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PERSPECTIVES IN KEROSENE PRODUCTION FROM *CAMELINA SATIVA* OIL

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Abstract

Global aviation industry is facing some significant challenges on reducing emissions of persistent greenhouse gases and on reducing 33% of total operating costs, represented by oscillating price of fuels. Thus, identifying new sources of biofuels is essential for the future of the industry. One promising research direction is provided by biofuels coming through processing oil, extracted from the seeds of Camelina sativa plant. Camelina sativa presents a lot of interest because of ecological plasticity, low-growing needs (low inputs, minimal nutrients) and the fact that it does not interfere with food resources of the population. The technology of obtaining biokerosen out of Camelina oil is partially similar to that of obtaining biodiesel. Camelina oil is initially subjected to a hydrodeoxigenation operation. Linear alkanes obtained are isomerized and are catalytic selective cracked, in order to obtain branched alkanes. These are more stable in aviation navigation areas, having lower boiling point than diesel biofuel. The last step is the catalytic closure of aromatic rings derived from existing paraffins. It can be avoided the expensive last step, by mixing the branched alkanes with conventional kerosene oil (which contains 25% aromatic hydrocarbons, in particular alkylbenzenes and alkylnaphthalenes). Also, by-products appear, such as natural pesticides, additives in the plastics industry, nutraceutical, forages etc. (alkanolamides, fatty alcohols, isopropyl esters, glycerol etc.). Advantages of using kerosene obtained from Camelina sativa oil are: the significantly low level of greenhouse gases coming from burning, compared with burning of fossil fuels, large base of unpretentious crops of Camelina sativa and highly pure biofuels, similar to fossil fuels. Disadvantages of using kerosene obtained from Camelina sativa oil are: high costs of technological equipment, low oil yields relative to plant production/hectare, large amount of hydrogen required, produced by high consumption of fossil energy and also, competition for the same resources as in the production of biodiesel .

Key words: *biofuels, Camelina sativa, greenhouse gases, kerosene, technology for obtaining kerosene.*

INTRODUCTION

Camelina sativa is a plant cultivated since ancient times (bronze and iron ages). It is not clear why since the Middle Ages, the crops of *Camelina sativa* were replaced by others.

The recent interest for this plant is supported by its reduced claims to a range of nutrients, claims which materialize into smaller amounts of inputs, but also in the ecological plasticity of the plant, which is capable to grow on half-arid soils, with low fertility (Zubr, 1997).

Certain features of *Camelina sativa* oil make it interesting for use as biofuel. Initially, as with other oilseeds, researchers efforts were directed towards obtaining biodiesel fuel.

In 2009, Japan Airlines and KLM Royal Dutch Airlines have successfully tested a mixture of aviation fuel, which contained 50% of a product derived from *Camelina sativa*.

In 2011, similar tests were performed by Honeywell and Boeing, Iberia company (using an A320 aircraft on the route Madrid - Barcelona) and Porter (Q400 aircraft on the route Montreal - Toronto) conducted similar tests on passenger flights.

The interest for the use of biofuels in aviation coming from *Camelina* starts from a series of studies according to which the products derived from this plant promise a decrease by 80% of the toxic impact over the environment (in particular by reducing greenhouse gas emissions, long cycle life), compared with their counterparts obtained from oil (Shonnard et. al., 2010).

In addition, the aviation industry is very vulnerable to world fluctuations in oil prices, fluctuations which are very common in the recent years. Prices vary depending on the geo-strategic policies of the great powers and the states in the Middle East, on the global financial market developments, factors that determine high volatility and instability. Considering that about 33% of the operational costs of the aviation industry are determined by fuel prices, fuel identification of additional resources is essential for the future of the industry.

In this paper we intend to conduct a study on the state of art regarding the potential use of *Camelina sativa* plant, in obtaining biofuels for aviation industry

MATERIALS AND METHODS

In order to accomplish this study were used data from specialized literature, and a series of official statistics (European Commission, reports of aviation companies etc.).

RESULTS AND DISCUSSIONS

Extensive studies concerning the technologies of culture and ecology of the plant *Camelina sativa* took place in several countries in the temperate regions of the world.

It was thus found, that although *Camelina* has reduced soil fertility claims, compared to the most oil-bearing crops, the treatments with nitrogen fertilizers affect the performance of the plant crop. The best results were obtained at additions of nitrogn fertilizers between 78.4 and 100.7 kg/ha, in Montana (USA) and 120 kg/ha in Germany (Eidhin et al. 2003; Frohlich and Rice 2005; Gilbertson et al., 2007; Shukla et al., 2002, Agegnehu and Honermeier 1997). Nitrogen fertilizers are critical for oil content in seeds, which decreases with the increase of nitrogen applied (Agegnehu and Honermeier 1997; Jackson 2008). This was revealed by a study conducted in Canada's maritime provinces by Urbaniak et al. (2008), that emphasized among others, the importance of choosing the variety of *Camelina*, for success or failure of the crop.

Camelina sativa responds to phosphorus fertilizer application, when its soil concentration exceeds 12 ppm (Jackson, 2008). Production per hectare reported by various researchers, is variable. Thus, in 2006 scientists from Huntley MT, obtained production of 1067 to 1093 kg/ha, at a sowing rate of 6.6 to 8.6 kg/ha. In Germany, Agegnehu and Honermeier (1997) reported a production of 2057 kg/ha for a sowing rate of 5.85 kg/ha.

Camelina sativa has reduced needs regarding pests control work. As potential pests we can mention *Phyllotreta cruciferae* (small fleas of cruciferous), which has a higher affinity for other cruciferous and *Peronospora camelinae* downy mildew. Some studies have shown that the *Camelina sativa* plant is competitive against weeds. In 1980, Lovett and Jackson have even suggested that *Camelina sativa* produces a series of allelopathy phytoherbicides, effective in growth stopping of *Linum usitatissimum* (flax).

The most important product of Camelina is oil, obtained by crushing and pressing the seeds. Oil content of seeds, reported to the dry matter, is ranging from 30 to 40% (Strasil, 1997). It is estimated that more than 50% of the composition of such oils is represented by polyunsaturated fatty acids. The contribution of the polyunsaturated fatty acids, in the composition of the oil, varies according to the applied phytotechny, but the main fatty acids identified are linoleic (18: 2) and α - linolenic acid (18: 3ώ3) (Eidhin et al., 2003). Most studies estimate that the content of erucic acid is less than 4% (Vollmann et al., 1996).

α - linolenic acid of *Camelina sativa* differs from its counterparts, extracted from other plants, by greater stability to oxidation. Wastes of oil extraction are an excellent source for animal forages, containing more than 5% α – linolenic acid. US Food and Drug Administration approved in 2009 the introduction of such forages in the chickens and cattle nutrition, in an amount up to 10% from the total ration.

Although polyunsaturated fatty acid composition makes interesting the *Camelina* oil for food consumption (source of omega 3 unsaturated fatty acid), must be considered that its stability is lower than usual edible oils from rapeseed, olive, corn, sesame or sunflower (Matthäus et al., 2004).

The fact that *Camelina* oil does not present popularity as food, makes it an interesting resource as biofuel, because it does not compete with crops for human consumption.

One of the most common fears, regarding the cultivation of certain plants for biofuel production, is the fact that most of the proposed plants have some agronomic traits, which can turn them into invasive plants (drought resistance, tolerance to soil fertility conditions, short cycle development, rapid accumulation of biomass etc.) (Raghu et. al. 2006; Barney and DiTomaso 2008). These native qualities can become overwhelming, in case these plants are widely cultivated (Minton and Mack, 2010). Although researches on invasive potential of *Camelina sativa* are limited, due to the high variability of the factors that must be taken into account, the findings of the current studies tend to evaluate this risks as being rather low (Davis et al., 2011). The quality of *Camelina sativa* seeds is significantly influenced by both phenotype and genotype factors. Vollman et al. (2007) showed that there is a significant number of genotypes of *Camelina sativa*, which can promise the simultaneous selection of superior qualities cultivars, both in terms of production achieved and seed oil content.

However, this selection does not have to take into account the increase in the size of the seeds, since this parameter correlates negative very significant both with total oil content, and the concentration of certain fatty acids (linolenic acid). This production sensitivity to phenotypic factors showes that the estimates regarding the amount of oil / hectare from Camelina crop, varies significantly from author to author, depending on geographic area of reference. In any case, most of the studies place the oil yield / ha of *Camelina*, under the oil yield / ha of rapeseed, over the oil yield / ha of soybeans and within range of variation of the sunflower crop (Table 1).

Table 1. Comparison between yields of different oilseeds (after Oil World Annual 2009, quoted by Moser, B., R., 2010)

Specification	**Camelina**	**Rapeseed**	**Soybean**	**Sunflower**
Production (t/ha)	0.9 – 2.24	2.68 – 3.39	2.14 – 2.84	1.44 – 1.70
Oil content (%, dry matter)	35 – 45	40 – 44	18 – 22	39 – 49
Oil yield (l/ha)	106 - 907	965 - 1342	347 - 562	505 - 750

The main constituent of *Camelina* oil fatty acids is linolenic acid (32-40% of dry weight), followed by linoleic, oleic and 11 - eicosenoic acids (Table 2).

Table 2. Fatty acids composition of Camelina oil (%), according to various authors

Fatty acid (No. of C)	**Authors**			
	Leonard (1998)	**Moser şi Vaughn (2010)**	**Zuhr şi Malthaus (2002)**	**Frohlich şi Rice (2005)**
Palmitic (16:0)	5.3	6.8	5.4	5.4
Stearic (18:0)	2.5	2.7	2.5	2.6
Oleic (18:1)	12.6	18.6	14.9	14.3
Linoleic (18:2)	15.6	19.6	15.2	14.3
α – Linolenic (18:3)	37.5	32.6	36.8	38.4
Arachidic (20:0)	1.2	1.5	1.3	1.4
11– Eicosenoic (20:1)	15.5	12.4	15.5	16.8
Eicosadienoic (20:2)	2.0	1.3	1.9	-
Eicosatrienoic (20:3)	1.7	0.8	1.6	-
Behenic (22:0)	0.3	0.2	0.3	0.2
Erucic (22:1)	2.9	2.3	2.8	2.9
Others	2.9	1.2	1.8	3.7

Initially, as with other oilseeds, researchers efforts were directed towards obtaining biodiesel fuel. Biodiesel is defined by the American Society for Testing and Materials (ASTM) as a mixture of monoalkyl esters of long chain fatty acids and is usually obtained by transesterification of the lipids, in the presence of an alkaline catalyst and an excess of methanol, at high temperature (600^0 C).

Another way of obtaining biodiesel from lipids, derived from biological materials, involves their hydrodeoxigenation, under pressure and high temperatures (40-150 atm, $350\text{-}450^0$ C), in the presence of hydrogen and heterogeneous catalysts. It is obtained a mixture of paraffins (linear alkanes with different carbon chain lengths), that are subject in the next stage, to isomerization operations. Isomerization serves to improve the flow properties to cold and also significantly decrease cetane number. The resulting mixture is made up of paraffins with 15-18 carbon atoms, and properties similar to the compounds obtained from counterparts oil.

There are several directions of research to obtain biofuels for aviation. These include starchy crops, lignocellulosic biomass (wood crops, residues from agriculture or forestry, algal biomass or municipal wastes) and oilseed crops. The conventional kerosene is a mixture of hydrocarbons consists of molecules that usually contain from 8 to 16 carbon atoms, obtained by refining crude oil. This must correspond to very strict standards related to safety in the aviation industry and to some essential goals, namely: providing a large amount of energy per unit of mass or volume; stability at low temperatures, to avoid freezing or gelling in thermic conditions existing at aircraft flying altitudes; compatibility with the materials used in aircraft.

In accordance with these objectives, kerosene must meet criteria that are related to a number of physico-chemical parameters, such as viscosity, surface tension, volatility, lubricity, sulfur amount, combustion properties, etc. The technical characteristics of kerosene, used in aviation industry, are defined by ASTM D1655 standards in United States or DEF STAN 91-91 in Europe (Table 3)

Table 3. Main technical specifications for kerosene used in aviation, in American standards (ASTM D 1655) and European (DEF STAN 91-91)

Fuel	**Jet A**	**Jet A - 1**
Standard	**ASTM D 1655**	**DEF STAN 91 - 91**
Acidity (mg KOH/g)	0.10	0.015
Aromatics (% vol, max.)	25	25
Sulphur, total (% wt)	0.30	0.30
Mercaptan (% wt)	0.003	0.003
Distillation temperature (^{0}C):		
Initial boiling point	-	Report
10% recovery, max	205	205
50% recovery, max	Report	Report
90% recovery, max	Report	Report
Final boiling point	300	300
Flash point (^{0}C, min)	38	38
Density (15^{0}C, kg/m^3)	775 - 840	775 – 840
Freezing point, (^{0}C, max)	- 40	- 47
Viscosity (- 20^{0}C, mm^2/sec, max)	8	8
Specific energy (MJ/kg, min)	42.8	42.8
Smoke point (mm, min)	18	19
Naphtalenes (vol %, max)	3	3
Copper strip (2h, 100^{0}C, max.)	1	1
Thermal Stability		
The pressure drop limit for the filter (mm Hg)	25	25
Tube deposit ratting (max)	< 3	< 3
Gum content (mg/100 ml, max)	7	7

Modern fuels used in aviation industry must satisfy a number of criteria associated with: efficiency, reliability and sustainability. Also, modern fuels must not affect the infrastructure in the field (Andriishin et. Al., 2010). Thus, they must: be supplied in good conditions and in constant quantity; have a good pumpability in different conditions of temperature or altitude; evaporate completely in the combustion chamber and burn in a wide range of variation of the fuel / air mixture; have high heat capacity, high density capability to ensure stable combustion; not contain mineral compounds, have no tendency to form soot or other specific deposits at high or low temperatures; not to be corrosive for aircraft components, to be compatible with non-metallic materials; have good anti-wear properties, for protection and cooling; have large storage life and stability to shipping; have low toxicity both for humans and environment.

Technology of obtaining biokerosen from *Camelina* is partial similar to obtaining biodiesel by hydrodeoxigenation of lipids. But isomerization operation is critical, since branched paraffins aims to achieve. The sequence of reactions required to obtain kerosene is summarized in Figure 1.

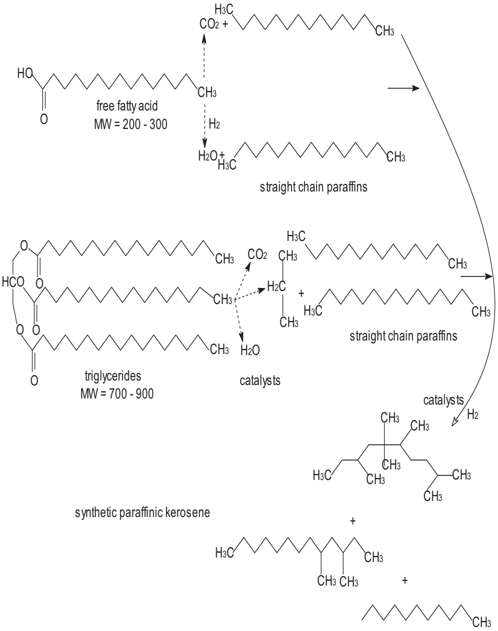

Figure 1. The sequence of chemical reactions for obtaining vegetable oil kerosene

The first reaction involves removing oxygen, with obtaining of linear paraffins, specific for biodiezel. Then, the cracking takes place, to obtain highly branched paraffins, similar to those components of synthetic kerosene.

In Figure 2 is shown a simplified scheme for industrial production of biokerosen from *Camelina* oil. In the first stage, *Camelina* oil is subjected to a hydrodeoxygenation process (HDO), whose result is obtaining linear alkanes.

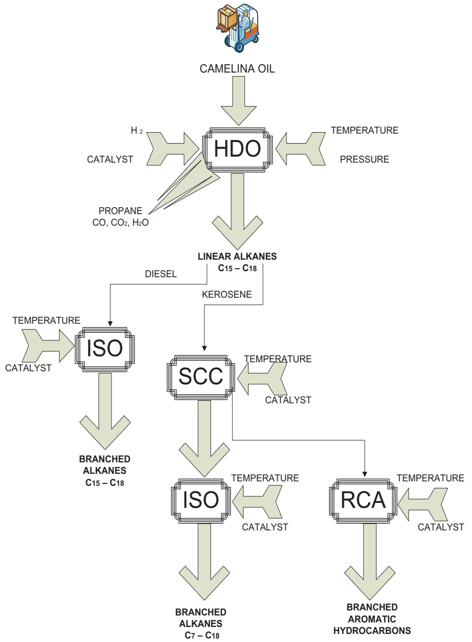

Figure 2. Simplified diagram for obtaining biokerosen of *Camelina sativa* oil (after Moser, 2010)

In second stage, occurs isomerisation (ISO) of linears alkanes to branched alkanes. This process is necessary, as linear alkanes freeze at temperatures encountered in specific areas of air navigation.

The third stage involves selective catalytic cracking (SCC) of the paraffins to shorter-chain hydrocarbons (C7–C18). The aim of this process is to obtain a mixture with a boiling temperature range lower than that of diesel biofuel.

With existing catalyst and reactor technologies, selective catalytic cracking and isomerization can be accomplished in a single stage.

Finally, the last step is the catalytic closure of the aromatic rings, for a part of existing paraffins, operation which for economic reasons may be replaced with conventional kerosene, mixed with biodiesel obtained from oil (biodiesel contains 25% aromatic hydrocarbons, especially alkylbenzenes or alkylnapthalenes).

Some studies have focused on improving the rate of transesterification of the fatty acids from *Camelina* oil. Thus, Patil et al (2011) have achieved a higher rate of production of fatty acid methyl esters (FAME) from *Camelina* oil, coupling catalysts based on metal oxides with heat treatment. With this treatment, two-fold higher yields of fatty acids methyl esters were obtained, using the catalyst of BaO and SrO and microwave heating, compared to conventional treatments. It was observed that BaO and SrO catalysts generated higher FAME yields than the CaO and MgO catalysts. A comparison between the conventional heating and the microwave-assisted transesterification processes, showed that the reaction rate constant obtained in the microwave-assisted transesterification process, are of two orders of magnitude higher than those obtained with the conventional heating method.

In Table 4 is shown the profile of fatty acid methyl esters identified in biokerosene obtained by methanol transesterification of *Camelina* oil (after Llamas et al., 2012; Matas et al., 2012).

Table 4. Profile of methyl esters of fatty acids identified in biokerosen, obtained by transesterification with methanol of *Camelina* oil (after Llamas et al., 2012)

Ester metilic	**CAM 100**
Methyl caprylate C8:0	-
Methyl caprate C10:0	-
Methyl laurate C12:0	0.84
Methyl miristate C14:0	0.41
Methy palmitate C16:0	5.83
Methyl stearate C18:0	2.97
Methyl oleate C18:1	15.83
Methyl linoleate C18:2	18.85
Methyl linolenate C18:3	33.99
Methylarachidate C20:0	1.54
Methyl-cis-11-eicosenoate C20:1	16.03
Methyl behenate C22:0	2.16
Methyl eructate C21:1	1.44
Iodine value	148.8
Mean formula	$C_{19.23}H_{34.97}O_2$
Molecular weight (g mol^{-1})	298.153
Stoichiometric air/fuel ratio	12.508
C (%)	77.45
H (%)	11.82
O (%)	10.73

(CAM = percent of *Camelina* oil)

The results of Llamas et al. (2012), in terms of biokerosen main physical - chemical parameters, obtained by transesterification with methanol of *Camelina* oil, but also with mixtures of *Camelina* oil and various proportions of conventional kerosene, led to the conclusion that only a mixture of 10% biokerosen and 90% conventional kerosene is feasible for use in the aviation industry (Table 5).

Table 5. Physico-chemical characteristics of the biokerosen obtained by transesterification of Camelina oil with methanol and various mixtures with conventional kerosene (as Llamas et al., 2012)

Parameter / Blends	**CAM 0**	**CAM 5**	**CAM 10**	**CAM 20**	**CAM 100**
	K1 100	**K1 95**	**K1 90**	**K1 80**	**K1 0**
Color and aspect	clear	clear	clear	clear	clear
Elemental composition (%, wt)					
C	85.9	85.29	8.23	83.86	77.09
H	13.88	12.99	1.,61	12.45	11.09
N	0.02	0.02	0.02	0.02	0.02
S	0.17	0.05	0.03	0.03	0.06
O		1.65	3.11	3.64	11.74
Another properties					
Density (15^{0}C, kg/m^{3})	793	806.7	810.8	819.8	889.3
Viscosity (45^{0}C, mm^{2}/s)	-	1.2591	1.3075	1.4539	4.3395
Viscosity (100^{0}C, mm^{2}/s)	-	0.6799	0.7053	0.7663	1.7077
Viscosity(-20^{0}C, mm^{2}/s)	-	3.11	3.20	3.80	6.96
Higher heating value (MJ/kg)	47.7	45.3	45.0	44.8	40.0
Lower heating value (MJ/kg)	44.44	42.52	42.30	42.14	37.71
Cold filter plugging point (^{0}C)	-	-31	-29	-26	-5
Cloud point (^{0}C)	-42.5	-26	-26	-22	1
Pour point (^{0}C)	-	-40	-32	-31	-5
Oxidative stability (hours)	-	>8	> 8	> 8	1.4

(CAM = percent of *Camelina* oil)

In addition to the products of interest resulted from the process of biokerosen obtaining, a number of other substances and residues can be exploited in different directions. This is a significant hope for economic profitability of the technology.

By-products can be used as: natural pesticides, additives in the plastics industry, nutraceuticals, forages and so on (alkanolamides, fatty alcohols, isopropyl esters, glycerol etc.).

It is estimated that the presented technology has a number of advantages, such as: obtaining products with a level of greenhouse gas emissions significantly lower, compared with fossil counterparts; large base of crops that can be processed in this direction and obtaining biofuels of high purity and chemical composition similar to that of fossil fuels. But the technology presents a number of disadvantages, which include: high costs of raw materials procurement and technological facilities, low yields relative to oil production/hectare of crops, competing for the same resources used in the production of biodiesel, high necessary of hydrogen produced today by a significant consumption of fossil energy.

Table 6 presents a summary of key aspects of technologies for obtaining biokerosen, currently proposed: Kerosen Parafinic Sintetic with hydrotreated esters and fatty acids (HFA-SPK), Kerosen Parafinic Sintetic Fischer - Tropsch (FT – SPK) and Kerosen Parafinic Sintetic from alcohol (ATJ – SPK). These technologies belong to the findings of a study by SINTEF ENERGI AS (Norway) for AVINOR, completed in 2012.

Table 6. Comparative analysis of the main constituent elements of the proposed technologies for obtaining biokerosen (SINTEF Energi I study for Avinor, 2012)

	HEFA - SPK	**FT-SPK**	**ATJ-SPK**
Feedstock	**Conventional oil crops**: soybean, palm oil, rapeseed, Coconut, corn; **New oil crops:** jatropha, camelina și halophyte; **Microalgae**	**Lignocellulosic biomass**: energy crops, agricultural and forestry residues, wastes	**Sugars**: sugarcane, sugar beets, molasses, and fruits **Starches:** corn, cassava, potatoes, and root crops **Lignocellulosic biomass**: energy crops, agricultural and forestry residues
By-products	Diesel, fractions of propane, naphtha and LPG, natural pesticides, nutriceuticals plastics, animal feed, heat and chemicals	Diesel, gasoline, naphtha, chemicals (hydrogen, methanol,)	**From sugars and starches**: diesel (from alcohol production), Proteins and fats (from jet-fuel production); **From lignocellulosic biomass**: diesel (from alcohol production), lignin and small amounts of proteins (from jet-fuel production)

	HEFA - SPK	FT-SPK	ATJ-SPK
Costs	Low CAPEX, High OPEX: high feedstock prices, low yields (little oil content in the crop), large hydrogen requirement	High CAPEX (gasification, gas cleaning and FT steps), Low OPEX (use of residues as feedstocks, high conversions)	Low CAPEX High OPEX: micro-organisms and pre-treatments
Certification	Certification since July 2011 by the ASTM D1655 standard, up to 50% blending with petroleum- based jet kerosene	Certification since September 2009 by the ASTM D1655 standard, up to 50% blending with petroleum-based jet kerosene	Under ASTM certification. Expected to be approved as fully synthetic aviation fuel with 100% replacement of the petroleum-based jet kerosene by 2014
Commercialization	Pilot plants under construction	Pilot plants under construction	Pilot plants under construction
Advantages	Wide range of feedstocks can be processed; Product life cycle emissions significantly lower compared to fossil fuels (80-85% including only biomass conversion processes); Very pure and high quality product with a chemical composition similar to conventional jet-fuel	Wide spectra of potential products; Large feedstock flexibility; Product life cycle emissions much lower compared to fossil fuels (90-95% including only biomass conversion processes); High conversions; Relatively low external hydrogen requirement when applying certain gasification systems (indirect gasification)	All steps necessary to convert alcohol to jet-fuel are at commercial scale in the petrochemical industry; Large feedstock flexibility; ATJ-SPK does not require blending with petroleum-derived jet-fuel; Little amount of external hydrogen required; High specificity when processing biomass to alcohols through fermentation
Challenges	High investment cost of the plants; High feedstock prices; Feedstock availability (competing with biodiesel producers for the same feedstock); Sustainability concerns; Low oil yields; Large amounts of hydrogen required	High capital costs; Biomass gasification still requires optimization, particularly with regards to tar minimization; Large amounts of hydrogen required	High alcohols production costs, particularly from lignocellulosic biomass; Limited experience with alcohols other than methanol/ethanol; Low production rates when working with microorganisms; High sensitivity of microorganisms towards impurities;

* CAPEX, Capital Expenditure, refers to investment expenditure required, ie all those costs that are amortized over the whole life of the investment;

** OPEX, operating expense refers to expenses necessary to support current activities

CONCLUSIONS

Since the technology for obtaining kerosene from vegetable oils, by transesterification, has already been certified, it is expected that its implementation will take place in the shortest time. There are a number of uncertainties that could influence the evolution of this technology, such as oil price relative to the price of the main raw materials of oilseeds.

It is expected that a decrease in the price of oil, or an increase of the raw materials of oilseeds, to limit investors enthusiasm. On the other hand, new technologies are proposed and enthusiastically embraced by researchers, as getting kerosene from microalgae.

Given the fact that *Camelina sativa* plant is not part of the crops which interfere with food resources of the population, its use for obtaining biokerosen may confer an advantage in discussed issue.

In addition, as we have shown, the technology for obtaining kerosene from oil plants can be improved, in terms of economical profitability, by increasing the use of the byproducts, which occur in the main reaction pathways of this technology.

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