Contribution of biodiesel fuel to environmental pollution

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This study offers an analysis of life cycle energy efficiency indicators of fatty acid methyl esters, their biodegradability in natural environment and of the concentration of toxic components in engine emissions. The life cycle energy efficiency indicator (R_{1}) of rapeseed oil fatty acid methyl esters (RME) depends directly on rapeseed productivity. When the rapeseed productivity is 1.79 t/ha, the energy consumption of RME production is higher than the energetic value of RME (the value of the life cycle energy efficiency indicator is lower than 1),thus RME cannot be attributed to renewable fuels. The fatty acid methyl esters produced from fatty waste are characterized by higher values of the life cycle energy efficiency indicator as compared with RME. Energy is used in the most efficient way when fatty acid methyl esters are produced from waste of animal origin – pork lard (PME) and beef tallow (TME). The value of $R₁$ is 3.17. Linseed oil fatty acid methyl esters (LSME) are characterized by a lower value of the life cycle energy efficiency indicator (R_1 – 2.6). When fatty acid methyl esters produced from waste fat and oil are used to make tri-component fuels (RME–LSME–TME, RME–LSME–PME), the value of the life cycle energy efficiency indicator is increasing proportionally to their content in the mixture. This indicator exceeds 1 even when the rapeseed productivity in Lithuania is moderate. Emissions of the tricomponent fuel mixtures RME90–LSME2–PME8, RME80–LSME4–PME16, RME70–LSME6– PME24, RME90–LSME2–TME8, RME80–LSME4–TME16, RME70–LSME6–TME24 differ not much from those of pure RME. Only the concentration of polycyclic aromatic hydrocarbons in exhaust gases decreased down to 57% at 1200 min⁻¹ when the mixture RME-LSME-TME was tested. The requirements of biodegradability were met by all the test fatty acid methyl esters and their mixtures. Of them, 90–98% degraded in 21 days, while in the case of fossil diesel fuel the biological decomposition reached only 57.3% during the same period. The biodegradability somewhat lower in mixtures that contain more rapeseed oil fatty acid methyl esters.

Key words: biodiesel, life cycle, biodegradability, oxide emissions

INTRODUCTION

Making and using the products may have negative effects on the environment, and this should be taken into account. One of the methods to evaluate the product influence on the environment is its life cycle analysis – assessment of the product's effect on the environment through the analysis of potential environmentadverse factors related to the product, starting with the acquisition of the raw materials and up to the utilization of waste. Fuel is considered to be a renewable energy resource if its $R₁$ (life cycle energy efficiency indicator) value is higher than 1 . The $R₁$ of fossil diesel fuel is 0.885 (Poitrat, 1993). The energy efficiency indicator of rapeseed oil fatty acid methyl esters (RME), which are usually used as biofuel, has been evaluated by researchers in various countries (Batshelor, 1995; Boo, 1993). Its values vary depending on the climatic conditions, rapeseed productivity, and production technologies. In order to use energy more effectively and to reduce pollution by waste, methods to use certain new materials for the production of biodiesel fuel are being searched for. Lithuania could use animal waste for this purpose; part of such waste is now being utilized by the JSC "Rietavo veterinarinė sanitarija" (Rietavas Veterinary Sanitation, Ltd.) by drying it and by incineration of technical fats. This process does not produce any additional energy, and a gas that causes the greenhouse effect is emitted. Furthermore, no useful product is made. The rest part of such kind of waste becomes a source of environmental pollution. Another potential waste material that could be rationally used for the production of biodiesel fuel is fiber flax linseed oil. It is not suitable for food because large amounts of various plant protection substances are used while growing technical flax.

With regard to the environmental protection, a positive property of biodiesel fuel is its quick biodegradability under natural conditions. A fuel is considered easily biodegradable if no less than 90% of it degrades in 21 days. There are several methods to analyse biodegradability (Industrial use of rape for biodiesel in Czech Republic, 1995, EPA (560/6-82-003), Lyman et al., 1990). Biodegradability in an aqueous environment may be determined by measuring the release of carbon dioxide $({\rm CO_2})$ (EPA, 1982) and applying the method of gas chromatography

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The direct effect of fuel on the environment is determined by analysing engine emissions. The Lithuanian normative documents regulate only the smokiness of the exhaust gases of diesel engines. However, the experience of other countries shows that it is also important to determine the content of CO , NO_x and soot in exhaust gases (Tschöke et al., 2002). The amount of soot is especially important because the polycyclic aromatic hydrocarbons and nitrogenous aromatic hydrocarbons accumulate on soot. Exhaust gases of diesel engines may contain more than 100 compounds characterized by carcinogenic and mutagenic effects (Jacob et al.,2003).

The survey of literature did not result in any data on the evaluation of environmental pollution (results of life cycle analysis, evaluation of engine emissions and fuel biodegradability) when fatty acid methyl esters produced from fatty waste are used.

The aim of this work was to determine the life cycle energy efficiency indicators of fatty acid methyl esters, to evaluate engine emissions and biofuel biodegradability.

To this end, the following tasks were solved:

• to determine the life cycle energy efficiency indicators of rapeseed oil fatty acid methyl esters, taking into account the productivity of rapeseed;

• to determine the life cycle energy efficiency indicators of fatty acid methyl esters produced from fatty waste, and of their mixtures with rapeseed oil fatty acid methyl esters;

• to analyse the composition of exhaust gases when the engine is fuelled with fatty acid methyl esters of different origin and their mixtures;

• to analyse the biodegradability of fatty acid methyl esters of different origin and of their mixtures in natural environments.

MATERIALS AND METHODS

To calculate the value of the life cycle energy efficiency indicator R₁ of fatty acid methyl esters (FAME), it is necessary to take into account energy consumption for the preparation of raw materials and during their processing into biodiesel fuel. When waste fat and oil are used for biodiesel fuel production, energy consumption for the preparation and remaking of waste into biofuel is taken into account.

Energy consumption in agriculture needed to produce 1 t of rapeseed oil fatty acid methyl esters (RME) is calculated with regard to direct (petroleum products and electricity) and indirect (energy accumulated in fertilizers, chemicals, tractors and agricultural machinery) energetic inputs. The amount of energy accumulated in agricultural machinery as well as fuel consumption in agriculture were calculated according to Tariffs of Mechanized Agroservice Works Pricelist (Mechanizuotų…, 1992). Energy accumulated in fertilizers and plant protection materials was calculated according to the norms recommended in R. Velička's "Rapsai" (2002) and in the KEMIRA catalogue and to energetic equivalents of the chemical substances used.

While assessing the life cycle energy efficiency indicator of fatty acid methyl esters produced from waste fat, we calculated energy consumption for all the production processes.

Energy consumption for rapeseed, linseed, and pork lard and beef tallow fatty acid methyl esters production was calculated on the basis of the technology and equipment specification used by the JSC "Rapsoila".

The life cycle energy efficiency indicator $(R₁)$ of rapeseed oil fatty acid methyl esters (RME) and linseed oil fatty acid methyl esters (LSME) was calculated using the following equation:

$$
R_{\scriptscriptstyle 1} = \frac{E_{\scriptscriptstyle c}}{E_{\scriptscriptstyle a\!g}} + E_{\scriptscriptstyle p}}\,,
$$

where: E_c – energy accumulated in RME or LSME (calorific value), MJ/t of biofuel;

 E_{aa} – energy consumption for oilseed plant growing and seed preparation for oil pressing, MJ/t of biofuel;

 E_p – energy consumption for oil pressing and transesterification, MJ/t of biofuel.

The life cycle energy efficiency indicator $(R₁)$ of waste fatty acid methyl esters (FAME) produced from fatty waste (pork lard and tallow) was calculated by the following equation:

$$
R_{\scriptscriptstyle 1} \, = \frac{E_{\scriptscriptstyle c}}{E_{\scriptscriptstyle w} \ + \ E_{\scriptscriptstyle p}},
$$

where: E_c – energy accumulated in FAME (calorific value), MJ/t of biofuel;

Ew – energy consumption for waste fat preparation (drying, filtration), MJ/t of biofuel;

E ^p – energy consumption for fat esterification and transesterification, MJ/t of biofuel.

To study engine emissions, an experimental one-cylinder direct-injection AVL-type engine 502.019 was used. Experiments were accomplished at the Coburg University of Applied Sciences (Germany). NO_x , CH, CO and CO₂ emissions in exhaust gases were determined using an AMA exhaust gas analyser (Pierburg Instruments). Polycyclic aromatic hydrocarbons (PAH) were analysed applying two methods: induction laser fluorescence spectrometry (LIF, Optimare) and high performance liquid chromatography (HPLC, Varian).

Biological decomposition of biodiesel fuel in water was determined applying the CEC L-33-T-82 method. Bacterial substrate for experiments was obtained from JSC "Kauno vandenys", sampled after the first wastewater cleaning (mechanical) stage. Samples after extraction were analysed using the infrared spectroscopic method at the KTU Chemical Engineering Department, using an FT-IR Spectrum GX spectrometer (Perkin-Elmer).

RESULTS AND DISCUSSION

To calculate the life cycle energy efficiency indicator R_1 of fatty acid methyl esters (FAME), it is necessary to take into account energy consumption for the preparation and processing of raw materials into biofuel.

The JSC "Rapsoila" produces 1 t of RME from 2.9 t of rapeseed. In order to calculate the dependence of energy consumption on rapeseed productivity, we chose different productivities of rapeseed: the optimistic with the highest yield of 2.8 t/ha (productivity in big farms), and the pessimistic one – 1.79 t/ha.

The energy consumption in agriculture for the production of 1 t of RME was calculated taking into account direct (petroleum products and electricity) and indirect (energy accumulated in fertilizers, chemicals, tractors and agricultural machinery) energetic inputs. Different tractors and agricultural machines most common in Lithuania were chosen to calculate the indirect energy inputs. In

the further calculations we used the average values. The energy accumulated in fertilizers and plant protection materials is shown in Table 1. The largest energy demands are related to fertilizers.Energy accumulated in fertilizers makes up more than 75% of total energy accumulated in fertilizers and plant protection materials.

To calculate the energy consumption for oil pressing and transesterification, a technological scheme of the production of biodiesel fuel used by the JSC "Rapsoila" was applied. The cold pressing method is used for oil extraction from rapeseed at this enterprise.

Figure 1 shows energy consumption for individual production processes (oil pressing and transesterification) and energy accumulated in the chemical substances used for fatty acid methyl esters production. The total energy demand to make one tonne of RME equals to 14349.8 MJ. The oil pressing procedure requires the highest energetic inputs – 6406.7 MJ.

Table 2 presents total energy consumption for 1 t of RME production in the cases of moderate and higher rapeseed productivity in big Lithuanian farms.

The obtained data imply that the largest energy demands are related to agriculture but not to the stages of oil pressing and transesterification. Most of energy input (30.2–36.2%) is presented by the energy accumulated in fertilizers and chemical materials used in agriculture. The energy accumulated in chemical materials used for transesterification is lower and reaches 8.7–11.2.3%.

The total energy consumption for RME production is inversely proportional to rapeseed productivity. When rapeseed fertility is 1.79 t/ha, the total energy consumption is bigger by 8815.9 MJ/t RME if compared to the energy consumption at the rapeseed productivity 2.8 t/ha. It is determined by a higher energy demand in agriculture for growing the same amount of rapeseed required for 1 t of RME production (2.9 t).

Fig. 1. Energy consumption for 1 t of RME production

As animal fat in this case is a waste material, energy consumption for its production was not calculated. Before transesterification, animal waste should be filtered (solid particles are eliminated) and dried because moisture stops the transesterification reaction. The acidity of the conventional raw material (rapeseed oil) used in the biodiesel fuel (RME) production cannot be exceed 2%. When the acidity of fatty waste is higher, before the traditional alkaline transesterification it is necessary to esterify free fatty acids, i. e. the production process is supplemented by one more stage of esterification, employing acid catalysts.

To produce one tonne of the fatty acid methyl esters of animal origin, 1075 kg of waste (pork lard and beef tallow) fats are needed, the moisture content of which is about 2.6%. Energy consumption for 1 t of methyl esters of animal origin production is 11907.55 MJ. For waste filtration 0.45 MJ, for drying 90.8 MJ and for esterification 3873.2 MJ are used, while transesterification needs similar energy amount as the transesterification of rapeseed oil (Table 2).

Figure 2 shows how the energy demand (%) is distributed in the case of the production of fatty acid methyl esters from fatty waste. The highest energy demands are allocated to the energy accumulated in chemical substances used for esterification and transesterification. Waste fat preparation for esterification (filtering and drying) needs quite little fossil energy (0.76% of total energy consumption) whereas the additional esterification stage of waste fat needs 3873.2 MJ/t of energy (32.8% of total consumption). Nevertheless, the total energy consumption for FAME production from fatty waste is 2.5–3.2 times lower as compared with energy consumption for RME production.

As flax in Lithuania is grown for textile industry, linseed here is not suitable for food due to its contamination by plant protection chemicals, so it is considered as a waste material. Thus,when energy consumption for linseed fatty acid methyl esters (LSME) production is calculated, only oil pressing and transesterification were taken into account. The transportation costs were also not assessed, either, as linseed is waste. So, it made 14349.8 MJ/t LSME and exceeded a little energy consumption for FAME production from animal waste, but it was sill twice lower than energy consumption for RME production.

The following are the values of energy accumulated in fatty acid methyl esters of different origin (calorific values) (Winne,1998):

- RME (rapeseed oil fatty acid methyl esters) 37.77 MJ/kg;
- TME (beef tallow fatty acid methyl esters) 37.25 MJ/kg;
- PME (pork lard fatty acid methyl esters) 37.25 MJ/kg;

• LSME (linseed oil fatty acid methyl esters) – 37.31 MJ/kg. Finally, we calculated the values of life cycle energy efficiency indicators of different fatty acid methyl esters: for TME and PME R_1 – 3.17, for LSME R_1 – 2.6. This allowed a conclusion that the methyl esters produced from fatty waste meet the requirement set for renewable energy resources ($R_1 > 1$). The value of R_1 for RME is above unity only when rapeseed productivity is 2.8 t/ha. When rapeseed productivity is 1.79 t/ha (average productivity in Lithuania), RME cannot be attributed to renewable fuels.

We also investigated the conformity of mixtures of animal and vegetable methyl esters to requirements of the standard EN 14214, and have found that pure TME, PME and LSME do not satisfy certain requirements regarding the quality of biodiesel fuel. We determined optimal concentrations of RME–LSME–

LME mixtures which by the iodine value, linolenic acid content and oxidation stability met the requirements of the above-mentioned standard (Janulis et al., 2005; Sendzikiene et al., 2005). The life cycle energy efficiency indicators were calculated for the mixtures that could be used as a biofuel (Table 3).

The $\mathrm{R}_{\text{\tiny{l}}}$ values of all the analysed mixtures are higher than 1, implying that all mixtures meet the requirements for renewable energy, even when productivity of rapeseed is low – 1.79 t/ha. When the content of fatty acid methyl esters produced from waste in a mixture is increasing the $R₁$ values also increase (the fossil energy is used more effectively). When the indexes of energetic efficiency of the test mixtures of methyl esters were compared to the indexes of pure RME as well as fossil diesel fuel $(R₁ = 0.885)$, has been determined that it is expedient to use fatty waste in the production of biodiesel fuel.

Upon determining the life cycle energy efficiency indicators of tri-component mixtures, engine emissions were investigated by fuelling with the above-mentioned mixtures, using a direct injection engine at two rotation speeds: 1200 min⁻¹ and 2000 min–1. The engine operated normally, even reaching the required rotation moment. The emissions have been analysed regarding NO_x , CO, CO₂, PAH (polycyclic aromatic hydrocarbons) and smokeness. The results are presented in Tables 4 and 5.

Emissions of tri-component fuel were compared with RME emissions and found to differ only slightly. Only the content of PAH decreased by 57% when the engine operated at a 1200 min⁻¹ and by 30% at a 2000 min–1 rotation speed. Comparing the emissions of RME–LSME–PME and RME–LSME–TME mixtures, no significant differences were determined.

The test results show that mixtures of fatty acid methyl esters made from waste materials (oil contaminated by pesticides and animal fats) with RME can be effectively used in diesel engines.

Fig. 2. Energy consumption (%) for production of 1 tonne of fatty acid methyl esters of animal origin

Table 3. Life cycle efficiency indicator R₁ of tri-component biodiesel fuel mixtures

	Fuel composition, %							
RME	LSME	TME,PME	Energy efficiency indicator R_1 , when rapeseed productivity					
			2.8 t/ha	1.79 t/ha				
100		0	1.24	0.96				
90		8	1.42	1.17				
80		16	1.60	1.38				
70		24	1.84	1.64				

Table 4. **Emission of exhaust gases by tri-component fuel RME–LSME–TME**

Table 5. **Emission of exhaust gases by tri-component fuel RME–LSME–PME**

No	Fuel composition, %		NO_{y} ppm		CO, ppm		$CO2$ %		Smokeness, Bosch		PAH, μ g/m ³		
	RME	LSME	PME	Rotation speed, min ⁻¹									
				1200	2000	1200	2000	1200	2000	1200	2000	1200	2000
. .	90			290	377	527	377ء	2.4	7.8	0.06	3.75	48.8	657.5
z.	80	4	16	305	443	540	360	2.4	7.9	0.05	3.7	45	570
J.	70	6	24	315	420	640	1400	2.5	7.9		3.7	28.1	654

The concentration of toxic components in exhaust gases differ only slightly if compared with that of pure RME.

Another important indicator used to evaluate the influence of fuel on the environment is assessment of fuel biodegradability. It was determined that during a 21-day period 89.6% of RME, 92.5% of PME, 98.3% of LSME degraded, while in case of fossil diesel fuel the biodegradability was only 57.3% during the same period. Data on RME biological decomposition differ slightly from data published in the European scientific literature, while data on fossil diesel fuel correspond to them (Industrial…, 1995).

Figure 3 presents the data of the biodegradability of tri-component fuel (RME-LSME-PME) in comparison to the degradability of fossil diesel fuel. The biodegradability of the tri-component fuel (90% fo RME, 2% LSME, 8% PME; 80% RME, 4% LSME, 16% PME; 70% RME, 6% LME, 24% PME) is similar. It is a little lower when the mixture contains more RME. The biodegradability of all the tri-component fuels after 21 days was higher than 90%, and about 1.5 times higher than that of fossil diesel fuel.

CONCLUSIONS

1. Energy consumption for RME production and the value of the life cycle energy efficiency indicator of RME $(R₁)$ depends directly on rapeseed productivity. When rapeseed productivity is 1.79 t/ha, the energy consumption for RME production is higher than the calorific value of RME (the value of R_i is lower than 1), thus RME cannot be attributed to renewable fuels.

2. Fatty acid methyl esters produced from fatty waste are characterized by higher values of the life cycle energy efficiency indicator as compared with RME. The energy is used in the most efficient way when waste animal fatty (pork lard and tallow) acid methyl esters are produced ($R_1 = 3.17$). Linseed oil fatty acid methyl esters are characterized by lower values of the life cycle energy efficiency indicator $(R₁ - 2.6)$.

3. The value of the life cycle energy efficiency indicator of tri-component mixtures (RME–LSME–TME, RME–LSME– PME) increases directly proportionally to the amount of fatty acid methyl esters produced from waste fat in the mixtures. It exceeds unity even when the rapeseed productivity in Lithuania is moderate.

4. The concentration of toxic components in exhaust gases of fuel mixtures corresponding to standard requirements (RME90–LSME2–PME8, RME80–LSME4–PME16, RME70– LSME6–PME24, RME90-–LSME2–TME8, RME80–LSME4– TME16, RME70–LSME6–TME24) is not much different from that of RME. Only the concentration of polycyclic aromatic hy-

Fig. 3. Dynamics of biological decomposition of tri-component fuel

drocarbons decreases down to 57% at 1200 min⁻¹ in the case of RME–LSME–TME mixtures.

5. The requirements of fuel biodegradability were met by all the test fatty acid methyl esters and their mixtures. 90–98% of them degraded in 21 days, while in the case of fossil diesel fuel the biodegradability was only 57% during the same period. Biodegradability was somewhat lower in the case of mixtures containing more rapeseed oil fatty acid methyl esters.

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BIODYZELINO POVEIKIS APLINKOS TARŠAI

S a n t r a u k a

Įvertintas riebalų rūgščių metilesterių gyvavimo ciklo energijos veiksmingumo rodiklis, deginių emisijos ir biologinis suirimas gamtinėje aplinkoje. Nustatyta, kad rapsų aliejaus riebalų rūgščių metilesterių (RME) gyvavimo ciklo energijos veiksmingumo rodiklis $(\mathbf{R}_\text{\tiny{l}})$ tiesiogiai priklauso nuo rapsų derlingumo. Esant 1,79 t/ha rapsų derlingumui, energijos kiekis, sunaudotas RME gamybai, yra didesnis už iš RME išgaunamos energijos kiekį (energijos veiksmingumo rodiklis mažesnis kaip 1), todėl RME negalima priskirti atsinaujinantiems energijos ištekliams. Riebalų rūgščių metilesteriai, gauti iš riebalingųjų atliekų, pasižymi didesnėmis energijos veiksmingumo rodiklio vertėmis, palyginti su RME. Efektyviausiai energija panaudojama gaminant atliekinių gyvūninių riebalų (kiaulių (PME) ir galvijų (TME)) rūgščių metilesterius ($R_1 - 3.17$). Mažesniu energijos veiksmingumo rodikliu pasižymi sėmenų aliejaus riebalų rūgščių metilesteriai ($R_1 - 2.6$). Naudojant atliekinių riebalų ir aliejaus riebalų rūgščių metilesterius trikomponenčių degalų mišinių (RME-LSME-TME, RME-LSME-PME) gamybai proporcingai jų kiekiui mišinyje didėja degalų energijos veiksmingumo rodiklis, kuris net esant vidutiniam Lietuvoje rapsų derlingumui viršija 1. Variklyje tirtų atitinkančių biodyzelino standartą degalų mišinių (RME90-LSME2-PME8, RME80-LSME4-PME16, RME70-LSME6- PME24, RME90-LSME2-TME8, RME80-LSME4-TME16, RME70- LSME6-TME24) emisijos nedaug skiriasi nuo RME. Tik policiklinių aromatinių angliavandenilių koncentracijos deginiuose sumažėja iki 57%, esant 1200 min⁻¹, tiriant mišinį RME-LSME-TME. Biologinio suirimo reikalavimus atitiko visi tirti riebalų rūgščių metilesteriai ir jų mišiniai. Per parą jų suiro 90–98%, o mineralinio dyzelino tik 57,3%. Biologinis suirimas šiek tiek mažesnis mišinių, kuriuose yra daugiau rapsų aliejaus riebalų rūgščių metilesterių.

Raktažodžiai: biodyzelinas, gyvavimo ciklas, biologinis suirimas, deginių emisijos