also called
transport reactor, the catalyst and the fluid flow co-currently at high speed.

Generally a cyclone system is used to separate the fluid, which can undergo
downstream processing, from the solid, which is recirculated to the reactor.
These reactors can be either upflow for risers, or downflow for downers.

Preferably, they are introduced continuously. In one embodiment, the
whole process is conducted continuously.

In the reactor, in presence of the catalyst, at least a portion of the mixed
waste plastic is converted to liquid fuels and waxes. This conversion preferably
takes place at an elevated temperature of for example above 350°C, preferably
above 400°C, more preferably above 410°C. Suitably, the conversion takes place
at a temperature in the range of above 410°C to 500°C, more preferably in the
range of from 420°C to 450°C, such as about 425°C.

The effect of the polystyrene content in the mixed waste plastic at a high
weight ratio of catalyst to mixed waste plastic in the reactor is now explained in
more detail with reference to the following examples and the attached figures
which show in

Figure 1 the effect of polystyrene loading on selectivity,
Figure 2 the effect of polystyrene loading on the quality of the diesel
fraction, and
Figure 3 the effect of polystyrene loading on the quality of the gasoline
fraction.

The examples below were conducted according to the following general
experimental procedure:

In each catalytic run in semibatch mode, 30 g of plastic (high density
polyethylene (HDPE) and variable amounts of polystyrene (PS)) and a defined
amount of the catalyst were loaded inside the reactor. The reactor was closed and
heated from room temperature to 200°C during 20 minutes, while simultaneously
purging with a 150 mL/min nitrogen flow. When the internal temperature
reached the melting point of the plastic, stirring was started and was slowly
increased until 690 rpm. The temperature was held at 200°C for 25-30 minutes.
During this heating process, nitrogen coming out from the reactor was not
collected.
After this first pretreatment step, the temperature was increased to the reaction temperature at a heating rate of 10°C/min, and the collection of gases and nitrogen in the corresponding gas sampling bag was started. When the internal temperature reached the reaction temperature, the circulation of the gaseous products was commuted to another pair of glass traps and corresponding gas sampling bag. This was considered as the zero reaction time.

During selected time periods, liquid and gaseous products were collected in a pair of glass traps and their associated gas sampling bag, respectively. At the end of the experiment the reactor was cooled to room temperature. During this cooling step, liquids and gases were also collected.

The reaction products were classified into 3 groups: i) gases, ii) liquid hydrocarbons and iii) residue (waxy compounds, ashes and coke accumulated on the catalyst). Quantification of the gases was done by gas chromatography (GC) using nitrogen as the internal standard, while quantification of liquids and residue was done by weight.

The simulated distillation (SIM-DIS) GC method allowed the determination of the different fractions in the liquid samples (according to the selected cuts); the detailed hydrocarbon analysis (DHA) GC method allowed the determination of the PIONA (paraffins, iso-paraffins, olefins, napthenes, aromatics) components in the gasoline fraction of the last withdrawn sample (C5-C11: Boiling point < 216.1°C; what includes C5-C6 in the gas sample and C5-C11 in the liquid samples), and GCxGC allowed the determination of saturates (everything that is not aromatic), mono-, di- and tri-aromatics in the diesel fraction of the last withdrawn liquid samples (C12-C21; 216.1 < BP < 359°C).

Should the disclosure of any patents, patent applications, and publications which are incorporated herein by reference conflict with the description of the present application to the extent that it may render a term unclear, the present description shall take precedence.

Example 1

The experiment was carried out following the general procedure described above. In this example, the raw material was pure HDPE (labelled 100HDPE). Reaction temperature was set at 425°C. In this example, 20 g of zeolite-type catalyst were used (catalyst: spent FCC catalyst provided by FCC Equilibrium Catalyst, Inc.). Catalyst to plastic weight ratio was equal to 20/30.
Example 2
The experiment was carried out following the general procedure described above. In this example, the raw material is a mixture containing 95 wt. % HDPE and 5 wt. % PS (labelled 95HDPE-5PS). Reaction temperature was set at 425°C. In this example, 20 g of zeolite-type catalyst have been used (catalyst: spent FCC catalyst provided by FCC Equilibrium Catalyst, Inc.). Catalyst to plastic weight ratio was equal to 20/30.

Example 3
The experiment was carried out following the general procedure described above. In this example, the raw material was a mixture containing 90 wt. % HDPE and 10 wt. % PS (labelled 90HDPE-10PS). Reaction temperature was set at 425°C. In this example, 20 g of zeolite-type catalyst were used (catalyst: spent FCC catalyst provided by FCC Equilibrium Catalyst, Inc.). Catalyst to plastic weight ratio was equal to 20/30.

Example 4
The experiment was carried out following the general procedure described above. In this example, the raw material was a mixture containing 85 wt. % HDPE and 15 wt. % PS (labelled 85HDPE-15PS). Reaction temperature was set at 425°C. In this example, 20 g of zeolite-type catalyst were used (catalyst: spent FCC catalyst provided by FCC Equilibrium Catalyst, Inc.). Catalyst to plastic weight ratio was equal to 20/30.

The effect of polystyrene loading on the selectivity of the process is shown in Figure 1. It is evident that the selectivity for gases and gasoline decreases with increasing polystyrene loading while the selectivity for diesel and waxes increases.

However, contrary to the expectation of the skilled person and contrary to what is described in the prior art, the process of the present invention provides a diesel fraction which in particular at low polystyrene loading comprises an even decreased amount of aromatic compounds. This effect is shown in Figure 2, which provides the amounts of saturated (S), monoaromatic (MA), diaromatic (DA), triaromatic (TA) and polyaromatic (PA) compounds in the obtained diesel fractions depending on the polystyrene loading.

Figure 3 shows the effect of the polystyrene loading on the quality of the gasoline fraction (P: paraffins, I: iso-paraffins, O: olefins, N: naphthenes, A: aromatics, U: unidentified). Figure 3 additionally shows the RON and MON of the gasoline fractions obtained with different polystyrene loadings.
Figures 1, 2 and 3 additionally provide the conversion (X) of the mixed waste plastic in each run.
CLAIMS

1. Process for converting mixed waste plastic into liquid fuels and waxes by catalytic cracking, the process comprising:

   introducing mixed waste plastic and a catalyst comprising a zeolite-type catalyst within a reactor;

   allowing at least a portion of the mixed waste plastic to be converted to liquid fuels and waxes within the reactor; and

   removing a product stream containing said liquid fuels and waxes from the reactor,

   characterized in that the mixed waste plastic contains from 2 to 50 % by weight of polystyrene and from 50 to 98 % by weight of polyolefin, each based on the total weight of polystyrene and polyolefin in the mixed waste plastic, and in that the weight ratio of catalyst to mixed waste plastic in the reactor is above 1:10.

2. Process according to claim 1, wherein the mixed waste plastic contains from 2 to 40 % by weight, preferably from 2 to 30 % by weight, more preferably from 2 to 20 % by weight of polystyrene, based on the total weight of polystyrene and polyolefin in the mixed waste plastic.

3. Process according to claim 1 or 2, wherein the weight ratio of catalyst to mixed waste plastic in the reactor is above 1:8, preferably above 1:5, more preferably above 1:3.

4. Process according to any of the preceding claims, wherein the weight ratio of catalyst to mixed waste plastic in the reactor is below 10:1, preferably below 7:1.

5. Process according to any of the preceding claims, wherein the catalyst predominantly is a zeolite-type catalyst.

6. Process according to any one of claims 1 to 4, wherein the catalyst consists of zeolite-type catalyst.
7. Process according to any one of claims 1 to 5, wherein the catalyst additionally comprises an amorphous-type catalyst.

8. Process according to claim 7, wherein the amorphous-type catalyst comprises silica, alumina, kaolin, or a mixture thereof.

9. Process according to any of the preceding claims, wherein the catalyst is fresh catalyst, equilibrated catalyst, or a mixture thereof.

10. Process according to any of the preceding claims, wherein the temperature at which at least part of the mixed waste plastic is converted to liquid fuels and waxes in the reactor is above 350°C, preferably above 410°C, more preferably in the range of above 410°C to 500°C, even more preferably in the range of from 420°C to 450°C.

11. Process according to any of the preceding claims, wherein the mixed waste plastic comprises more than 50 % by weight of polystyrene and polyolefin, based on the total weight of the mixed waste plastic.

12. Process according to any of the preceding claims, which is conducted continuously.

13. Process according to any of the preceding claims, wherein the waste plastic is selected from post consumer waste plastic, off-spec plastic and industrial scrap plastic.

14. Process according to any of the preceding claims, wherein the waste plastic is essentially free of thermosetting polymers.
**Figure 1**

Cumulative selectivity (%) for different products:
- **Gases**
- **Gasolines**
- **Kerosene**
- **Diesel**
- **HCO**

Legend:
- 100HDPE
- 95HDPE-5PS
- 90HDPE-10PS
- 85HDPE-15PS

X = 88-93%
Figure 3
A. CLASSIFICATION OF SUBJECT MATTER

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
CIOG

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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<td>WO 2015/128033 AI (SAUDI BASIC IND CORP [SA]; SABIC GLOBAL TECHNOLOGIES BV [NL]; WARD AND) 3 September 2015 (2015-09-03) example 1</td>
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<td>WO 2014/040634 AI (OUTOTEC 0YJ [FI]; SCHAAF TANJA [DE]; BINDER CHRISTIAN [DE]; ANASTASJE) 20 March 2014 (2014-03-20) page 7, line 12 - line 24</td>
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Further documents are listed in the continuation of Box C.

See patent family annex.

Date of the actual completion of the international search

10 March 2017

Date of mailing of the international search report

17/03/2017

Authorized officer

Bernet, Olivier
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