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Lake blue clay - sapropel - flax shive briquettes for water absorption and desorption

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Abstract. Latvian lakes are rich in sapropel sediments and below the sapropel layer there is another valuable natural resource, namely blue clay. Flax shives are formed in large quantities as a waste in flax processing factories. The problem involves processing and rational use of these resources. The paper studies the obtaining of briquettes from flax shives using sapropel and lake clay as a binding material. Briquettes are intended for use in cases when problems with regular plant watering occur. For example, provision of plants with water in greenhouses, indoor spaces for a long period of time without human presence. Briquettes are placed into the soil next to the plant roots. Briquettes are initially moistened and absorb a significant amount of water (moisture content to be expressed in comparison to the sample's dry weight 250–380%). The presence of clay in them contributes to slow drying of briquettes and provides a long lasting supply of moisture to the plant roots. The paper deals with the issues of the optimal composition of components and the amount of pressure for the production of briquettes, as well as water absorption and desorption properties of briquettes. The aim of the study is to develop a production method of clay – sapropel – flax shive briquettes with good water absorption and desorption properties.

Key words: clay, sapropel, flax shives, briquettes.

INTRODUCTION

Latvia has 2256 lakes with the total area of 1,001 km^2. Most lakes and bogs contain sapropel deposits. Overall resources of sapropel in Latvia comprise approximately 2 billion m^3 (Stankevica & Kļavins, 2013). The sapropel is a renewable and natural resource. Numerous studies have shown the possibility of the effective use of sapropel in building, bio-energy, food, chemical industry, agriculture, veterinary medicine, cattle-breeding, forestry, medicine and cosmetics (Kireicheva & Khokhlova, 1998; Shinkarev et al., 2000; Schepetkin et al., 2002; Nikolaeva et al., 2009; Obuka et al., 2013; Ostrovskij, 2014; Platonov et al., 2014; Tsukanov et al., 2014). A sufficient number of studies has been done to determine the sapropel presence in Latvia, its use in solid soap, glue and composite material manufacturing, as well as its humus impact on hydroponics (Research of modification possibilities and properties of peat, sapropel and clays, 2015).

Clay is one of the most common and most accessible minerals in Latvia. Latvia is rich in clay and it has the largest amount of clay resources in Europe. Most of Latvia's clay resources are used in manufacturing building ceramics, however, the potential of local clay is not sufficiently used (Stinkule, 2014). In the world clays have a tremendous number of miscellaneous uses. The use of clays as soaps, absorbents and construction material has been reported since ancient times. Clays are used in the manufacture of porcelain, whiteware, refractories, bricks, tiles, stoneware, glazed products, catalysts, in preparation of muds for drilling oil wells, as effective barriers to isolate radioactive wastes, and may absorb various pollutants including organic compounds and inorganic trace metals (Kodama & Grim, 2018). The effectiveness of clay in the pharmaceutical industry, in medicine and cosmetology has been proved (Lopez-Galindo et al., 2007; Ray & Ferrell, 2008; Carretero & Pozo, 2009; Ghiaci et al., 2009; Williams & Haydel, 2010; Abdel-Motelib et al., 2011; Matike et al., 2011). The structure, composition and formation of clay deposites is very variable. The lake blue clay is located below the sapropel layer and is considered to be the recent sediment. The natural composition of clay has admixtures of various dispersions - quartz, calcite, dolomite, feldspar, organic compounds etc. (Stinkule, 2014), therefore the use of clay minerals in composites requires pre-treatment of clay, namely release from coarse-grained admixtures. The lake blue clay is free of coarse-grained admixtures.

Since ancient times the flax has always taken a significant place in the agriculture of Latvia (Ivanovs & Stramkale, 2001). Flax fibre (shives) is considered to be waste product remaining after the cellulose fibre removal from flax straw. The core flax shive is generally composed of ~46% cellulose, ~26% hemicellulose, ~23% lignin and ~2% ash (Stemergy renewable bio-fibre, 2018). For every tonne of flax fibre there can be produced 2.5 tonnes of shive (Cox et al., 1999). Flax shives are an industrial and agricultural by-product and it is important to find a solution for its rational use.

The aim of the research is to develop environmentally friendly, biodegradable briquettes with good water absorption and desorption properties using local resources such as lake clay, sapropel and industrial by-product, namely flax shive, and offer a method for their production. The briquettes, which accumulate water in large quantities and slowly return it providing soil with additional water reserves for a long period of time, are intended for use in agriculture. They are placed into the soil where it is necessary to provide water for extended period of time or in areas where the access to water is limited, for example, provision of plants with water in greenhouses or indoor spaces for a long period of time without human presence. Briquettes will gradually provide the soil with water and plants will absorb from it. In the study the mixture of sapropel and blue lake clay is used in briquettes as binders. There are indications that the clay mineral surface (surface charge) and the organic component form a chemical bond (Švinka et al., 2016). Sapropel has an adhesive and hydrophobic quality (Stankeviča & Kļaviņš, 2013; Obuka et al., 2015). Clay in briquettes also serves as water absorbent. Clay materials contain water in several forms. The water may be held in pores and may be removed by drying under ambient conditions. Water may also be adsorbed on the surface of clay mineral structures (Kodama & Grim, 2018). Flax shive performs the function of filler and water absorbent in briquettes. The core flask shive has a porous structure making it very absorbent (Stemergy renewable bio-fibre, 2018).

MATERIALS AND METHODS

In the study briquettes with different composition of mass of sapropel, flax shive and clay are used (Table 1). Separate tests are carried out on a binder of briquettes, specifically dried solid sapropel.

Table 1. Composition and mechanical properties of briquettes used in the research

Type of briquettes	1	2	3	4	5	6	7	8	9
Composition in mass %; sapropel : flax shive : clay	80: 10:10	80: 15:5	80: 5:15	75: 10:15	75: 15:10	75: 20:5	70: 15:15	85: 7.5:7.5	85: 10:5
Hardness, Shore A	30 ± 16	52 ± 12	58 ± 17	40 ± 10	40 ± 24	51 ± 11	40 ± 14	41 ± 13	52 ± 29
Compressive strength 10% relative deformation; MPa; measurement error 1.6%	0.051	0.104	0.038	0.040	0.124	0.103	0.061	0.061	0.063
Density, kg m^{-3}; measurement error 2.0%	232	196	202	195	207	205	199	209	184

Blue clay and sapropel were collected from the lake Plusons (Latvia). Sapropel colour is black–grey; odour – weak, wet soil; consistency – plastic, soft, easily applied and adhesive; moisture 90.53%, dry matter – 9.47%; pH 7.51; contains Na, Mg, K, Ca, Fe, Cr, Mn, Co, Ni, Cu, Zn, Cd, Pb, Al, Ag, Ba. Blue clay has a colloid composition, sludge odour, grey colour, plastic, soft, smooth consistency, moisture 80.16%, dry matter – 19.84%, pH 7.74., which contains Na, Mg, K, Ca, Fe, Cr, Mn, Co, Ni, Cu, Zn, Cd, Pb, Al, Ag, Ba (Tretjakova et al., 2017).

Drying of sapropel samples (until the mass is left unchanged) is carried out in a laboratory room at the temperature of 20 ± 1 °C with a relative air humidity of 31 ± 3% and in a drying oven Memmert UNE 400 at a high temperature (80, 100, 140, 1,600 °C). Shore D Hardness Tester is used to measure the hardness of dried sapropel.

Sapropel, flax shives and clay are mixed to uniform consistency, filled in cubic shapes having dimensions of 50 x 50 x 50 mm and dried in a laboratory room in still air (temperature 20 ± 1 °C, relative humidity of air 31 ± 3%)(Fig. 1).

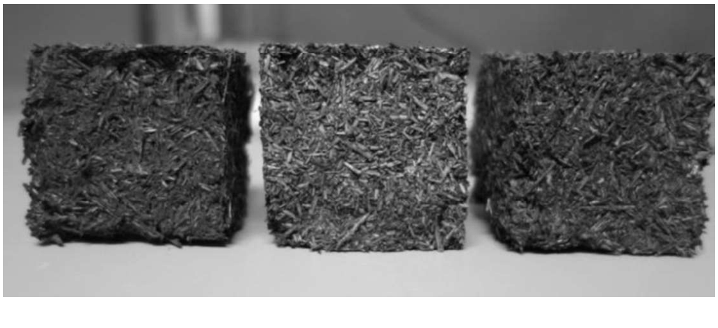

Figure 1. Sapropel, flax shives and clay briquettes.

The obtained briquettes are tested for water content:

$$u = \frac{m_w}{m_{dry}} \cdot 100\%, \tag{1}$$

where m_{dry} – dry mass of the material (oven-dried material), m_w – the mass of water in the material; $m_w = m - m_{dry}$; m – the mass of wet material.

Dry mass of dried briquettes is determined m_{dry}. Then the briquettes are completely immersed in water at a temperature of 20 °C for 1 hour. In the beginning when the briquettes are removed from water the water content (time t = 0) is determined. Then the drying process of briquettes in the soil is studied. A flat bath (length-500 mm, width-400 mm, depth-80 mm), which is fully filled with soil is used. The initial moisture of soil is 21%. Water-impregnated briquettes are placed in the soil at least 50 mm from each other. The samples are kept in a room with a temperature of 20 ± 1 °C, relative humidity of air is 31 ±%. From time to time briquettes are removed out of soil, carefully cleaned of soil particles, weighed (wet mass m is determined) and placed back into the soil. Their water content is calculated. The process of measurement continues until the briquettes have completely released the water to the soil (until the mass of the briquettes remains unchanged).

Using universal tensile machine Zwick / Roell Z-150 (Fig. 2), briquettes are subjected to pressure testing; strain rate-10mm min^{-1}; maximum compression load of 2kN; temperature 20 °C. The compressive strength is determined from the compression coefficients at a relative deformation of 10%. Shore A Hardness Tester is used to measure hardness of briquettes.

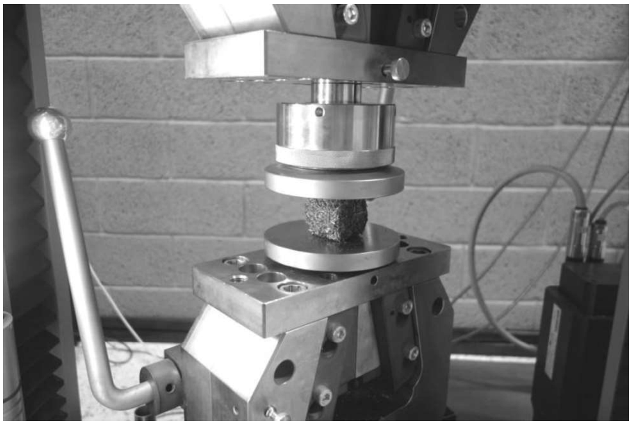

Figure 2. Briquette pressure testing using Zwick / Roell Z-150 universal testing machine.

RESULTS AND DISCUSSION

It is important to accelerate the technological process of manufacturing briquettes. The greatest amount of time it consumes is due to drying of the sapropel, clay and flax shive briquettes. The drying process can be accelerated by raising the temperature in the dryer and providing an intense flow of dry air and discharge of the wet one. It is important (especially during transportation) that the mechanical resistance of the briquettes is as high as possible. Sapropel serves as a binder in briquettes, so the mechanical properties of sapropel greatly affect the mechanical resistance of briquettes. In order to determine the optimum temperature during drying, the hardness of the dried sapropel is used initially as an indicator. It has been identified that with increasing drying temperature the hardness decreases (Fig. 3). For example, the sapropel just dried at 20 °C according to Shore D is 66 ± 3.7, but the hardness of freshly dried sapropel at 160 °C is only 49 ± 6. Hardness increases over time after complete drying of the sapropel, for example, within 1 week (samples are located in a room with a temperature of 20 ± 1 °C and a relative humidity of 31 ± 3%) after drying out the sapropel, the Shore D hardness increases from 66 ± 3.7 to 74 ± 3 (if the drying temperature was 20 °C), 49 ± 6 to 63 ± 4.5 (if the drying temperature was 160 °C). The explanation may be related to the hydration processes during sapropel drying (similar to the way it occurs during hardening of the concrete), which result in the formation of hydrosilicates, hydroaluminates, and calcium hydroferrates. The probability may be proven by the presence of silicon, calcium, iron, aluminium and oxygen in the content of sapropel (Martinovs et al., 2017; Pleiksnis et al., 2017). This means that drying must be done slowly (for a long time without forced ventilation) at a temperature of 20 °C to obtain more resistant material. In this case, sapropel is dried to have the highest degree of hardness. The briquettes must be kept at least 1 week in a warehouse after drying. During this time the hardness of sapropel increases by 12% (if the drying is at 20 °C, Fig. 3). Only then briquettes can be transported. This means that accelerating the production technological process at the expense of drying time is problematic, therefore additional research is still needed. Consequently, it must be taken into consideration that briquette manufacturing requires large areas for drying and storing products.

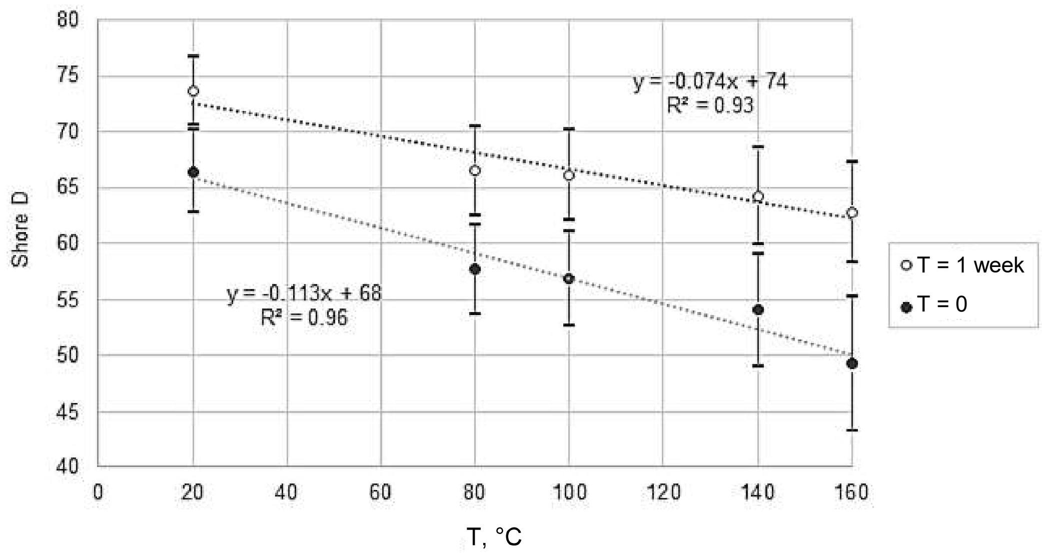

Figure 3. Sapropel hardness (Shore D) depending on the drying temperature (T) and the time(t) after sapropel has been completely dried (t = 0-freshly dried sapropel; t = 1 week-solid sapropel which has been kept in a warehouse for one week).

The press test charts for all types of briquettes are given in Fig. 4. It shows that the briquette type No.5 deforms the least in the result of pressure (maximum load for all samples 2kN). Composition, hardness, compressive strength at 10% relative deformation and density for various types of briquettes are given in Table 1. These results (compressive strength, density) are close to sapropel and hemp shives thermal insulation material (Pleiksnis et al.,2017). It has been proven that hemp shives are especially strong and flexible (Adamovics et al., 2013). This means that it would be possible to use hemp shives or other fillers, such as sawdust, wood chips or their blends instead of flax shives. In Table 2 there are given the results of the drying process of water-impregnated briquettes. It shows the changes in water content % in different types of briquettes over some period of time. Figures for the characterization of the composition of the briquettes mean the proportion of the mass of sapropel-shives- blue clay, for example, 80:10:10 means that 80% of briquettes contain sapropel, 10% of flax shives and 10% of blue clay. The data in Table 2 shows that the best water desorption properties are in briquettes with composition 75:15:10 (briquette type No. 5).

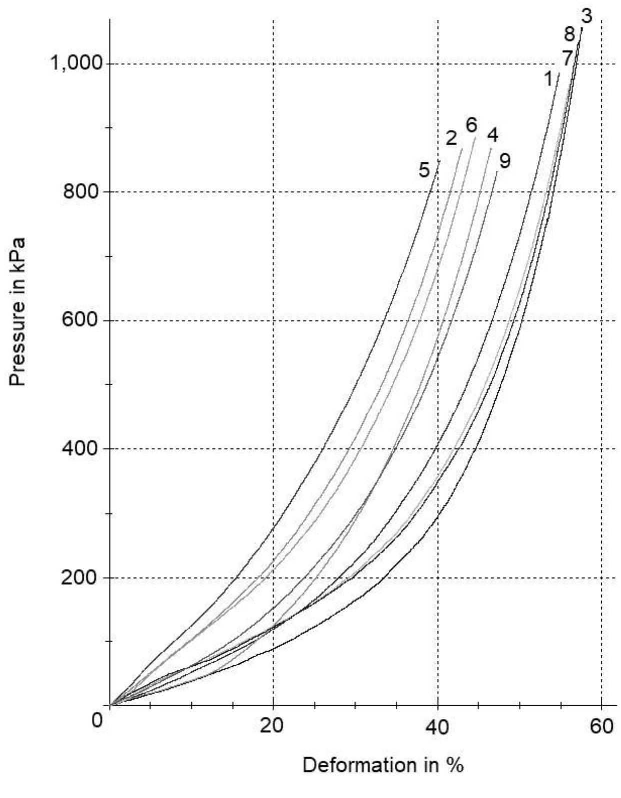

Figure 4. Briquette press characteristic curves in the range up to 2kN of compressive force.

Table 2. Changes in absolute water content (u) (%) of briquettes during a period of time t (when they are drying in soil)

t, day	Type of briquettes								
	1	2	3	4	5	6	7	8	9
0	320	317	289	297	243	290	316	329	320
1	191	205	178	194	185	211	207	197	202
2	161	167	148	157	161	180	171	168	174
3	141	148	131	136	142	154	148	146	146
4	125	131	102	116	124	133	124	125	126
7	74	71	52	77	90	71	73	80	77
8	61	59	32	61	78	59	58	60	61
9	49	50	20	47	66	50	45	46	50
10	38	40	9	32	55	40	33	30	37
11	26	29		22	44	29	20	17	24
14	12	12			22	17	12	11	11
15					19				

Briquettes have low mechanical resistance and they can easily be damaged during transportation. To solve this problem, a briquetting machine (Fig. 5) is offered. The certain proportions of sapropel, flax shive and clay are mixed to uniform consistency and put in a briquetting machine. Briquettes have a cubic shape with dimensions of 50 x 50 x 50 mm. For forming, drying, storing and transportation of briquettes, thermoplastic forms that have been manufactured using a plastic vacuum forming method are used. The prepared form is placed in the molding machine. Constructively the briquetting equipment consists of a rack (made of steel tubes), a table surface (on which there are placed linear movement knobs, namely linear rails, electric drive), the feed hopper and a rammer roller to tamp the mixture in a mold. The mixture gets into the shapes after the shutter with pneumatic drive built in the feed hopper is opened. The amount of mixture in forms can be adjusted by changing the speed of the hopper and the roller. This can be done by adjusting the speed of the electric drive motor with a variable frequency drive. When the hopper reaches the end position, the end switch is actuated, the shutter is closed, the electric drive is reversed and the hopper gets to the starting position. A new briquetting cycle can be started after inserting a new shape and pressing the start button done by the machine operator.

Traditionally, sapropel is used as a soil improver or supplementary fertilizer (Stankeviča & Kļaviņš, 2013). Thus, briquettes will simultaneously promote soil regeneration both in water return process and in the process of decay by returning minerals and organic matter to the soil. Sapropel in composition of fertilizers participates as physiologically active binder of the base having high ion exchange and sorption properties. It is also an additional supplier of humic substances, amino acids, including aspartic, glutamic, as well as glycine, alanine and histidine, into nutrient medium of plants (Agafonova et al., 2015). The plants need macro elements such as H, C, O, N, Mg, K, Ca, P, microelements Cl, B, Fe, Mn, Co, Ni, Zn, Cu, Mo (Mohr & Schopfer, 1995). There is lack of Cu in the soil of Latvia. The elemental analysis on sapropel showed that lake sediments in comparison with peat have a higher N concentration. Higher concentrations in organic lake sediments are common for metals of natural origin such as Ca, Fe and Mg. Heavy metal concentration compared to its maximum permissible concentration in soil showed that the sapropelic sediments are not contaminated (Stankevica et al., 2012). The Plusons lake sapropel has a high concentration of such elements as K, Fe, Co, Ni, Zn, Cu that are important to a plant, but in comparison with sapropel the lake clay contains significantly more Mg, Ca, Mn and less heavy metals such as Cd, Zn, Cu (Tretjakova et al., 2017). Additional additives can be added in briquettes to adapt them to different soils.

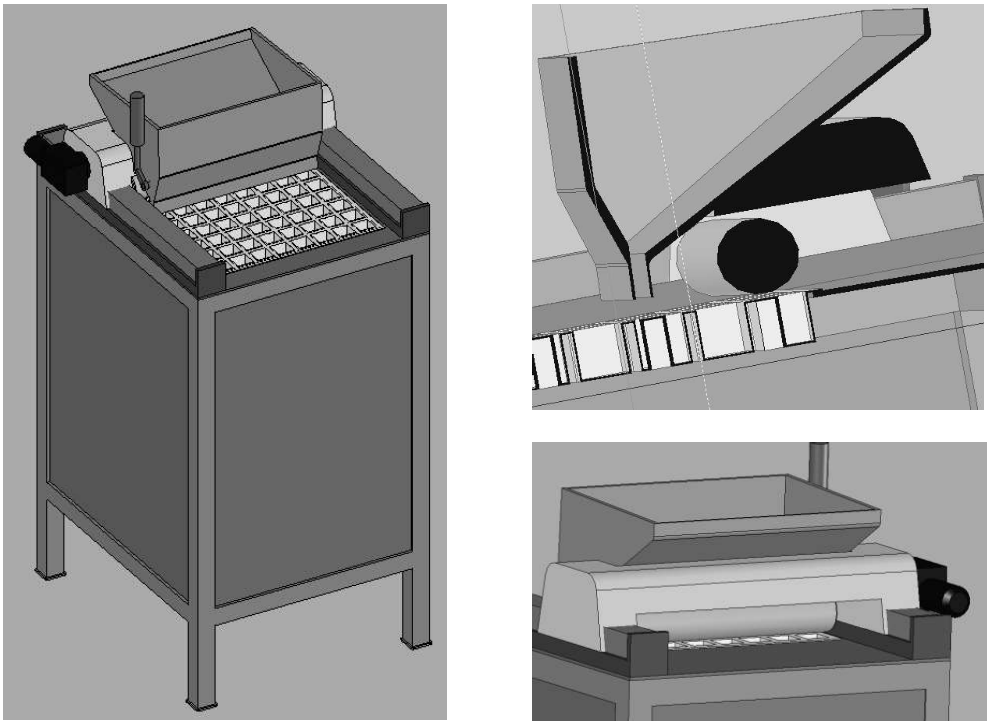

Figure 5. 3D model of briquetting equipment from different views.

Both sapropel and clay are microbiologically active and form an appropriate environment for microorganisms, while some clays have antibacterial properties, therefore microbiological stability of briquettes is important. Clay and sapropel contamination with microorganisms must be controlled, given that both can originate diseases or damage the product when it is stored. Results of the microbiological testing of the Plusons lake blue clays show that the presence of *Candida albicans, Pseudomonas aeruginosa, Staphilacoccus aureus has* not been stated. The number of mesophyll aerobic microorganisms varied < 1 to 3.6 X 10 CFU 0.1 g^{-1} (Tretjakova et al., 2017). *C. albicans, P. aeruginosa, S. aureus* have not been found in the sapropel of the Plusons lake. The number of mesophyll aerobic microorganisms varied from 3.6 x 10 to 5.0 x 10^2 CFU 0.1 g^{-1} (unpublished). Platonov et al. (2014) have demonstrated sapropel protective antibiotic activity against *Escherichia coli, S.aureus, C.diphterie* and fungi Candida. Strus et al. (2014) studies have shown a slight antibacterial activity of sapropel against selected test cultures (*E. coli, P. aeruginosa, Bacillus subtilis, Proteus vulgaris, C. albicans*). Sapropel has antimicrobial activity on *S. aureus* reference culture (Tretjakova et al., 2015). Fungus and actinomycetes synthesize antibiotics and sulfanilamides in sapropel (Stankeviča & Kļaviņš, 2013). Clay has antibacterial and antifungal properties. Clay shows antibacterial effect against *E. coli*, *S. aureus, P. aeruginosa* (Williams & Haydel, 2010; Parolo et al., 2011; Lafi & Al-Dulaimy, 2011). Antimicrobial testing of two clays proved that while one clay promotes bacterial growth, the other one eliminates bacteria or significantly inhibits its growth (Williams et al., 2008). The research results showed that some natural clay samples were devoid of antimicrobial and antifungal activity (Nikolajeva et al., 2013). The biological effects of clay minerals are influenced by their mineral composition and particle size (World Health Organization), therefore in-depth studies on the microbial resistance of the briquettes are needed.

Macroporous sapropel support seemed to be promising for adhesion of various microorganisms in order to prepare commercially attractive whole-cell biocatalysts (Kovalenko et al., 2016). In Žvagiņa et al. (2015) studies the clay was tested for the immobilization of bacteria. Muter et al. (2011), Muter et al. (2012), Nikolajeva et al. (2012) concluded in their studies that ceramic materials can be used for immobilization of microorganisms. Griba (2011) has compared in her study the clay extracted in Russia and Latvia and has come to conclusion that bacteria is more adhesive in clays extracted in Latvia. Thus, briquettes could be the material for the immobilization of such genera of bacteria as Azotobacter, Bacillus, Rhizobium and Streptomyces that promote the growth of plants. Further research is also needed in this area.

CONCLUSIONS

Briquettes containing 75% sapropel, 15% flax shive and 10% blue clay have the best mechanical and water desorption properties.

To obtain more durable briquettes, they should be dried slowly (for a long time without forced ventilation) at 20 °C.

After drying the briquettes must be places in a warehouse for at least 1 week. During this time sapropel hardness increases by 12%, then the briquettes can be transported.

Forming, drying, storing and transportation of briquettes should be done in the thermoplastic forms or other materials (ideally, a biodegradable material).

It is recommended to carry out further studies on the microbial activity of the briquettes and the possibilities of immobilization of bacteria that furthers the growth of plants.

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