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SUGARCANE BIOMASS AS A POTENTIAL CARRIER FOR DRUG DELIVERY SYSTEM

Non-steroidal anti-inflammatory drugs are well-known medications for reducing pain and a group of drugs that can cause mucosal damage of the stomach. The negative effects on the digestive system can be reduced by immobilization of drugs on various carriers, for instance, on the components of plant biomass, for the creation of drug delivery system. Plant biomass is a lignocellulosic complex consisting of lignin, cellulose and hemicellulose that can potentially be regarded as a carrier of pharmaceuticals. Sugarcane residues such as bagasse and straw are biomass by-products of the sugarcane industry. One of the prospective ways for their efficient utilization can include chemical processing with the aim of obtaining effective biosorbents or so-called carriers of different composition and structure. The aim of the work was to study the structural, morphological, and sorption properties of cellulose, lignin, and lignocellulose, derived from sugarcane biomass (bagasse and straw) by means of delignification and hydrolysis, as potential components for drug delivery system. Sugarcane straw samples show higher densities in comparison with bagasse samples. Both lignin samples from bagasse and straw have greater bulk and true density if compared to other materials from sugarcane biomass of cellulosic and lignocellulosic nature. The increase in adsorption pore volume in lignins is observed, being indicative of better sorption ability. Both samples of cellulose and lignocellulose from straw have greater pore structure if compared to the initial material. The values of sodium diclofenac sorption efficiency correlate with the values of pore volume for corresponding materials. Lignin from sugarcane straw, which shows greater porosity, has greater sorption properties. SEM images show that the initial materials and treated materials have complex morphology. FTIR spectra show a clear difference in the structure of lignocellulose, cellulose, and lignin from sugarcane bagasse and straw. The potential application of biopolymers from bagasse and straw as organic carriers of sodium diclofenac was studied. With this purpose, plant polymers were impregnated with an alcoholic solution of sodium diclofenac and the desorption process was investigated. The lignin sample from sugarcane straw has a longer period of drug release, which indicates the obtained effect of prolongation.

Keywords: *biomass, lignin, cellulose, sodium diclofenac, sorbent*

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Problem statement. Non-steroidal anti-inflammatory drugs (NSAID) are a special group of medications that relieve or reduce pain. NSAID are widely used for inflammation in chronic conditions but at the same time they can cause mucosal damage of the stomach [1]. This problem can be solved through immobilization of NSAID on various carriers for more safe and efficient drug delivery [2]. Natural biodegradable polymers obtained from different sources like proteins, carbohydrates and chemically modified carbohydrates can be used among others for this purpose [3].

Literature review. Biomass is the complex system of polysaccharides and biopolymers of aromatic nature [4]. Cellulose is a linear homopolymer consisting of D-glucose units linked with β(1→4) bonds. It is the main constituent of plant fibers and one of the most abundant polymers in nature. Unlike cellulose, lignin is a heteropolymer of aromatic nature of quite complex structure and one of the most abundant natural polymers of aromatic origin on Earth. Due to its aromatic structure, lignin can be considered as a potential source of valuable goods, chemicals, fuels, etc. It can be separated from cellulose by different pulping processes or by acidic hydrolysis. Due to the presence of various functional groups in cellulose and lignin, these biopolymers have a high sorption ability [5]. Lignocellulosic biocomplexe consisting of lignin, cellulose and hemicellulose in various proportions can potentially be regarded as an effective organic carrier of pharmaceuticals in the drug delivery systems [6].

Biomass residues of agriculture and food industry are a great source of lignin and cellulose with unique chemical and physical properties [7]. Sugarcane is an important crop in tropical and subtropical countries, and bagasse and straw are huge by-products of the sugarcane industry. Different methods for their utilization have been proposed, for example, bioconversion into fuels [8] or pulp production for the purpose of the paper industry [9]. Another way of prospective processing of sugarcane biomass is their deep chemical treatment and modification with the aim of obtaining effective biosorbents of different composition and structure for the application in ecology and medicine.

Aim. The aim of the work was to study the structural, morphological and sorption properties of cellulose, lignin, and lignocellulose, derived from sugarcane biomass (bagasse and straw) by means of delignification and hydrolysis, as potential components for drug delivery system.

Materials and methods. The sugarcane biomass (bagasse and straw i.e. tips and leaves), was obtained from Center Sugarcane Experimentation (Brazil). The biomass was ground, sieved and fraction from 0.5 to 1 mm was used in experiments.

Chemical composition of the sugarcane bagasse and straw was determined with the application of chlorite method, the Kurschner-Hoffer method, and TAPPI standards (T 222 cm-02, T 207 cm-99, T 212 cm-02, T 204 cm-97 and T 211 om-02) [10].

In order to isolate lignin, cellulose and lignocelluloses, sulphuric acid hydrolysis [11], hydrogen peroxide delignification in acetic acid and acetic acid hydrolysis, respectively, were used [10].

The textural characterization of the materials was carried out using the adsorption of benzene vapor in the desiccators [12].

The bulk density of the biomaterials was studied using a glass cup with an internal volume (V_b) of 100 cm^3. The cup was filled with biomaterials and weighed and the mass of the sample (m_s) was determined by subtraction of the mass of the empty cup. The bulk density (ρ_b, g/cm^3) of materials was determined as:

$$\rho_b = m_s / V_b \quad (1)$$

The true density (ρ_t, g/cm^3) was determined with the application of water displacement. Water was added to a glass cup filled with biomaterial to fill the empty space between the particles. True volume V_t was determined by subtraction of the volume of water and the true density was calculated as:

$$\rho_t = m_s / V_t \quad (2)$$

Changes in structure of biomass after treatment were investigated. A Cressington 208HR high-resolution sputter coater was used to coat the samples with gold. The thickness was controlled to be 2-3 nm. Then, a Field Emission SEM (Hitachi S-4800) was used to observe the morphological structures of the samples.

Fourier transform infrared (FTIR) spectra were obtained using a Perkin-Elmer Spectrum 2000 FTIR spectrometer (Waltham, MA, USA) equipped with an ATR system, Spectac MKII Golden Gate (Creecstone Ridge, GA, USA). The samples were analysed at wavelengths ranging from 600 to 4000 cm^{-1}. All spectra were obtained from dry samples which were subjected to 16 scans at a resolution of 4 cm^{-1} and an interval was 1 cm^{-1}. FTIR were recorded at room temperature. Before data recording, background scanning was done for background correction.

The sorption ability of modified sugarcane biomass towards sodium diclofenac was investigated in batch experiments. For this purpose, a model solution with sodium diclofenac concentration of 32 mg/l was used. The solution was prepared on a phosphate buffer at pH 6.0. The mass of sorbent was 0.2 g, the volume of the solution was 50 ml. Experiments were carried out during 24 h at 25 °C. Initial and equilibrium concentrations of sodium diclofenac of were determined by spectrophotometric method at λ = 275 nm.

The possibility of application of biopolymers such as lignin cellulose and lignocellulose as carriers of sodium diclofenac was studied. With this purpose, plant polymers were impregnated with an alcoholic solution of sodium diclofenac with the concentration of drug 32 mg/l. Impregnation of the samples was performed at periodic manual stirring at the temperature of 80 °C until complete evaporation of the solvent. The obtained composite materials were compressed into tablets. Desorption process was studied using a phosphate buffer solution with pH 6.8 at 37 °C.

Results and discussion. The chemical composition of both sugarcane biomass is shown in Table 1. As can be seen, the contents of cellulose and hemicelluloses in sugarcane straw are lower than in bagasse. The sugarcane bagasse is characterized by the presence of a low portion of extractives (resins, wax and fats). Analysis of straw composition showed that it contains higher content of extractives soluble in hot water and in 1 % NaOH if compared to bagasse. As can be seen, the lignin content is greater in sugarcane bagasse. The chemical composition of sugarcane bagasse agrees with data reported by other authors [13].

Table 1 – Chemical composition of sugarcane bagasse and straw

Component	Content, %	
	Bagasse	Straw
Hollocellulose	75.9	67.8
Cellulose	42.1	37.2
Lignin	21.4	19.6
Ash	2.3	7.8
Ethanol-benzene extractives	0.8	4.3
Hot water solubility	8.2	17.5
1 % NaOH solubility	28.4	38.7

In order to isolate lignin, cellulose and lignocelluloses, sulphuric acid hydrolysis, hydrogen peroxide delignification in acetic acid and acetic acid hydrolysis, respectively, were used. The content of the main components such as cellulose and lignin in materials obtained is shown in Table 2. The density of the initial biomasses and biopolymers as well as specific surface area and adsorption pore volume are also given in Table 2. At moisture content 8%, all samples differed from each other in values of bulk and true densities. Initial straw shows higher density in comparison with bagasse. Both lignin samples have greater bulk and true density in comparison with other materials. The increase in specific surface area and adsorption pore volume for lignin samples was also observed. Cellulose and lignocellulose from sugarcane straw have greater pore structure when compared to initial biomass. All samples from straw are characterized by greater ash content in comparison with samples from bagasse.

Table 2 – Structural characterization of sugarcane bagasse-derived materials

Materials	Parameters					
	Lignin/cellulose content, %	Bulk density, g/cm^3	True density, g/cm^3	Specific surface area, m^2/g	Adsorption pore volume, cm^3/g	Ash, %
Sugarcane bagasse						
Initial biomass	21.4/42.1	0.049	0.096	1.35	0.08	2.3
Lignin	90.5/3	0.121	0.412	2.98	0.19	6.5
Cellulose	3.03/67.3	0.048	0.069	1.25	0.15	2.5
Lignocellulose	29/34.3	0.116	0.383	1.23	0.11	1.27
Sugarcane straw						
Initial biomass	19.6/37.2	0.087	0.119	1.93	0.05	7.8
Lignin	81.8/1	0.141	0.159	6.68	0.24	17.2
Cellulose	8.6/57.5	0.038	0.138	5.37	0.13	9.7
Lignocellulose	27.7/30.3	0.124	0.487	2.68	0.18	6.06

SEM images (Fig. 1) show that original biomass and treated biomaterials have complex morphologies. The surface of lignocellulosic materials after acetic acid treatment of sugarcane biomass has lower content of small impurities and the surface is more heterogeneous (Fig. 1A2 and 1B2) After the treatment with the mixture of hydrogen peroxide and acetic acid, the surface morphology of both cellulosic materials changed significantly (Fig. 1A3 and 1B3) due to delignification. Lignin samples characterized by the presence of condensed lignin particles of various sizes (Fig. 1A4 and 1B4).

FTIR spectra for initial biomass and for treated biomaterials based on sugarcane bagasse and straw are shown in Fig. 2.

The stretching vibrations of hydrogen bonds in -OH groups in the region of 3000–3700 cm^{-1} were identified in all the spectra but less pronounced in lignin. C-H stretching vibration at 2924 cm^{-1} due to hydrogen bonding and the water absorption at 1641 cm^{-1} were also identified in all the spectra [14]. At the same time peak at 2924 cm^{-1} can be assigned to C-H stretching in -CH_3 groups and peak at 2851 cm^{-1} to C-H stretching in -CH_2- groups of lignin and esterified polysaccharide [15]. The peak at 1034 cm^{-1} can be attributed at the same time to C-O, C-C stretching and C-OH bending in polysaccharides. Bands in the range of 1463-1430 cm^{-1} can be assigned to asymmetric C-H deformation of -CH_3 and

-CH_2- groups of aliphatic nature. The presence of lignin in biomass and in treated materials can be confirmed by the band at 1510-1513 cm^{-1} and at 900-700 cm^{-1} which assigned to the stretching of lignin rings [14].

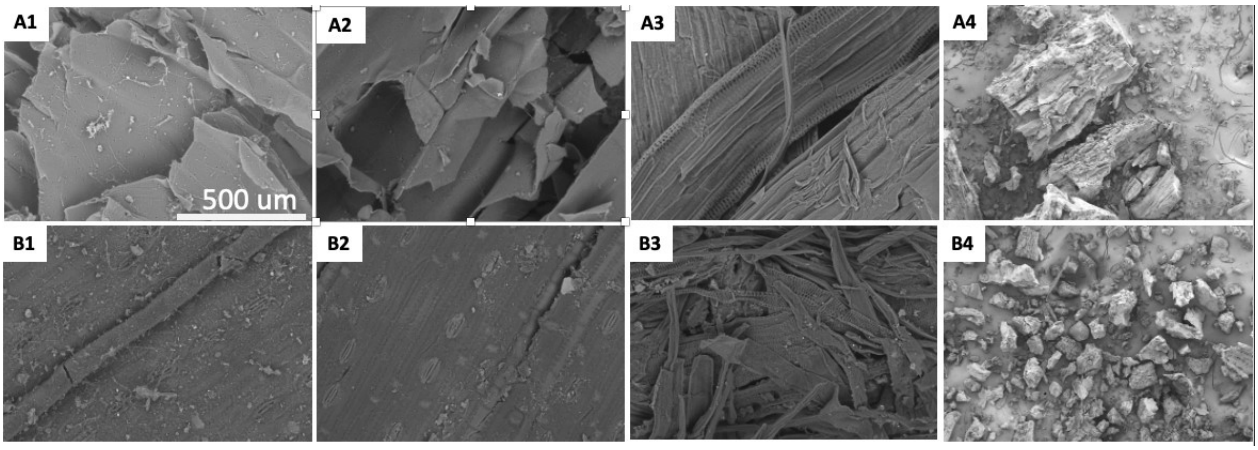

Fig. 1 – SEM images for (A) bagasse and (B) straw: (1) initial biomass, (2) lignocellulose after acetic acid treatment, (3) cellulose after delignification with the mixture of hydrogen peroxide and acetic acid, (4) lignin after acid hydrolysis

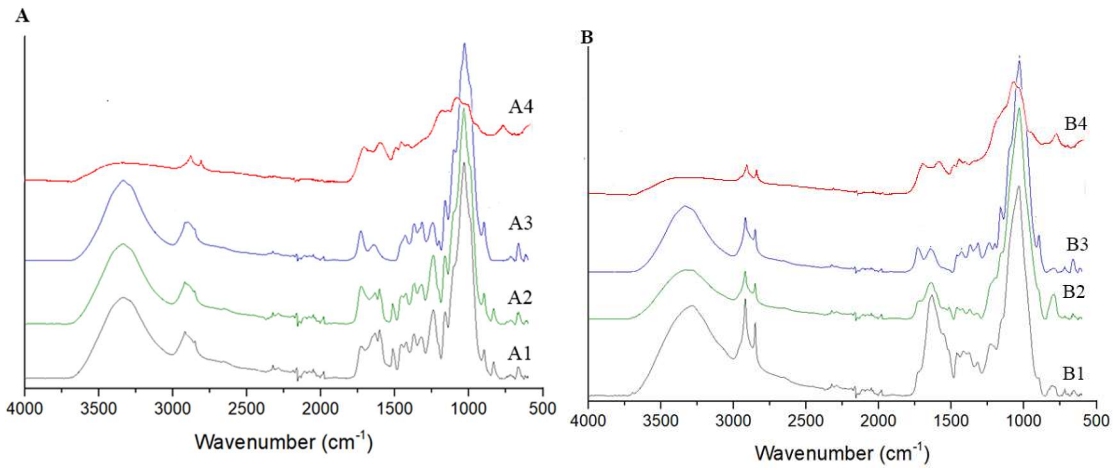

Fig. 2 – FTIR spectra for (A) bagasse and (B) straw: (1) initial biomass, (2) lignocellulose after acetic acid treatment, (3) cellulose after delignification with the mixture of hydrogen peroxide and acetic acid, (4) lignin after acid hydrolysis

The main structural difference between the biomass and the treated materials was that the band in the ranges 1510-1513 cm^{-1} and 900-700 cm^{-1} was not observed in the cellulosic samples confirming the efficient delignification, and the lignin samples showed intense absorbance in that regions.

The results of the investigation of sorption ability of initial biomass and obtained biopolymers are shown in Table 3. According to the results, the values of the efficiency of sodium diclofenac sorption are correlated with the values of the specific surface area and pore volume. The greater sorption ability corresponds to lignin from sugarcane straw, which shows greater structural porosity.

The possibility of application of biopolymers such as lignin cellulose and lignocellulose as carriers of sodium diclofenac was studied. The kinetics of the release of the medication from the obtained composites indicate the direct dependence of the efficiency of the sodium diclofenac desorption on the porosity of the organic carriers (Fig. 3).

Lignin sample from sugarcane straw shows longer period of drug release indicating the achievement of the prolongation effect.

Table 3 – Sorption properties of biopolymers from sugarcane residue

Indicator	Materials from bagasse			Materials from straw		
	Lignin	Cellulose	Lignocellulose	Lignin	Cellulose	Lignocellulose
Efficiency of sodium diclofenac sorption, %	36.5	15.6	2.13	85.8	21.5	44.4

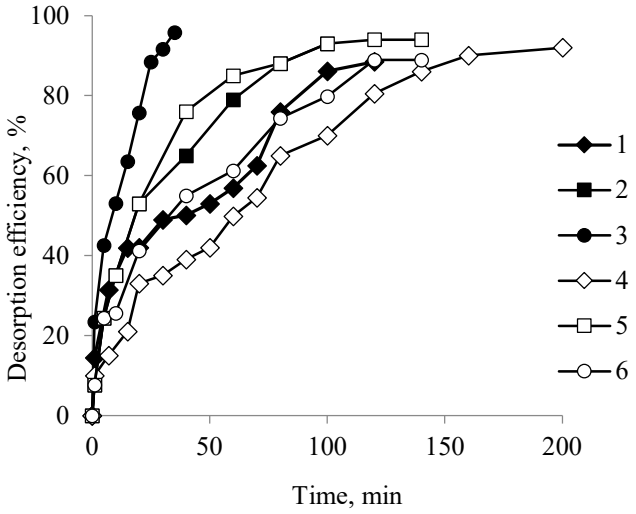

Fig. 3 – Sodium diclofenac efficiency desorption from the volume of lignin (1), cellulose (2) and lignocellulose (3) from sugarcane bagasse and lignin (4), cellulose (5) and lignocellulose (6) from sugarcane straw

The obtained data give ground to assert that sugarcane lignin can be effectively used in drug delivery systems due to longer period of NSAID desorption.

Conclusions. This work suggests that efficient carriers for drug delivery systems can be prepared from sugarcane biomass. The treatment of bagasse and straw with acetic acid, mixture of hydrogen peroxide and acetic acid, acid hydrolysis gave materials with different structural features and sorption ability correlated with the porosity of the materials. The greater sorption ability towards sodium diclofenac corresponds to lignin sample from sugarcane straw, which also shows greater structural porosity. Greater adsorption pore volume can also be the main reason for longer period of drug release from lignin-carrier based on sugarcane straw indicating the achievement of the prolongation effect. These results may serve as a basis for the development of efficient methods for the utilization of sugarcane residues in the pharmaceutical industry.

Prospects for further research. Further study of the structure of materials from sugarcane biomass to determine the content of various functional groups and determine the possibility of chemical modification of biosorbents for preparation of highly effective carriers for drug delivery system is quite promising.

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ЛІГНОЦЕЛЮЛОЗНА БІОМАСА ЯК ПОТЕНЦІЙНИЙ НОСІЙ ДЛЯ СИСТЕМИ ДОСТАВКИ ЛІКІВ

Як відомо, нестероїдні протизапальні засоби – це ліки для зменшення болю та група препаратів, які можуть викликати ураження слизової оболонки шлунка. Негативний вплив на систему травлення можна зменшити шляхом іммобілізації ліків на різних носіях, наприклад на компонентах рослинної біомаси, для створення системи доставки ліків. Рослинна біомаса – лігноцелюлозний комплекс, що складається з лігніну, целюлози та геміцелюлоз, потенційно може бути використаний як носій фармацевтичного препарату. Залишки цукрової тростини, такі як багаса і солома, є побічними продуктами переробки цукрової тростини. Одним із перспективних шляхів їх ефективного використання може бути хімічна обробка з метою отримання ефективних біосорбентів або так званих носіїв різного складу та структури. Метою роботи було вивчення структурних, морфологічних та сорбційних властивостей целюлози, лігніну та лігноцелюлози, отриманих із біомаси цукрової тростини (багаси та соломи) шляхом делігніфікації та гідролізу як потенційних компонентів системи доставки ліків. Зразки соломи цукрової тростини демонструють більш високу щільність у порівнянні зі зразками багаси. Обидва зразки лігніну з багаси та соломи мають більшу щільність порівняно з іншими матеріалами з біомаси сахарної тростини целюлозної та лігноцелюлозної природи. Спостерігається збільшення об'єму адсорбційних пор у лігнінах, що вказує на кращу здатність до адсорбції. Обидва зразки целюлози та лігноцелюлози з соломи мають більшу пористість у порівнянні з вихідним матеріалом. Значення ефективності сорбції диклофенаку натрію корелюються із значеннями об'єму пор для відповідних матеріалів. Більшими сорбційними властивостями характеризується лігнін із соломи цукрової

тростини, який має більшу пористість. Зображення СЕМ показують, що вихідні та оброблені матеріали мають складну морфологію. Спектри ІК показують чітку відмінність у структурі лігноцелюлози, целюлози та лігніну багаси та соломи цукрової тростини. Досліджено можливість застосування біополімерів з як органічних носіїв диклофенаку натрію. З цією метою рослинні полімери просочували спиртовим розчином диклофенаку натрію та досліджували процес десорбції. Зразок лігніну із соломи цукрової тростини має більший період вивільнення препарату, що свідчить про отриманий ефект пролонгації.

***Ключові слова**: біомаса; лігнін; целюлоза; диклофенак натрію; сорбент*

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