Increasing the Oxidative Stability of Oil Extracted from *Jatropha curcas L.* Seeds by Adding Oil Extracted from Roasted *Vitis vinifera* Seeds

MIHAELA GABRIELA DUMITRU*
University of Craiova, Faculty of Scienece, Department of Chemistry, 107i Calea Bucuresti, 200585, România

This study was conducted to investigate the efficacy of supplementing the oil extracted from Jatropha curcas L. seeds with the oil extracted from roasted Vitis vinifera seeds regarding its oxidation stability during storage. Two sets of samples were formed: 1-Oil obtained from Jatropha curcas L. seeds; 2-a blend made from 95% oil extracted from Jatropha curcas L. seeds and 5% oil extracted from the roasted Vitis vinifera seeds. The samples were placed in dark bottles and stored in laboratory at an average temperature of 25°C for 12 months. Lipid oxidation kinetics was assessed according to the value of peroxide, acidity index, total polyphenol content and oxidation stability determined through the Handorn-Zürcher method (Rancimat). The samples supplemented with oil obtained from roasted Vitis vinifera seeds showed a much higher stability compared to oil samples obtained from non-supplemented Jatropha curcas L. seeds.

Keywords: oil Jatropha curcas L.; oil Vitis vinifera; oxidation stability

The search for fuel sources alternative to current foss fuels used has been reported by various authors. The o extracted from *Jatropha curcas L.* seeds was identified a an important source of alternative fuel.

The literature indicates that the fatty acid methyl este of *Jatropha curcas* is one of the 26 fatty acid methyl este of oil that are most suitable for biodiesel product [1, 2].

Revealed that the fatty acid composition of *Jatroph curcas* oil was analyzed by gas chromatography, majo long chain fatty acids present in the *Jatropha curcas* o which are palmitic acid (16.69%), stearic acid (7.67% oleic acid (40.39%) linoleic acid (33.09%) and Linoleic aci (0.28%). *Jatropha curcas* oil contains high percentage o unsaturated fatty acid, which is about 75.64% [3].

Oil from the *Jatropha* seeds has excellent propertie including low acidity, low viscosity compared to castor o and a better cloud point and pour point when compared t palm oil [4-7] but also the disadvantage of high percentag of unsaturated fatty acids that cause susceptibility t oxidation [8-13].

The lipids contained are susceptible to oxidation in th presence of catalytic systems such as light, heat, enzyme metals, metallic-proteins and microorganisms leading t complex processes of autoxidation, photo-oxidatio thermal or enzymatic oxidation, most of which involve fre radicals and/or other reactive species as the intermediat Autoxidation is the most common process among all an is defined as the spontaneous reaction of lipids wit atmospheric oxygen through a chain reaction of fre radicals.

Autoxidation is a chain reaction including three basi steps: initiation, propagation, and termination [14-15 figure 1.

Although the process cannot be stopped, there are som methods that may be taken to slow down this proces Addition of antioxidants in the oils may help to slo oxidation or degradation processes and increase thei oxidation stability [16].

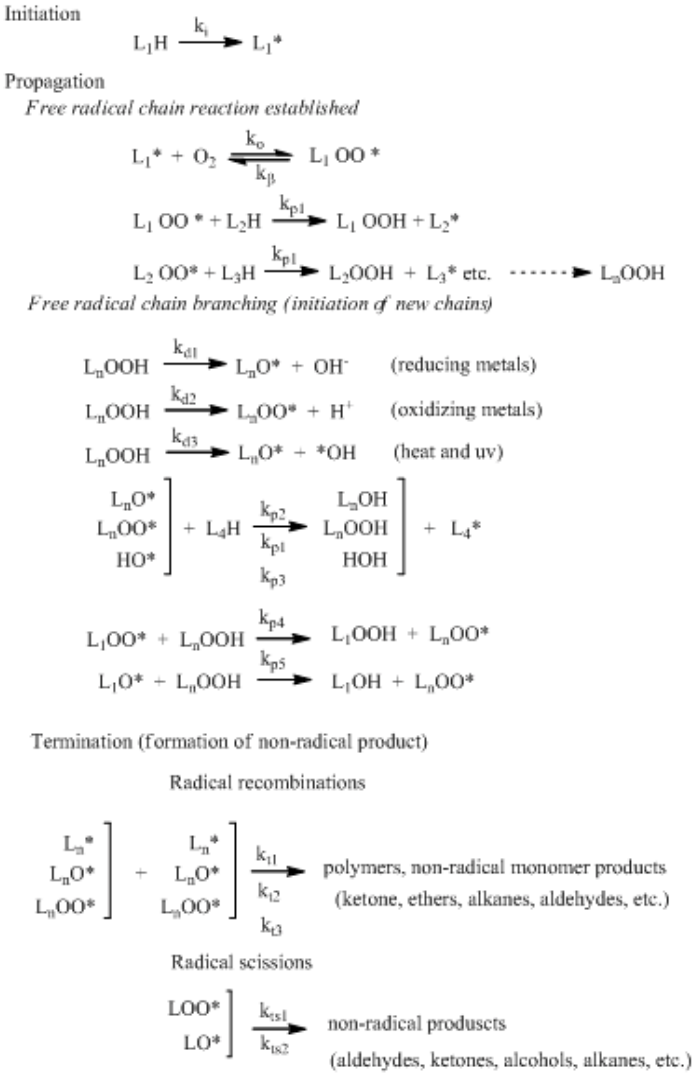

Fig. 1. Lipid oxidation mechanism

* email: dummgs@yahoo.com; Tel.: +40 251597048

This study was conducted to investigate the efficacy of supplementing the oil extracted from *Jatropha curcas* L. seeds with oil extracted from roasted grape seeds on the oxidation stability during storage. The progress of lipid oxidation was evaluated based on the peroxide index, acidity index, iodine index, content in total polyphenols and oxidation stability determined through the Hadorn-Zürcher method (Rancimat).

Experimental part

Materials and methods

To achieve the experimental part we used *Vitis vinifera* seeds purchased from the Teaching Research Resort of Craiova University-Romania, having the following characteristics [17]: *the appearance*: the dry surface without any trace of mold; *the color:* brown or dark red, various shades, characteristic for the variety of provenance; *smell*: is specific from the seed of *Vitis vinifera*, but it is not allowed the smell of mould; *seeds altered*: less than 5%; impurities (pieces of the bunch, skin)-below 3%; *the moisture*: 35-40%; *density*: 1.1-1.3 g/cm^3; *hygroscopicity, mL/100 g*: 7-15.

Roasting seeds was done in a toaster type Rombat for 10 minutes at 220°C and their grinding was done in a machine type Viacenza 200 adjusted to obtain a particle size of 1-1.25 mm.

Jatropha curcas L. seeds were purchased from Thai trade with the following characteristics: diameter 7.5-9 mm; length 17-19 mm; dark brown; 7.8% moisture and shredded by crushing.

Extraction of oil from seeds of Vitis vinifera and Jatropha curcas

The extraction of oil from the *Jatropha curcas* L. and *Vitis vinifera* seeds was carried out in a Soxhlet-type device using light petroleum solvent. The extraction time was 6 h. After oil extraction, the excess solvent was distilled off reduced vacuum using a rotary evaporator [18.19].

Characterization of oil obtained from Jatropha curcas L seeds and the blend of Oil Jatropha curcas L and Oil Vitis vinífera

Determination of peroxide value level by Hara-Totani method

Peroxide of oil samples was determined at the beginning and after determining oxidation stability by Hara-Totani [20-22]. In a 250 mL conical bottle was weighed on analytical balance a fixed quantity of oil (20 mg) and were dissolved in 10 mL chloroform. Add 15 mL glacial acetic acid and mix. After replacing air with nitrogen or CO_2, add 0.3 mL saturated KI solution and shake for 1 minute, followed by cooling in ice water bath in the dark. Add 100 mL cold distilled water and shake and then titrated potentiometrically with sodium thiosulfate solution 0.001 N keeping the vial in ice bath. During titration, there is a change in potential that in the equivalence moment shows a sudden drop. Parallel running a blank solution without oil where volume 0.001 N sodium thiosulphate used in titration until the equivalence point should not exceed 0.15 mL. If this amount is exceeded then another will prepare a saturated solution of KI. Index value is calculated in milliequivalents peroxide oxygen/kg oil using formula:

IP = (V - Vm) . F . N . 1000/m mE O_2/Kg oil

where: V - number ml 0.001 N sodium thiosulphate solution used for titration of sample to be analyzed;

Vm - number ml 0.001 N sodium thiosulphate solution used in blank titration;

m - mass of oil sample (g);

F - hactor solution 0.001 N sodium thiosulphate;

N - normality of 0.001 N sodium thiosulphate.

Acid Value

Two gram of the pure oil was weighed accurately by transfer method into a 250 mL conical flask. Neutral ethanol (20 mL) was added by means of a pipette and the flask heated on a steam bath for 3 min. Then the flask was cooled and the contents titrated with 0.1 N alcoholic potassium hydroxide solution using phenolphthalein as an indicator. A blank titration was also conducted side by side [2].

Determination of total phenolic content (TPC)

The TPC values of the seed oil extracts were measured using the Folin-Ciocalteu reagent [23,24]. The reaction mixtu;re contained 100µL of antioxidant extract or solvent, 500 ìL of the Folin-Ciocalteu reagent, 1.5 mL of 20% sodium carbonate, and 1.5 mL of pure water. After 2 h of reaction at ambient temperature, absorbance was read at 725 nm and used to calculate the TPC, using Gallic acid as the standard [25]. Triplicate measurements were taken and the results were expressed as mg Gallic acid equivalents (GAE)/g of seed oil using a standard curve generated with 50µg, 100, 300 and 500µg Gallic acid per 100 mL.

Oxidation stability through the Hadorn-Zürcher method (Rancimat)

This method consists in the oil oxidation in accelerate conditions. The method permits the establishment of the induction period, which corresponds, with the initiation step of the oil auto-oxidation. To determine the stability in oxidation it was used an installation, which used oxidized oil samples (10 g) at a temperature of 110°C [21]. Through the oil samples, it was bubbled air with a debit of 8 L per hour. Because of the oxidation reactions, which take place in a reactor, the formed volatile acids are trained by the air current and absorbed in the measurement cell where there is bidistilled water. The measurement of the solution conductibility is done with a conductometer of Radelkis type. In the beginning, we notice a slow increasing of the solution conductibility, after that it appears a sudden increasing of this because of the formation of volatile acids. The induction period is considered the interval until the moment of the suddenly curve's change, (fig. 2).

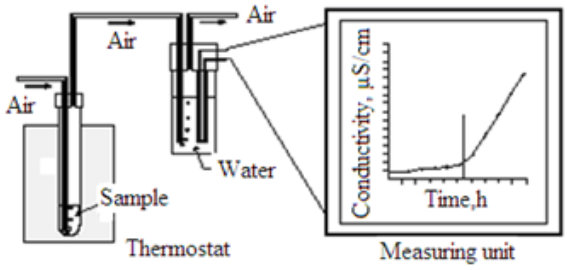

Fig. 2. Installation for the determination of stability in the Biodiesel oxidation

Two sets of samples were formed:

- oil obtained from *Jatropha curcas L* sedes;
- a blend made from 95% oil from *Jatropha curcas L* seeds and 5% oil from roasted *Vitis vinifera* sedes;

The samples were placed in dark bottles and stored in laboratory at the average temperature of 25°C for 12 months. Every month, major qualitative oil indexes were determined from oil samples: the index of peroxide, index of acidity, content in total polyphenols and oxidation stability.

Results with discussions

The literature shows the oil extracted from the Vitis vinifera seeds compared to other oils as a source of

Component name	Concentration mg/Kg oil	Source	Component name	Concentration mg/Kg oil	Source
α-tocopherol	36-325	26,27,30-34	α-tocotrienol	56-319	27,30-34
β- tocopherol	25-48	27,31,32	β-tocotrienol	4-18	26
γ- tocopherol	3-39	26,27,30-34	γ-tocotrienol	261-1575	26,27,31,32
δ-tocopherol	0.6-163	30	δ- tocotrienol	6-17	26, 27,31,32
Total phenols	100-238	30	Phytosterols	82-136	26

Table 1
THE MAIN CHEMICAL COMPONENTS CONTAINED IN THE OIL EXTRACTED FROM *VITIS VINIFERA* SEDES

Table 2
THE MAIN QUALITATIVE INDEXES OF OIL SAMPLES DETERMINED DURING STORAGE

Period	Oil *Jatropha curcas* L				Oil *Jatropha curcas* L + Oil *Vitis vinifera*			
	PV	AV	TP	OS	PV	AV	TP	OS
Ianuarie	3.5	2.31	39.0	181	2.1	2.32	169.0	724
Februarie	5.1	2.80	38.5	179	3.7	2.75	167.0	721
Martie	7.4	3.00	35.6	159	6.5	2.81	160.0	718
Aprilie	12.9	4.10	28.7	141	10.2	3.78	157.0	710
Mai	26.0	9.58	19.0	121	16.3	5.35	145.0	690
Iunie	30.0	15.75	16.0	115	20.0	7.32	130.7	679
Iulie	42.8	20.66	11.5	109	24.5	10.60	110.0	624
August	53.0	24.10	8.3	100	38.0	15.25	101.0	490
Septembrie	65.0	30.81	6.5	95	49.3	20.24	98.2	450
Octombrie	77.1	35.30	4.0	88	58.1	25.50	88.4	404
Noiembrie	85.8	38.20	3.5	75	62.0	27.11	75.0	368
Decembrie	91.2	59.10	2.0	59	70.4	30.45	69.4	213

antioxidants directly related to the high concentration in tocopherol and tocotrienol isomers of vitamin E, vitamin E that is rarely found in other oils [26-34].

In table 1, are shown the main chemical components contained in the oil extracted from *Vitis vinifera* seeds.

In addition, grape seed oil contains tannins in higher concentrations than other seed oils [35] such as Gallic acid, catechin, epicatechin, and a large amount of procyanidins [36]. By roasting *Vitis vinifera* seeds at 220°C by hydrolysable tannins form, through decarboxylation, pyrogallol, a strong antioxidant [37]. In this context we used the oil extracted from roasted *Vitis vinifera* seeds in supplementing the oil extracted from *Jatropha curcas* L seeds.

To determine the role played by supplementing the *Jatropha curcas* L. oil with the oil obtained from roasted *Vitis vinifera* seeds, we used as indicators: the index of peroxide, index of acidity, content in total polyphenols and oxidation stability in the samples studied.

Following the determinations carried out on oil samples, we obtained these results (table 2).

The experimental values indicate lipid oxidation in two periods of time. In the first 4 months of storage of non-supplemented oil extracted from *Jatropha curcas* L. we observed a slower growth in peroxide values (12.9 mEq O_2/Kg oil). In the next period, up to 12 months, the rate of oxidation increases and peroxide values increase toward the value of 91.2 mEq O_2/Kg oil.

The main factor affecting lipid peroxidation reaction rate is deemed to be the reaction initiation type (monomolecular or bimolecular). The monomolecular or bimolecular reaction can be responsible for initiating the lipid oxidative chain of oil by decomposing peroxides. During the first 4 months of storing non-supplemented *Jatropha curcas* L. oil, the low concentration of peroxides favors monomolecular initiation:

$$PV^{1/2} = PV_0^{1/2} + \frac{1}{2}k_a^t \quad 0 \le t < t_b$$

and when it reaches the critical value of 26.0 mE O_2/Kg oil (the sudden increase of the peroxide index value), the reaction is controlled by the bimolecular mechanism [38, 39]:

$$PV^{1/2} = PV_b \times e^{k_b \times (t - t_b)} \quad t_b \le t$$

PV–the value of peroxide, mE O_2/Kg oil;

PV_0–the value of peroxide at the beginning of experiment (time t_0);

PV_b–the value of peroxide at time t, months;

k_a, k_b–constants, (mE O_2/Kg);

t_b–the time needed to reach the critical point, months.

In *Jatropha curcas* L. oil supplemented with the oil extracted from roasted *Vitis vinifera* seeds, the critical value is reached after 7 months of final storage reaching a peroxide index value of 70.4 mE O_2/Kg oil, value 22.8% less than non-supplemented oil.

Acidity is one of the essential properties for determining the oil quality. Changing the acidity of oils occurs in the formation of peroxides which decompose and interact with the formation of several oxidation products, including aldehydes, which are oxidized in acids [40,41].

It was noted that the samples of oil extracted from *Jatropha curcas* L. seeds supplemented with oil extracted from *Vitis vinifera* seeds show, in the beginning of the experiment, an acidity of 2.32 mg KOH/g oil and after 7 months of storage, when it reaches the critical period of 10.60 mg KOH/g oil to finally reach the value of 30.45 mg KOH/g oil. The samples of non-supplemented oil have, in the beginning of the experiment, the value 2.31 mg KOH/g oil and after 4 months of storage to achieve the critical value of 9.58 mg KOH/g oil and finally reaching the value of 59.10 mg KOH/g oil.

To minimize oxidation reactions that occur during storage, we use antioxidants.

In a system, an antioxidant can be defined as any substance that is present in low concentrations compared to that of an oxidized substrate, which delays or prevents the oxidation of that substrate [39]. Antioxidants are natural or synthetic substances that react with atmospheric oxygen or free radicals in the environment, protecting compounds against autoxidation. The study carried out is based on the antioxidant properties of polyphenols contained in the oil extracted from the roasted *Vitis vinifera* seeds.

The antioxidant activity of phenolic compounds is regarded to be related to scavenging free radicals, chelating transition-metals involved in free-radical production and inhibiting the enzymes participating in free-radical generation [43-46].

The free radical scavenging activity of phenolic compounds is generally attributed to their ability to donate a hydrogen atom to reduce ROS radicals [42].

$$ROO\bullet + ArOH \rightarrow ROOH + ArO\bullet$$
$$HO\bullet + ArOH \rightarrow HOH + ArO\bullet$$
$$ROO\bullet + ArO\bullet \rightarrow ROOArO$$

During the experiment, the value of total polyphenol content of non-supplemented *Jatropha curcas* L. oil ranges from 39 mg GAE/g oil to 2 mg GAE/g oil at the end of the storage period.

The polyphenol content in the oil samples extracted from the supplemented *Jatropha curcas* L. seeds has a range from 169 mg GAE/g oil at the beginning of the experiment and 69.4 mg GAE/g oil at the end of the experiment.

The fact that the *Vitis vinifera* seeds, through their content in polyphenols (Gallic acid, monomers flavan-3-ol (catechin, epicatechin, galocatechin, epicatechin 3-O-gallate, dimers, trimers and polymers procyanidins), develop an antioxidant activity 20 times higher than vitamin C and 50 times stronger than vitamin E [47,48] have helped to protect the oil obtained from *Jatropha curcas* L. seeds during storage. This antioxidant effect is highlighted by the values obtained by the samples supplemented with the oil extracted from roasted *Vitis vinifera* seeds. We consider that an important role is played by the pyrogallol formation from hydrolysable tannins which increase the antioxidant effect of oil.

The parameters determined by the Handorn-Zürcher method is a variation visible throughout the test. It shows a correlation between the oxidation stability and the degradation of non-supplemented oil samples quality compared to supplemented samples. Therefore, non-supplemented samples show a value of oxidation stability between 180 min in the beginning of the experiment and 59 min at the end, while supplemented oil samples had 724 min in the beginning of the experiment and 213 min at the end. The data in table 2 express the changes of parameters registered during the experiment in response to supplementing the oil extracted from *Jatropha curcas* L. seeds with the oil extracted from roasted *Vitis vinifera* seeds.

Conclusions

The antioxidant effect of the oil from roasted *Vitis vinifera* seeds was identified through the results obtained as a viable means of improving the stability of oil obtained from *Jatropha curcas* L seeds.

The small values of peroxides, low acidity, high content in total polyphenols and greater stability of supplemented oil samples during storage clearly show the antioxidant potential of the oil extracted from roasted *Vitis vinifera* seeds.

Abundant raw materials, high content in substances with antioxidant potential and very low price of *Vitis vinifera* seeds recommend their use in the protection activity of oils from the oxidative degradation effect.

References

1.MOHIBBE, A.M., WARIS, A., NAHAR, N.M., Biomass Bioenerg.,**29**, 2005, p.293.
2.JOSHI, A., PANKAJ, S., BACHHETI, R.K., Int. J. Appl.. Biol Pharm., **2**, 2011, p.2123.
3.UGBOGU, A.E., AKUBUGWO, E.I.,UHEGBU, F.O., CHINYERE, C.G., UGBOGU, O.C., ODUSE, K. A., Int. Food Res., **21**, nr. 2, 2014, 735.
4.NEIDA, C., OM TAPANES DONATO, A., GOMES, A., JOSE, W., DE MESQUITA, C., and OCTAVIO, A., CEVA, ANTUNES., Fuel, **87**, 2008 p. 2286.
5.SARIN, R., SHARMA, M., SINHARAY, S., MALHOTRA, R.K., Fuel, **86**, 2007, p. 1365.
6.McCORMICK, R.L., RATCLIF, M., MOENS, L., LAWRENCE, R., Fuel Process. Technol., **88**, 2007, p. 651.
7.DUANPEN, C., JARUN, C. and PENJIT, S., Kasetsart J. (Nat. Sci.), **44**, 2010, p. 243.
8.NZIKOU, J.M., MATOS, L., MBEMBA, F., NDANGUI, C.B., PAMBOU-TOBI, N.P.G., KIMBONGUILA, A., SILOU, T.H., LINDER, M., DESOBRY, S., Res. J. App. Sci. Eng. Technol., **1,** nr. 3, 2009, p. 154.
9.AKINTAYO, E.T., Bioresour. Technol., **92,** 2004, p. 307.
10.AUGUSTUS, G.D.P.S., JAVA BEANS, M., SEILER, G.J., Biomass Bioenerg., **23,** 2002, p. 164.
11.KPOVIESSI, D.S.S., ACCROMBESSI, G.C., KOSSOUOH, C., SOUMANOU, M.M., MOUDACHIROU, M., C.R. Chim., **7,** 2004, p. 1007.
12.BASHAR, M.A., RAHIMI, M.Y., JUMAT, S., EMAD, Y., NADIA, S., Chem. Mol. Eng., **7**, nr. 12, 2013, p. 893.
13.MADARASZ, J., AMIT, K., J. Eng. Sci., **1,** nr. 3**,** 2011, p. 186.
14.DUMITRU (BITA), M.G., GRECU, D.R., TUTUNEA, D., POPESCU, A., BICA, M., Rev. Chim.(Bucharest), **61,** no. 9, 2009, p. 882.
15.DUMITRU, M.G., NAGI, M., BUCULEI, M.E., PICIOREA, G.D., HITICAS, I.D., STANCIU, T., Bulg. J. Agric. Sci., **19,** nr. 6, 2013, p. 1445.
16.FOXA, N.J., STACHOWIAK, G.W., Tribol. Int., **40,** 2007, p. 1035.
17.BITA, M.G., PREDA, M., Riv. Ital. Sostanze Gr., **LXXXI,** nr. 5, 2005, p.326.
18.*** SR 13258/96
19.BITA, M.G., PREDA, M., Rev. Chim.(Bucharest), **56,** no. 7, 2005, p.716.
20.HARA, S., TOTANI, Y., J. Am. Oil Chem. Soc., **65,** nr. 12**,** 1988, p. 1948.
21. BITA, M.G., PREDA, M., Riv. Ital. Sostanze Gr., **80,** nr. 6**,** 2004, p. 377.
22.DUMITRU, M.G., GRECU, D.R., Bulg J. Agric. Sci., **16,** nr. 2, 2010, p. 227.
23.JOSHI, A., PANKAJ, S., BACHHETI, R.K., J. Appl. Biol. Pharm**., 2,** nr. 1, 2011, p. 123.
24.YU, L., PERRET, J., HARRIS, M., WILSON, J., HALEY, S., J. Agri. Food Chem., **51,** nr. 6, 2003, p. 1566.
25.ARSLAN, D., Grasas aceites, **63,** nr. 2**,** 2012, p. 158.
26.FERNANDES, L., CASAL, S., CRUZ, R., PEREIRA, J.A., RAMALHOSA, E., Food Res. Int., **50,** nr. 1**,** 2013, p. 161.
27.FERNANDA, B.S., FERNANDA, C.D.S., LUCILLIA, R.O., MANCINI-FILHO, J., Food Sci. Technol (Campinas), **35,** nr. 3, 2015, p. 399.
28.FREITAS, L.S., JACQUES, R.A., RICHTER, M.F., SILVA, A.L., CARAMÃO, E.B., J Chromatogr A, **1200,** nr. 1**,** p. 80**,** 2008.
29.CREWS, C., HOUGH, P., GODWARD, J., BRERETON, P., LEES, M., GUIET, S., WINKELMANN, W., J. Agr. Food Chem., **54,** nr. 17**,** 2006, p. 6261.
30.GOKTURK, B.N., GULCAN, O., EMINE, S.C., Grasas aceites, **58,** nr. 1, 2007, p. 29.
31.NAVAS, P.B., Arch. Latinoam Nutr., **59,** nr. 2, 2009, p. 214.
32.NAVAS, P.B., Rev. Fac. Agron. (LUZ), **27**, 2010, p. 270.
33.OVCHAROVA, T., ZLATANOV, M., IVANOV, A., EIJT, **3**, nr. 9, 2014, p. 178.
34.ILKNUR, D., EBRU, P., YBRAHIM, S.O., ALASALVAR, C., ERDAL, E., Eur. J. Lipid Sci. Tech., **115,** nr. 6, 2013, p. 641.
35.LUQUE-RODRÍGUEZ, J.M., DE CASTRO, M.D.L., PEREZ-JUAN, P., Talanta, **68**, nr. 1, 2005, p. 126.
36.BEVERIDGE, T.H.J., GIRARD, B., KOPP, T., DROVER, J.C.J., J. Agr. Food Chem., **53,** nr. 5, 2005, p. 1799.
37.MINERVA, M.F., RONALD, A.H., MICHAEL, E.W., DIANE, G., MILTON, H., MICHELLE, H., Cutan. Ocul. Toxicol., **32,** nr. 3, 2013, p. 234.
38.BITA, M.G., Rev. Chim., **59,** nr. 9, 2008, p. 1014.
39.ORTOLA, M.D., GUTIERREZET, C.L., CHIRALT, A., FITO, P., Technol. Int., **4,** 1998**,** p. 67.
40.KNOTHE, G., DUNN, R.O., J. Am. Oil Chem. Soc., **80**, 2003, p. 1021.
41.FRANKEL, E.N., Lipid oxidation, vol. **19**, Pergamon Press Ltd.Ed Printed in Great Britain, 1980, p. 1-22.
42.HALLIWELL, B., MURCIA, M.A., CHIRICO, S., ARUOMA, O.I., Crit. Rev. Food Sci. Nutr**., 35,** 1995, p. 7.
43.YANG, B., KOTANI, A., ARAI, K., KUSU, F., Anal. Sci., **17**, nr. 5, 2001, p. 599.
44.ARUOMA, O.I., Mutat. Res., **523–524,** nr. 9, 2002, p. 9.
45.HENSLEY, K., MOU, S., PYE, Q.N., DIXON, R.A., SUMMNER, L.W., FLOYD, R.A., Curr. Top. Nutraceutical Res., **2,** 2004, p. 13.
46.GREEN, R.C., 2007. Physicochemical properties and phenolic composition of selected Saskatchewan fruits: buffalo berry, chokecherry, and sea buckthorn. [Thesis]. The Department of Applied Microbiology and Food Science University of Saskatchewan, Saskatoon, Canada
47.SHI, J., YU, J., POHORLY, J., KAKUDA, Y., J. Med. Food, **6,** nr. 4,2004, p. 291.
48.BITA, M.G. (DUMITRU), PREDA, M., Rev. Chim.(Bucharest), **58,** no. **6**, 2007, p. 494.

Manuscript received: 6.06.2015