



## Description

### TECHNICAL FIELD OF THE INVENTION

[0001] This invention refers to a process aimed at converting plastic wastes into materials which can be used, as it is common in the so-called circular economy. Also, a plant for carrying out the process is referred to, according to this invention.

### BACKGROUND OF THE INVENTION

[0002] Waste disposal has been a problem since the industrial production has become massive. The massive production has made the most common products easily available: very often, to replace a broken item is much easier and results in higher cost savings than to repair the same. Of course, this leads to the disposal of any kind of object when it breaks, when it does not function any more, in case of obsolescence, when it is no longer fashionable, when there is not enough room at home for storing it, and so on.

[0003] While waste disposal was rather simple until relatively recently, some big drawbacks have arisen recently. Pollution has become an issue and air and water pollution and soil contamination are now not accepted any more by people. Incinerators are disliked because of their fumes, which are normally considered polluting, and people normally try to avoid their setting up. Landfills consume soil, result in leaks of harmful liquids, which can contaminate soils and waters. Moreover, the massive production and its related production of wastes leads to shortage of raw materials, which are becoming more and more expensive. The so-called greenhouse effect and the related global warming have also given their contribution to the problem, giving another reason to avoid waste incineration whenever unnecessary.

[0004] This situation has led to intensive investigations, aimed at avoiding, on one hand, the need of disposing of wastes and, on the other hand, to push as much as possible towards a reuse, whenever possible, which is called "circular economy".

[0005] In order to achieve these two results, a proper waste collection is very important. In the past, wastes were collected together and disposed of either in incinerators or in landfills, altogether, with no sorting at all. In order to reuse wasted products, this collection is completely unsuitable. Regeneration or reclamation can take place only with at least relatively homogeneous wastes and this can be achieved only in two ways: 1) wastes are collected separately or 2) wastes are sorted out after their collection. It is apparent that route 2) is much more complicated, requires unqualified manpower doing a long-lasting job, with a very high likelihood of error. Route 1) appears much more viable. A differentiated collection at the origin is much simpler, much more accurate, is normally performed by the wasting subject which has only to decide where any item should be put before

bringing wastes to the public collection. In spite of some deficiencies, this route is proving rather successful, and people are more and more learning how to separate wastes.

5 [0006] One of the fractions to be collected is normally made up by plastic wastes which is a large percentage of the overall waste. The regeneration of plastic materials is often rather difficult, not always can be performed successfully and normally the recycled charge can be added  
10 to the virgin material only in a rather small percentage (normally below 30 wt.%). Therefore, a large part of plastic wastes is still to be disposed of; namely, plastic recycling is only partial and cannot lead to a complete reuse. This is a big issue, because of the long-lasting  
15 features of plastic materials which do not decompose spontaneously.

[0007] The fact that, with some exceptions, plastic wastes can be recycled only partially has pushed researchers to develop other processes. In particular, since  
20 plastic materials are polymers, depolymerisation processes to obtain monomers and/or oligomers are a way to dispose of plastic wastes and obtain raw materials for other reactions. In this view, Politecnico di Milano elaborated a process for disposing of plastic wastes which involves the transformation into lubricants and  
25 hydrogen under mild process conditions. This process is very suitable; however, it involves some problems, especially in moving plastic wastes during at least the first steps thereof and produces pyrolysis oil, which is  
30 normally non-regenerable or very hardly regenerable and should be disposed of.

[0008] Another problem related to wastes is the disposal of waste lubricants, which is the core business of the Applicant. The Applicant has filed a number of patents  
35 aimed at recovering waste mineral oils, leading to a recover of more than 95 wt.% of the collected waste oils. This is surely a good achievement, which allows to get lubricants at a lower cost, to reduce the oil consumption for the production of lubricants and avoids waste combustion or contamination of soils. There is also a good  
40 saving in energy costs.

[0009] Although the recovery of waste lubricants is becoming more and more important, a part of the waste lubricants is not yet collected properly or is even simply  
45 poured in the environment and a part of it cannot be regenerated, particularly when the contents in water are higher than 15 wt.%, flash point of the oil is below 120 °C, content in zinc and/or lead is higher than 2000 mg/kg, thinners are more than 15 vol.%, PCB/PCT is  
50 higher than 25 mg/kg, saponification is higher than 30 mg/kg and/or neutralisation number is higher than 10 mg KOH per g of oil. Under such conditions, waste lubricants are normally either incinerated or somehow disposed of. It would be very useful to find a use therefor, so as to  
55 reduce the environmental impact and the oil depletion.

## SUMMARY OF THE INVENTION

**[0010]** Problem underlying this invention is to propose a process for recovering waste plastics overcoming the above drawbacks and allowing also to employ, without disposing of them, lubricants which cannot be regenerated. This purpose is achieved, according to a first aspect, through a process for the conversion of plastic wastes, involving the depolymerisation of the plastic material making up the waste, wherein the plastic waste is extruded producing a molten mass which is mixed with hydrogen and fed to a hydrocracking reactor, characterised in that the waste plastic, upon extrusion, is mixed with lubricants which cannot be regenerated and the resulting molten mass contains both plastic and lubricant wastes. According to a second aspect, the problem is solved by a plant for carrying out the process above, characterised in that it comprises a unit for the pre-treatment of the lubricants which cannot be regenerated, a melting unit for melting the plastic wastes and mixing them to the pre-treated lubricants and a depolymerisation unit. Subclaims disclose preferential features of the invention.

**[0011]** According to an embodiment, lubricants which cannot be regenerated are mineral oils, pyrolysis oils or used oils.

**[0012]** According to an embodiment, the lubricants which cannot be regenerated are pre-treated by a pre-flash distillation step.

**[0013]** According to an embodiment, the pre-flash column is a hole plate or an open packed column.

**[0014]** According to an embodiment, pre-flash takes place at a temperature between 120 and 160 °C and a pressure ranging from 0.2 to 0.7 bar.

**[0015]** According to an embodiment, the lubricants are distilled in a distillation column; preferably, such column is a plate or a packed column.

**[0016]** According to an embodiment, such a distillation step takes place at 250 to 400 °C under a pressure of 0.025 to 0.055 bar.

**[0017]** According to an embodiment, lubricants are temporarily stored in a tank after the pre-treatment steps.

**[0018]** According to the invention, plastic wastes are shredded in a shredder before being mixed with the pre-treated lubricants, before being fed to an extruder.

**[0019]** According to an embodiment, plastic wastes are a mixture containing PE and/or PP and/or PS and/or PET and/or PVC and/or ABS and/or BR. No separation of the materials is required.

**[0020]** According to an embodiment, the mixture of plastic wastes and pre-treated lubricants is molten within the said extruder.

**[0021]** According to an embodiment, the molten material is temporarily stored in a tank. The material within the said tank is kept in a molten state. The temperature within the said tank is kept at 270 to 280 °C.

**[0022]** According to an embodiment, the molten material is further moved by a gear pump.

**[0023]** According to an embodiment, the molten material undergoes a removal of impurities. Impurities are normally sulphur, nitrogen, chlorine and metals.

**[0024]** According to an embodiment, the de-polymerisation takes place within a hydrocracker; the hydrocracking reaction takes place under catalysis, preferably with a heterogeneous catalyst.

**[0025]** According to an embodiment, the product of the depolymerisation is distilled into a number of different fractions; distillation preferably takes place in a packed column.

**[0026]** According to an embodiment, four feeds are fed to the process: waste lubricants, plastic wastes, hydrogen.

**[0027]** According to an embodiment, wastewater, light distillate, gasoline, bitumen, bases for lubricants, ultra-low sulphur diesel are products of the inventive process.

## BRIEF DESCRIPTION OF THE DRAWING

**[0028]** Further features and advantages of this invention better appear when reading the following detailed description of a preferred embodiment, disclosed by way of non-limiting example only and shown in the appended drawing, wherein the only figure is a flow diagram of the process according to this invention.

## BEST WAY TO CARRY OUT THE INVENTION

**[0029]** A line 1 feeds waste lubricants which cannot be regenerated to the process, entering a pre-flash column 2; the column 2 has two outlets: an outlet 3 (head) for water and other solvents to be removed from the lubricant and a line 4 (tail), feeding a furnace 5, for reaching a proper temperature.

**[0030]** The pre-flash column 2 can be of any suitable kind. Preferably, it is a hole plate or an open packed column, which are the columns used most frequently.

**[0031]** The line 4, after the furnace 5, feeds a distillation column 6. Also the column 6 can be a plate or a packed column.

**[0032]** The column 6 has four outlets.

**[0033]** A first outlet 7 removes light distillates, which are normally to be oxidised for producing at least part of the heat which is needed for carrying out the process.

**[0034]** A second line 8 brings away gasoline, which is a valuable product for further use or storage.

**[0035]** A third outlet 9 downloads bitumen, which can be stored or sold.

**[0036]** Finally, the outlet 10 feeds semi-finished lubricants to the subsequent steps of the process; generally, the semi-finished lubricants are stored in a tank 11 for the next step of the inventive process.

**[0037]** The part of the plant just described serves for preparing the lubricant to be added to the plastic materials to be depolymerised, so as to improve its extrudability.

**[0038]** An extruder 12 receives semi-finished lubricants from the tank 11 via a tube 13, with the aid of a

pump 14, and plastic wastes, fed from a line 15 and a shredder 16, in order to better prepare the plastic wastes for the subsequent melting. A line 17, leaving the extruder 12, feeds a tank 18, where the material is kept in a molten state.

**[0039]** A line 19 brings the molten material, by means of a gear pump 20, to a heater 21 and to a furnace 22, after mixing the molten material with hydrogen coming from a line 23. The gear pump 20 is particularly suitable for moving viscous or even very viscous liquid, like these molten materials.

**[0040]** The heated material is fed to a reactor 24, where a demetallisation reaction takes place, removing metal impurities from the material. The outlet 25 thereof is fed to a hydrocracker 25 through a line 26; the line 26 merges with a line 27, which feeds hydrogen to the same reactor 25, so as to be able to carry out the intended reactions. The outlet 28 from the reactor 25 feeds a distillation column 29; such a column is preferably a packed column. The distillation leads to outlets 30, 31, 32, 33, 34 and 35.

**[0041]** Products come from the column 29, through six outlets 30 to 35.

**[0042]** Now, turning to the process according to this invention, it should be appreciated that four feeds to the plant are provided: waste lubricants (1), plastic wastes (15) and hydrogen (23 and 27). On the other hand, ten outlets, with products, marketable byproducts and wastes, are provided: wastewater (3), light distillates (7), gasoline (8), bitumen (9 and 35), bases for lubricants (32, 33 and 34), ultra-low sulphur diesel (31) and light distillates (30).

**[0043]** Lubricants fed through the pipe 1 are waste lubricants which cannot usually be regenerated and which are normally incinerated. They can be mineral oils, pyrolysis oils or used oils. As it has been stated above, they have contents in water higher than 15 wt.%, flash point of the oil below 120 °C, contents in zinc and/or lead higher than 2000 mg/kg, thinners which are more than 15 vol.%, PCB/PCT higher than 25 mg/kg, saponification higher than 30 mg/kg and/or neutralisation number higher than 10 mg KOH per g of oil. The process according to the present invention offers an alternative to the combustion of such wastes, increasing the percentage of reused lubricants and reducing the CO<sub>2</sub> build up, which is an advantage from a point of view both economical and ecological. Waste lubricants are pretreated through a flash distillation, in order to remove water therefrom: they are fed into the column 2, undergo a flash process, normally with a temperature between 120 and 160 °C, preferably 140 °C, and a pressure ranging from 0.2 to 0.7 bar, preferably being 0.33 bar. These conditions allow the removal of a large amount of water from the waste lubricants, with the best yield. The water exiting the column 2 can be reused for cooling purposes within the same industrial premises or to produce steam; otherwise, it can be safely wasted in a way which is known per se.

**[0044]** The lubricants are then pre-treated through a

distillation, in order to remove bitumen and light distillates therefrom. In this view, column 6 is fed with the lubricants having left the column 2 and the furnace 5. Such lubricants arrive into the column 6 at a temperature ranging from 250 to 400 °C, preferably at 360 °C, and under a pressure ranging from 0.025 to 0.055 bar, preferably 0.04 bar. According to a preferred embodiment, the temperature in the head of column 6 is 50 °C and in the bottom 280 to 300 °C, while the pressure is around 0.027 bar in the head and 0.04 bar in the bottom. Three fractions leave the column 5. Light distillates, which are normally used within the plant to get energy through combustion, gasoline through the pipe 8 and bitumen through the pipe 9, which are byproducts which can be sold. The fraction leaving the column 6 through the pipe 10 is the one used in the subsequent steps of the process. This fraction is mixed with the shredded plastic wastes, lubricating them in the extruder 12. Namely, the melting unit includes an extruder 12, fed with pre-treated lubricants and shredded plastic wastes.

**[0045]** The wastes fed through the line 15 are made up by any kind of plastic waste. Some not limiting examples are: PE, PP, PS, PET, PVC, ABS and BR. To be clear: different kind of plastic wastes, made up by any kind of polymer in any respective ratio, can be fed together, with no need of separating them from each other. They are shredded together in the shredder 16 and fed to the extruder 12, where they admix with lubricants coming from the line 13. The possibility of stocking the pre-treated lubricants in the tank 11 can be very useful when the pre-treatment of lubricants and/or the treatment of the plastic wastes is performed in a batch fashion. Owed to this, the process according to this invention can be carried out continuously, batchwise or semicontinuously.

**[0046]** The waste/oil ratio within the extruder 12 is not particularly critical. However, a preferred ratio ranges from 75:25 to 95:5 wt.% and is preferably 90:10 wt.%. This mixture is molten within the extruder 12 and leaves the extruder 12 itself in the liquid state; such a liquid can be pumped in a relatively easy way. In order to keep liquid the molten material, the temperature within the extruder 12 ranges from 230 to 300 °C, preferably from 250 to 270 °C.

**[0047]** The choice of a gear pump 20 is due to the fact that the molten plastic material is rather viscous, and this kind of a pump is able to move even this kind of viscous liquids with no particular problem.

**[0048]** Also, this molten material can be temporarily stored in a tank 18, where it should be kept heated and stirred, in order to prevent an undesired solidification. The temperature within the tank 18 ranges from 270 to 280 °C. Insulation of the tank 18 is surely useful, if not even necessary.

**[0049]** The molten mass, before entering the hydrocracking reactor 25, is fed to the hydrofinishing reactor 24, in order to remove sulphur, nitrogen, metals and other impurities. When the pump 20 is operated, the liquid exits the tank 18 through the line 19, is further heated in the

heat exchanger 21 to a temperature ranging from 300 to 400 °C, preferably from 300 to 350 °C. The heated liquid is fed with hydrogen, coming from the inlet 23, under a pressure preferably ranging from 150 to 200 bar and is fed to the furnace 22. The heated liquid is fed to the reactor 24 at a temperature of 300-500 °C, preferably 350-450 °C. The reaction within the reactor 24 is aimed at removing metals and in hydrotreating the material. This step removes sulphur, nitrogen, aromatic carbon, chlorine and metals from the treated material.

[0050] The material leaving the reactor 24 through the pipe 26 is admixed again with hydrogen coming from the pipe 27, in order to have an appropriate quantity of reducing agent in the reaction mixture and is reacted in the reactor 25. The reactor 25 is in fact a hydrocracker -as it has already been mentioned-, operating with a catalyst conventionally used for hydrocracking processes, which breaks the molecular chains, transforming the polymers into hydrocarbons having a lower molecular weight, in fact depolymerising the charge. The product, leaving the reactor 25 through the line 28, is then fed to a distillation column 29, where it is distilled into a number of products, which are mostly valuable and can be sold profitably. The distillation takes place at a temperature which is around 50 °C at the head and 280-300 °C at the bottom, whereas the pressure is 0.0027 to 0.0040 bar at the head and 0.010 to 0.020 bar at the bottom.

[0051] As it has already been stated, outlet 30 downloads light distillates which, as the ones coming from the outlet 7, are normally used to produce energy for the plant by combustion. Outlet 31 downloads ultra -low sulphur diesel, outlets 32 to 34 lubricant bases and outlet 35 bitumen, as the outlet 9.

[0052] The process according to this invention allows to produce lubricants, diesel of high quality and bitumen, with virtually no waste, allowing to reuse lubricants which are usually not suitable for undergoing a regeneration process. The addition of the lubricants to the plastic wastes allows a better melting and to get a more homogeneous flow; it also helps in the depolymerisation step process does not need a preliminary step of separating from each other the plastic wastes, which results in a saving of costs and manpower.

[0053] It should be understood, anyway, that the invention should not be considered limited to the particular arrangement illustrated above, which is only an exemplary embodiment thereof, but that various variants are possible, all within the reach of the skilled person, without departing from the scope of protection of the invention itself, as defined by the appended claims.

#### LIST OF REFERENCES

##### [0054]

- 1 Feed
- 2 Pre-flash column
- 3 Head outlet

- 4 Tail outlet
- 5 Furnace
- 6 Distillation column
- 7 Outlet
- 8 Outlet
- 9 Outlet
- 10 Outlet
- 11 Tank
- 12 Extruder
- 13 Tube
- 14 Pump
- 15 Feed
- 16 Shredder
- 17 Line
- 18 Tank
- 19 Line
- 20 Gear pump
- 21 Heater
- 22 Furnace
- 23 Line
- 24 Reactor
- 25 Hydrocracker
- 26 Line
- 27 Line
- 28 Outlet
- 29 Column
- 30 Outlet
- 31 Outlet
- 32 Outlet
- 33 Outlet
- 34 Outlet
- 35 Outlet

#### Claims

1. Process for the conversion of plastic wastes, involving the depolymerisation of the plastic material making up the waste, wherein the plastic waste is extruded producing a molten mass which is mixed with hydrogen and fed to a hydrocracking reactor, **characterised in that** the waste plastic, upon extrusion, is mixed with lubricants which cannot be regenerated and the resulting molten mass contains both plastic and lubricant wastes.
2. Process as claimed in claim 1), **characterised in that** the lubricants are pre-treated through a flash distillation, in order to remove water and thinners therefrom.
3. Process as claimed in claim 1) or 2), **characterised in that** the lubricants are pre-treated through a distillation, in order to remove bitumen and light distillates therefrom.
4. Process as in any previous claim, **characterised in that** the molten mass, before entering the hydrocracking reactor, is fed to a hydrofinishing reactor,

in order to remove sulphur, nitrogen, chlorine, metals and other impurities.

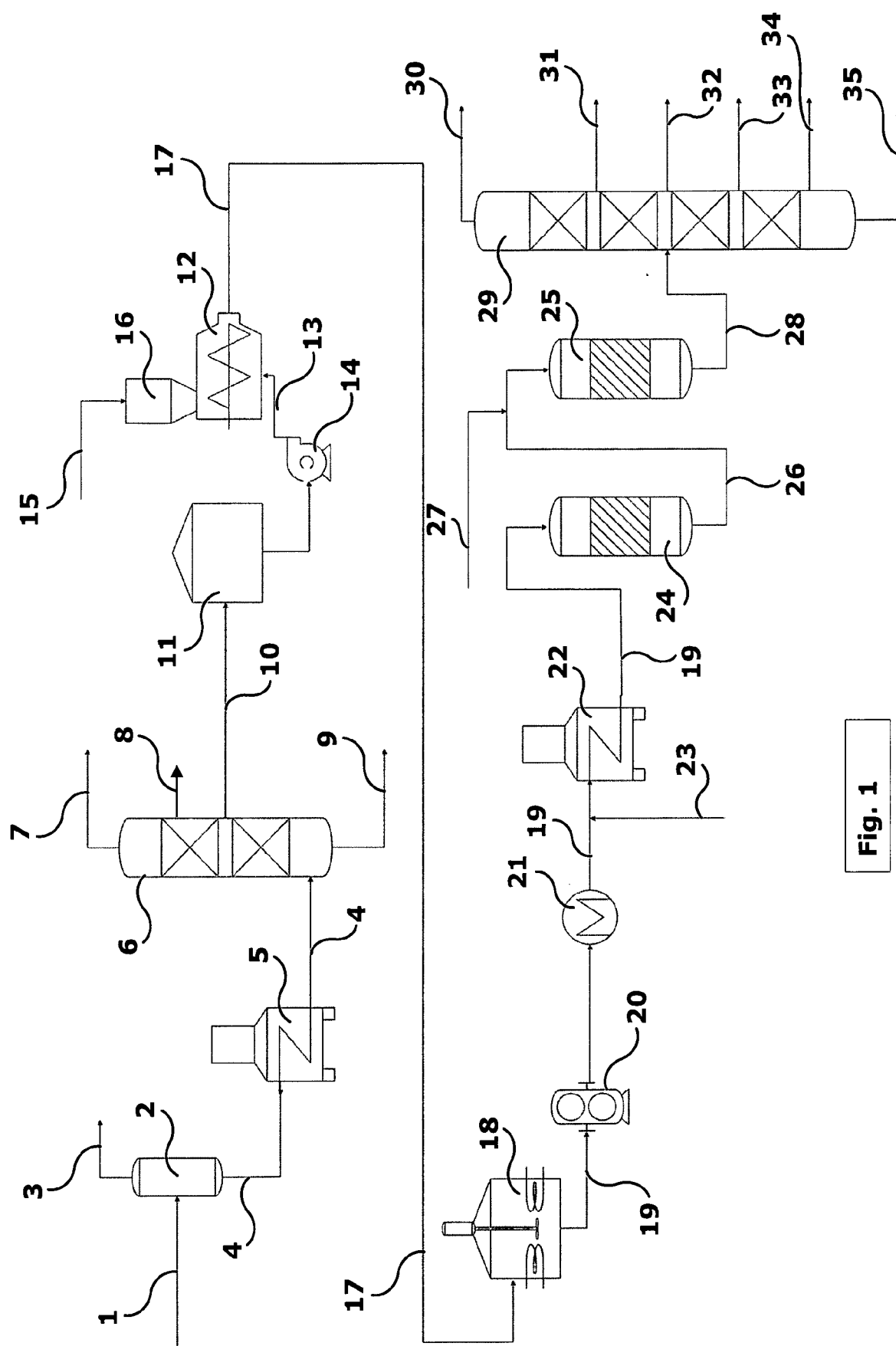
5. Process as in any previous claim, **characterised in that** it is carried out continuously. 5
6. Process as in any claim 1) to 4), **characterised in that** it is carried out batchwise or semicontinuously.
7. Process as in claim 6), **characterised in that** pre-treated lubricants and/or the molten mass of lubricants and plastic wastes are temporarily stocked in tanks before undergoing next steps. 10
8. Process as in claim 7), **characterised in that** the tank where the molten mass is temporarily stocked is heated and stirred, in order to prevent the solidification of the mass itself. 15
9. Process as in any previous claim, **characterised in that** the product leaving the hydrocracking reactor is distilled into a number of products. 20
10. Plant for carrying out the process according to any previous claim, **characterised in that** it comprises a unit (2; 6) for the pre-treatment of the lubricants which cannot be regenerated, a melting unit (12) for melting the plastic wastes and mixing them to the pre-treated lubricants and a depolymerisation unit (25). 25  
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11. Plant as claimed in claim 10), **characterised in that** the melting unit includes an extruder (12), fed with pre-treated lubricants and shredded plastic wastes. 35

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## EUROPEAN SEARCH REPORT

Application Number

EP 23 42 5035

## DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
X	WO 2020/129020 A1 (ENI SPA [IT]; VERSALIS SPA [IT]) 25 June 2020 (2020-06-25) * page 14, line 12 - page 24, line 2; claims; figures *	1-11	INV. C10G1/10 C10G47/00
A	----- CZESLAW KAJDAS: "Major pathways for used oil disposal and recycling. Part 1", TRIBOTEST, vol. 7, no. 1, 1 September 2000 (2000-09-01), pages 61-74, XP055133449, ISSN: 1354-4063, DOI: 10.1002/tt.3020070107 * the whole document *	1-11	
A	----- EP 3 960 838 A1 (RESONANTE LLC [US]) 2 March 2022 (2022-03-02) * claims *	1-11	
A	----- WO 2023/111975 A1 (ITELYUM REGENERATION S P A [IT]; MILANO POLITECNICO [IT]) 22 June 2023 (2023-06-22) * claims; figures *	1-11	TECHNICAL FIELDS SEARCHED (IPC) C10G
The present search report has been drawn up for all claims			
Place of search <b>The Hague</b>		Date of completion of the search <b>20 November 2023</b>	Examiner <b>Van der Poel, Wim</b>
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ..... & : member of the same patent family, corresponding document	



# **ANNEX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN PATENT APPLICATION NO.**

EP 23 42 5035

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This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on  
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		<b>WO 2020129020 A1</b>	<b>25-06-2020</b>
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For more details about this annex : see Official Journal of the European Patent Office, No. 12/82