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Silica synthesis by the sol-gel method and its use in the preparation of multifunctional biocomposites

Research Article

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Abstract: This study focuses on the optimization process of silica synthesis using the sol-gel method while applying a statistical design of experiments which was based on a multilevel mathematical model. The product obtained in the process of optimized synthesis, characterized by the best dispersive and morphological parameters, was used for the preparation of organic/inorganic composites. The organic precursor was Kraft lignin, a high-molecular natural polymer. Synthesis of silica/lignin biocomposites was carried out by three proposed methods. The physicochemical properties and dispersive–morphological properties of each product were determined using the following available methods: Scanning Electron Microscopy – SEM, Non-Invasive Back-Scattering – NIBS, Fourier Transform Infrared Spectroscopy – FT-IR, Thermogravimetric analysis – TG and others. The electrokinetic and thermal properties of the biocomposites sufficed to be applied for example, as a cheap and biodegradable polymer filler. Further areas of application of these composites were sought, especially in electrochemistry as the advanced electrode materials.

Keywords: *Sol-gel method • Silica • Silica/lignin biocomposites • Physicochemical and structural properties*

1. Introduction

Modern technology and engineering aim to develop materials with improved physicochemical properties and established optimum methods for their production. A technique that assists the development of methods for obtaining an optimum product with minimum investment of time and/or energy is that of a statistical design of experiments. This technique is a modified investigation which is followed by the observations and measurements in regard to its properties.

Organic/inorganic composites are materials of high quality and functionality. These composites are made frequently from an inorganic component and an organic modifier. One of the most attractive and often used inorganic materials is silica which offers many unique properties such as hardness, chemical resistance, high reactivity and highly developed surface area. Silicon dioxide can be obtained in many processes, resulting in products with different morphological properties; these processes include the sol-gel method [1] a high-temperature process [2], and precipitation in a polar or nonpolar medium [3-5]. The above methods for silica synthesis enable the production of silica with a specific particle shape, size and dispersion in relation to the particle size distribution [6]. This support has been used primarily in the production of materials for everyday use. For example, silica has been widely applied in the glass and ceramic industries, metallurgy and construction [7].

In addition to cellulose, lignin is the main component of wood tissue and one of the most abundant aromatic biopolymers on Earth [8]. The annual production of lignin, which stems mainly from industrial waste, exceeds 50 million tons [9]. Lignin is a complex phenol biopolymer [10] formed in a polymerization reaction via the so-called phenylpropanoid pathway [11,12]. The exact mechanism of lignin synthesis is described in [13]. As lignin has

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different functional groups in its structure, it has become an interesting material with prospects of application in many areas, including adsorption of heavy metals [14-16], as a substrate for the synthesis of low-molecular organic compounds [17,18], and in particular, as an active component of biocomposites [19-22]. Lignin is isolated from wood by hydrolysis or extraction [23], which gives different forms of lignin, including Klason lignin [24], cellulolytic enzyme lignin [25], Brauns lignin [26], milled wood lignin [27], and Kraft lignin [28]. Kraft lignin, which is a by-product of the process of paper production, has been widely applied, although not on a large scale [29]. There are also alternative methods of obtaining and purifying lignin such as, employing ultrafiltration or nanofiltration [30].

The fact that lignin is of natural origin and silica is an inert material, also contributes to the interest in their use in modern chemical technology. Modern chemical technology promotes sustainability and "green chemistry" in the production of advanced materials [31]. This paper attempts to optimize the sol-gel process of silica synthesis based on the use of a multilevel mathematical model. The optimization of the process made it possible to obtain a product with the best dispersive–morphological properties. This product was used for the synthesis of silica/lignin biocomposite materials. The obtained composites were subjected to comprehensive physicochemical analysis using multiple methods, including Non-Invasive Back-Scattering – NIBS, Scanning Electron Microscopy – SEM, Fourier Transform Infrared Spectroscopy – FT-IR, Thermogravimetric analysis – TG and Electrophoretic Light Scattering – ELS.

2. Experimental procedure

2.1. Synthesis of silica by the sol-gel method

Silica was synthesized by a modified Stöber method involving simultaneous hydrolysis and condensation of tetraethoxysilane TEOS (analytical grade) purchased from Sigma-Aldrich (Germany), in a medium of 95% ethyl alcohol (analytical grade; Chempur, Poland) and 25% ammonia (analytical grade; POCh SA, Poland). The following process was optimized by testing the effects of the following factors to obtain a product with the best morphological properties: .

- Quantity of TEOS, in three variants: 11, 14 and 17 cm^3;
- Quantity of NH_4OH, in three variants: 11, 14 and 17 cm^3;
- Temperature of the process, in two variants: 20 and 40°C;
- Time of the process, in three variants: 30, 60 and 90 minutes;
- Type of TEOS dosing into the reaction medium, in two variants (0/1): dosing with a peristaltic pump (1) and addition of a single portion of TEOS to the reaction medium (0).

The amount of ethyl alcohol used as the reaction medium was kept constant (50 cm^3). When dosing TEOS with a peristaltic pump, the rate was set to ensure that the whole volume of TEOS would be introduced into the reaction medium in one half of the process time. With the use of a demonstration version of the DesignExpert program from Stat-Ease Inc., a mixed experimental list was generated. The list was generated based on a multilevel mathematical model and intended to reduce the number of samples prepared and studied from 108 to 39.

The reaction medium consisted of 50 cm^3 of ethyl alcohol and the appropriate quantity of ammonia solution. The medium was subjected to intense stirring (1800 rpm) using a EUROSTAR digital stirrer (IKA-Werke GmbH & Co.) and heated/cooled to the desired temperature in a water bath (JULABO Labortechnik GmbH). The appropriate quantity of TEOS was either added (0) or dosed (1) into the medium, and the reaction was conducted for the time set for the sample in question. The white silica precipitate obtained was separated from the post-reaction mixture by filtration under reduced pressure. The product was washed out three times by repulpation with ethyl alcohol and a suitable quantity of water. The SiO_2 precipitate was dried in a stationary drier (Memmert) at 105°C for about 12 h to eliminate moisture.

2.2. Synthesis of silica/lignin biocomposites

The process of optimization based on the mathematical multilevel model made it possible to determine the parameters of which the product with the best dispersive–morphological properties could be obtained. The silica obtained was modified with *N*-2-(aminoethyl)-3-aminopropyltrimethoxysilane (Sigma-Aldrich). The appropriate amount of the modifying substance was first hydrolyzed in a methanol/water (4:1, v/v) system, and it was then deposited on the silica surface by the method described in [32]. Another method of silica modification, with silane coupling agents, has been proposed [33]. The silica modified with aminosilane was used in further studies as a support. The final synthesis of composites, based on the optimized silica support and Kraft lignin (Sigma-Aldrich), was performed in three variants – the three proposed methods – each time using a quantity of lignin corresponding to 1 part to 5 parts by weight of SiO_2. This weight ratio has been found to ensure the best dispersive and morphological properties of the products [20].

2.2.1. Method I

A reactor was charged with solution 1, containing 1.0 g of lignin dissolved in a dioxane/water mixture with a ratio of 1:1 (v/v). Solution 2 (oxidizing) was made up of 1.3 g of sodium periodate (Sigma-Aldrich) dissolved in 30 cm^3 of distilled water. Solution 2 was dosed into solution 1 under vigorous stirring (1000 rpm). An important condition was for the lignin activation to be performed in the dark. After completing the dosage, the system continued to be stirred for 30 minutes. Then the aminosilane-activated silica was added to the system, and the contents were stirred for about 1 h. After this time, the solvents were evaporated in a vacuum evaporator (Büchi Labortechnik GmbH). The precipitate obtained was dried at 105°C for about 1 h.

2.2.2. Method II

A reactor was charged with solution 1 (oxidizing) made of 1.3 g sodium periodate dissolved in 30 cm^3 of distilled water. Solution 2 was made from 1.0 g lignin dissolved in a mixture of dioxane/water at a ratio of 1:1 (v/v). Solution 2 was dosed into solution 1 using a peristaltic pump for 15 minutes, and then followed by the whole content being vigorously stirred (1000 rpm) for 15 minutes. The process was performed in the dark. The obtained lignin solution was deposited by atomization onto the previously prepared modified silica support. The obtained suspension was subjected to solvent evaporation in a vacuum evaporator, and the precipitate was dried for about 1 h at 105°C.

2.2.3. Method III

The third method was an attempt to directly produce a silica/lignin biocomposite. In this procedure a reactor was charged with 5.0 g silica modified with aminosilane, 1.0 g lignin and 1.3 g sodium periodate. The contents were stirred, adding dropwise 40 cm^3 of a dioxane/water solution at 1:1 (v/v) using a peristaltic pump. This approach was used to ensure that the process of addition would last for 15 minutes. The solvents were evaporated in a vacuum evaporator, and the precipitate was dried in a stationary drier for 1 h at 105°C.

2.3. Physicochemical characterization of silica and final biocomposites

SEM images of all samples were made on a scanning electron microscope (Zeiss EVO40). The information inferred from the SEM images was verified by the particle size distribution, measured using a Zetasizer Nano ZS and Mastersizer 2000 (Malvern Instruments Ltd.), employing the non-invasive back scattering (NIBS) and laser diffraction methods.

The silica/lignin biocomposites obtained using the three methods described above were also subjected to SEM image analysis and particle size distribution determination in the range 0.6–6000 nm using a Zetasizer Nano ZS and in the range 0.2–2000 μm using a Mastersizer 2000. The electrophoretic mobility was measured to evaluate the dispersion stability, and then the zeta potential values were calculated using the Henry equation. The measurements were made using a Zetasizer Nano ZS (Malvern Instruments Ltd.) equipped with an autotitrator. Thermal analysis was performed using a Jupiter STA449F3 (Netzsch). Precise determination of the thermal stability of biocomposites is of key importance for their applications. To verify that the products obtained were indeed silica/lignin biocomposites, and to evaluate the efficiency of the process of their production, the FT-IR spectra of the products were obtained using an Alpha apparatus with a Platinum ATR reflection attachment (Bruker Optics GmbH). The FT-IR spectra confirmed the presence of characteristic functional groups, which provides that the proposed methods of functionalization are effective.

3. Results and discussion

3.1. Dispersive–morphological characterization of silicas. Statistical analysis

Prior to the synthesis of biocomposites, the synthesis of silica was performed by a modified Stöber method. The parameters of the synthesis were optimized to obtain the best physico-chemical properties, in particular high homogeneity and small particle size distribution, which were adopted as criteria for the use of a sample as a support. Table 1 presents the fundamental parameters characterizing the silica samples, including the mean size of particles, verified by SEM images taken for all samples.

The samples of which the mean particle size was determined by the Zetasizer Nano ZS was greater than 1 μm, namely samples showing a tendency for agglomeration, were rejected. This criterion for sample acceptance (mean particle size smaller than 1 μm) permitted a reduction in the number of samples subjected to statistical analysis. Statistical analysis was performed concerning the correlations between the pore size distribution in particular samples and the assumed

Table 1. List of silica samples and the Z–Average particle size results.

Sample No.	Amount of TEOS (cm^3)	Amount of NH_4OH (cm^3)	Time (min)	Temperature (°C)	Dosage (0/1)	Z–Average* (nm)
1	11	11	90	40	0	800
2	11	11	30	20	1	rejected
3	11	17	30	40	0	rejected
4	14	17	30	40	1	588
5	17	14	90	20	0	rejected
6	11	17	60	20	1	625
7	11	14	60	40	0	875
8	11	14	90	40	0	rejected
9	17	17	60	20	0	rejected
10	17	11	30	20	0	rejected
11	14	14	90	40	0	rejected
12	14	11	60	20	1	584
13	17	11	60	40	0	510
14	14	14	60	20	0	rejected
15	11	11	60	40	1	609
16	14	11	90	20	0	rejected
17	14	11	30	40	0	713
18	14	17	90	40	0	rejected
19	17	17	30	20	1	933
20	17	11	90	20	1	975
21	17	14	60	20	1	rejected
22	11	14	90	20	1	640
23	17	14	30	40	0	rejected
24	14	17	60	40	0	rejected
25	14	14	30	20	1	rejected
26	11	11	60	20	0	rejected
27	14	11	90	40	1	312
28	17	17	60	40	1	500
29	14	17	90	20	1	rejected
30	11	17	90	40	1	rejected
31	17	17	90	40	0	rejected
32	14	17	30	20	0	rejected
33	17	14	90	40	1	543
34	14	14	60	40	1	rejected
35	17	11	30	40	1	290
36	11	14	30	40	1	478
37	11	17	90	20	0	rejected
38	11	14	30	20	0	rejected
39	14	17	60	20	1	rejected

* *„rejected" means particles showing a tendency to agglomerate, whose Z–Average size is greater than 1000 nm*

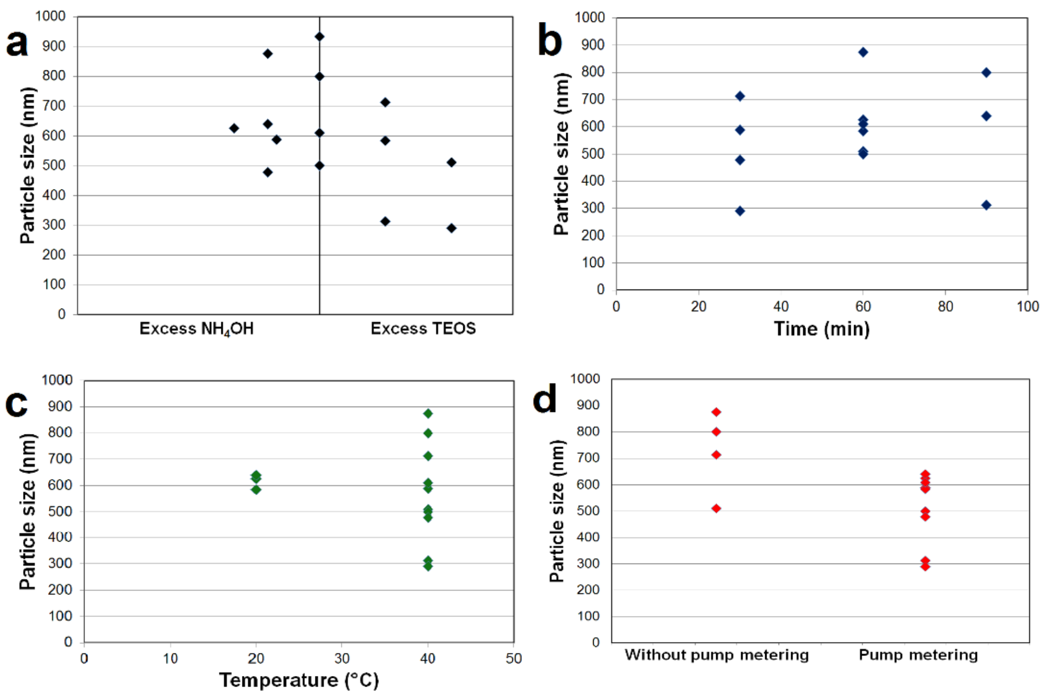

Figure 1. Results of statistical analysis. Particle size distribution *versus* the following initial variables: (a) excess of one of the reagents, (b) time, (c) temperature, and (d) mode of TEOS dosing.

variable parameters of the reaction: the excess of reagents, time of reaction, temperature and mode of TEOS introduction. The results are presented in Fig. 1.

The use of ammonia solution in excess over TEOS (Fig. 1a) resulted in obtaining five samples with spherically-shaped particles. The mean particle size distribution of these samples covered the range 478–875 nm. When the substrates NH_4OH:TEOS were used in the ratio 1:1 v/v, four samples satisfied the criterion of acceptance, and their particle size distribution covered the range 500–933 nm. The points on the right side of Fig. 1a were obtained for the samples synthesized in the presence of excess TEOS with respect to NH_4OH. These samples had the smallest particles, with sizes below 300 nm, and displayed high uniformity.

As depicted in Fig. 1b, the time of reaction had no significant influence on the particle size in the samples. The smaller number of samples satisfying the criterion of acceptance for a reaction time of 90 minutes indicates that when the system is left longer in the reaction medium, it shows a greater tendency to form highly undesirable clusters of agglomerates. Moreover, for technological reasons it should be acknowledged that a time of 30 minutes is sufficient to obtain a product with beneficial dispersive–morphological properties.

To test the effect of temperature of synthesis, the samples were obtained at two different temperatures, 20 and 40°C. As seen in Fig. 1c, a greater number of samples satisfying the criterion of acceptance were obtained at the higher temperature of synthesis. Moreover, it was indicated by the larger number of positive results obtained at the higher temperature and the wide range of particle sizes (290–980 nm), that the elevated temperature of the reaction medium permitted spherically-shaped silica samples to be obtained , but had no direct influence on their size.

The final parameter of synthesis the effect of which was tested, was the mode of introduction of TEOS into the reaction medium (Fig. 1d). As many as nineout of 13 samples meeting the particle size criterion were obtained when TEOS was dosed into the reaction medium. Moreover, the systems obtained with TEOS and introduced by dosing had smaller-sized particles than those obtained when TEOS was added in a single portion.

As the results above signify, the time of reaction has no effect on the size and morphology of the silica particles. All time variants applied had similar particle sizes and shapes. The main factors determining the size and shape of silica particles were the mode of TEOS introduction (size) and elevated temperature (regular spherical shape). The use of excess of TEOS with respect to NH_4OH permitted the obtaining of silicas with the smallest diameters (close to 300 nm); however, no

Table 2. Silica samples of the best dispersive–morphological properties and their synthesis parameters.

Sample No.	Amount of TEOS (cm^3)	Amount of NH_4OH (cm^3)	Time (min)	Temperature (°C)	Dosage (0/1)	Z-Average (nm)
27	14	11	90	40	1	312
35	17	11	30	40	1	290

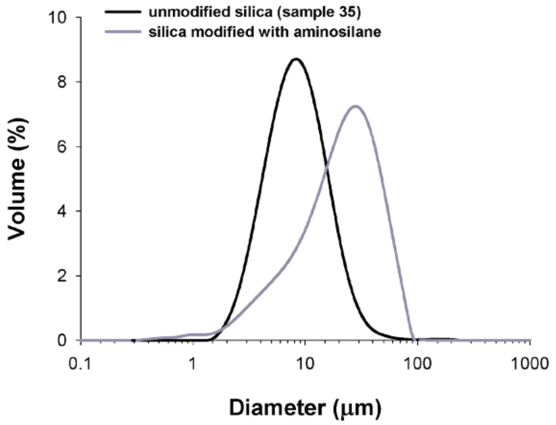

Figure 2. Particle size distributions of unmodified and aminosilane-grafted silicas (Mastersizer 2000).

direct correlation between these parameters was found. On the basis of the statistical analysis two silica samples with the best morphological and dispersive properties were chosen; their parameters are illustrated in Table 2.

The sample that had the more conveniently technological production was selected of the two. Namely sample 35 was obtained in a reaction time of 30 minutes. The parameters of this sample are given in Figs. 2 and 3.

Additionally, the particle size distribution was measured for this silica sample modified with aminosilane. According to the results of measurements on a Mastersizer 2000 (Fig. 2) this sample is uniform and has a narrow particle size distribution. The measurements on a Zetasizer Nano ZS (Fig. 3a) show that the unmodified sample 35 is built of particles with diameters in the range 220–531 nm, with the main contribution of 21.4% coming from particles 295 nm in diameter. SEM images of unmodified and modified sample 35 (Figs. 3c, 3d) fully confirm the above results. Fig. 3b presents the particle size distributions for silica (sample 35) modified with aminosilane. The modification was applied to enhance the affinity of silica to lignin activated with sodium periodate. The results are as expected, and show an increase in the volume contribution of larger particles as a result of the modification.

3.2. Analysis of properties of silica/lignin biocomposites

3.2.1. Dispersive-morphological properties

Silica sample 35 modified with aminosilane was used to prepare silica/lignin biocomposites. Three methods for biocomposite production were investigated, as described above in chapter 2.2. Table 3 presents the parameters of the samples obtained by the three methods.

Analysis of these data shows that method II produces biocomposite samples with the best parameters. A detailed characterization of the biocomposites obtained by this method at different temperatures, samples 3 and 4, is shown in Figs. 4 and 5 respectively.

The particle size distributions obtained for the two samples on a Mastersizer 2000 (Fig. 4) revealed similar ranges of particle diameters: 1.0–52.5 µm and 1.0–45.8 µm for samples 3 and 4 respectively. Better properties were recorded for composite 4, obtained at the higher temperature. The difference was a greater number of particles of smaller sizes in the sample synthesized at the higher temperature (composite 4). In sample 3, 50% of the volume was occupied by particles with diameters smaller than 10.5 µm and 90% by particles with diameters smaller than 27.1 µm, while in sample 4, the corresponding values were 7.8 µm and 21.1 µm. These data are consistent with the particle size distributions measured on a Zetasizer Nano ZS (Figs. 5a, 5b). Particles with diameters smaller than 200 nm accounted for 42.1% of sample 3 and 53.7% of sample 4. The SEM images of composites 3 and 4 (Figs. 5c, 5d) confirmed the presence of primary particles, aggregates (up to 1000 nm) and agglomerates (diameters higher than 1000 nm) in both samples.

3.2.2. Electrokinetic characterization

The dispersion stability of the samples was evaluated on the basis of zeta potential measurements. Results are presented in Fig. 6 for the composites obtained by all three proposed methods , and – for comparison purposes – for the native precursors (silica and lignin).

As Fig. 6a shows, the isoelectric point for unmodified silica (sample 35) was attained at a pH close to 4.5.

Table 3. Dispersive characteristic of silica/lignin biocomposites obtained by different methods.

Biocomp.No.	Process temperature (°C)	Dispersive properties				
		Particle size distribution range from Zetasizer Nano ZS (nm)	Particle diameter from Mastersizer 2000 (μm)			
			d(0.1)	d(0.5)	d(0.9)	D[4.3]
METHOD 1						
1	20	68–106 1110–4800	2.6	9.4	24.7	11.8
2	40	91–220 1720–5560	3.2	15.3	32.9	16.9
METHOD 2						
3	20	79–122 955–5560	2.5	10.5	27.1	12.8
4	40	68–220 955–1720	2.5	7.8	21.1	10.1
METHOD 3						
5	20	59–91 1990–5560	3.2	14.8	31.7	16.3
6	40	79–106 396–712	4.3	15.9	32.0	17.3

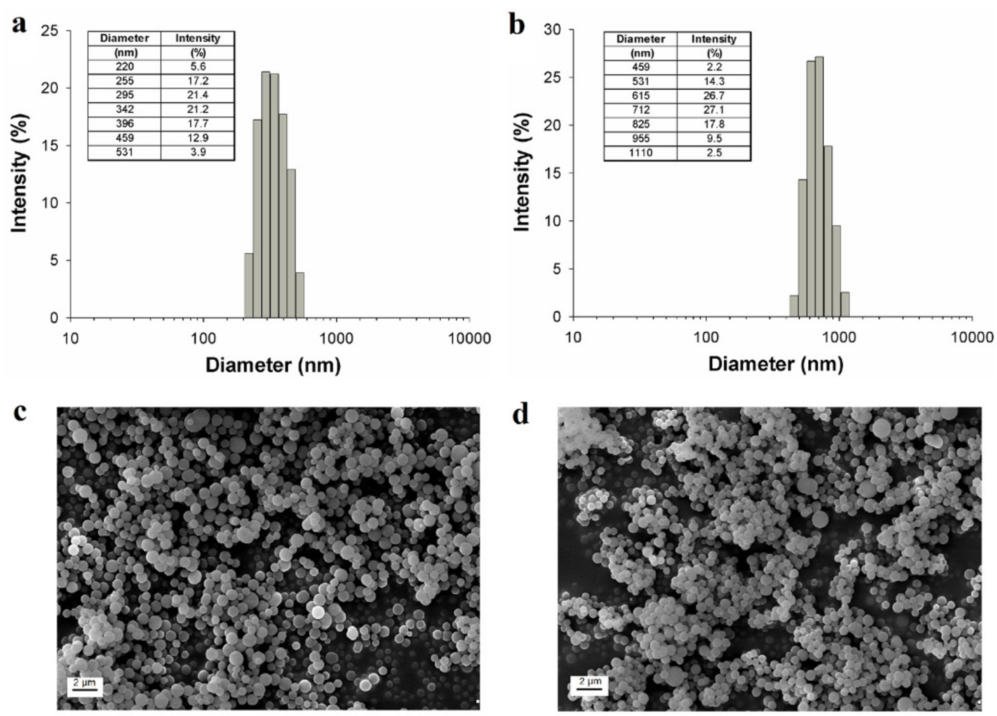

Figure 3. Particle size distributions measured on a Zetasizer Nano ZS and SEM images of (a, c) unmodified silica (sample 35) and (b, d) silica (sample 35) modified with aminosilane.

This value is somewhat raised compared with the data obtained for the silica synthesized by precipitation in a polar or nonpolar medium. This can be explained by the effect of the ammonia used as the medium in the sol-gel method. The use of ammonia results in incorporation of $-NH_3^+$ groups into the silica skeleton, as was established in [34,35]. Thanks to the modification with silane, the modified sample 35 displayed a high stability in the acidic pH range, while the unmodified sample 35 showed a high stability for the alkaline pH range. Lignin displayed high stability for pH>2. The zeta potential characteristics recorded for silica-lignin composites are presented in Fig. 6b. These data show that the samples obtained at the lower temperature have a tendency towards smaller zeta potential values. The influence of temperature on zeta potential values at a given pH was previously indicated in [36,37]. The composites were stable at pH values higher than 3, and thus over the whole pH range. The composites obtained at 40°C showed a very interesting rapid decrease in zeta potential in the range pH 2–5, while for higher pH values they were electrokinetically stable (zeta potential below –30 mV). Isoelectric points were observed for only one of the produced six composite samples : for sample 4 at a pH close to 2.2. The most stable proved

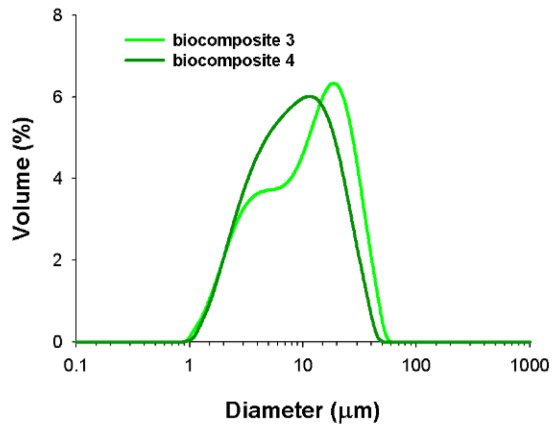

Figure 4. Particle size distributions of two selected biocomposites (Mastersizer 2000).

to be composite 6, obtained by method III, which was generally stable over the whole pH range considered, its zeta potential varying from -25 to -49 mV.

3.2.3. FT-IR spectroscopy

FT-IR analysis was performed on an Alpha spectrometer (Bruker) equipped with an attenuated total reflection (ATR) attachment, to verify the production of composites and to identify the characteristic groups present in the structure of silica, lignin and SiO_2/lignin biocomposites (Fig. 7).

The spectrum recorded for pure silica prior to its modification with silane revealed the presence of physically bound water, as the broad band in the range 3600–3200 cm^{-1} attributed to the stretching vibrations of O–H groups. An additional confirmation of this fact was the band at 1642 cm^{-1}, attributed to the bending vibrations of the same group. The other important bands were those at 1120 cm^{-1} and 817 cm^{-1}, assigned to the stretching vibrations of Si–O–Si, that at 960 cm^{-1} attributed to the stretching vibrations of Si–OH, and that at 466 cm^{-1} attributed to the stretching vibrations of Si–O. The disturbances at 2200–1900 cm^{-1} in all of the analysed spectra were assigned to the presence of a diamond in the ATR attachment. The IR spectrum of lignin confirmed the presence of functional groups specific to that substance, including hydroxyl groups –

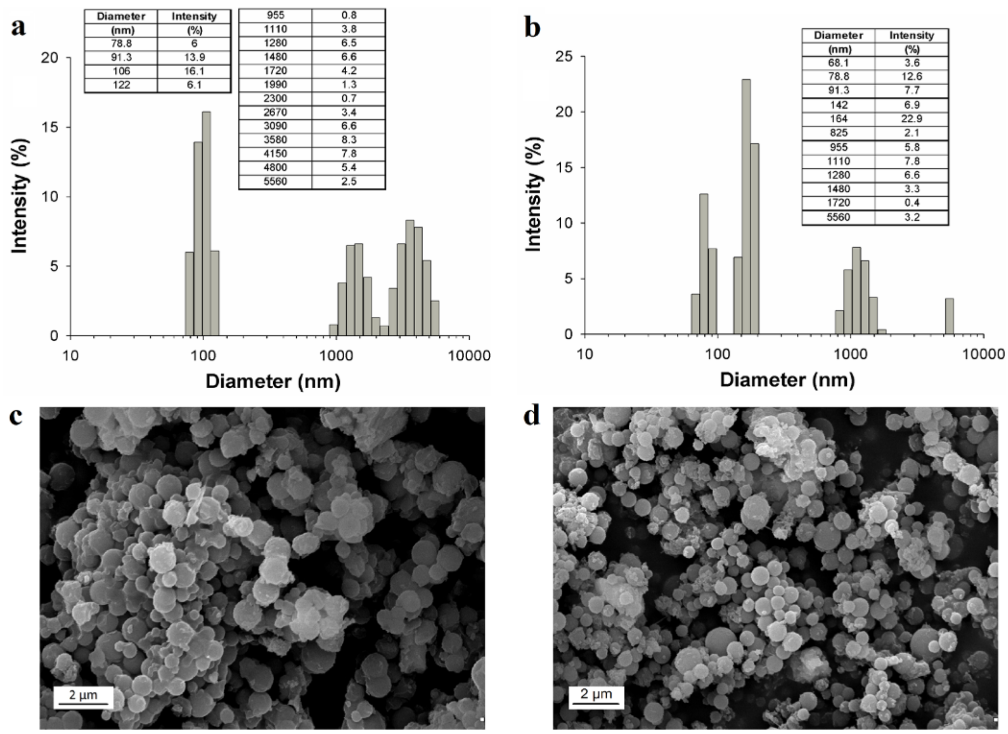

Figure 5. Particle size distributions measured on a Zetasizer Nano ZS and SEM images of (a, c) biocomposite 3 and (b, d) biocomposite 4.

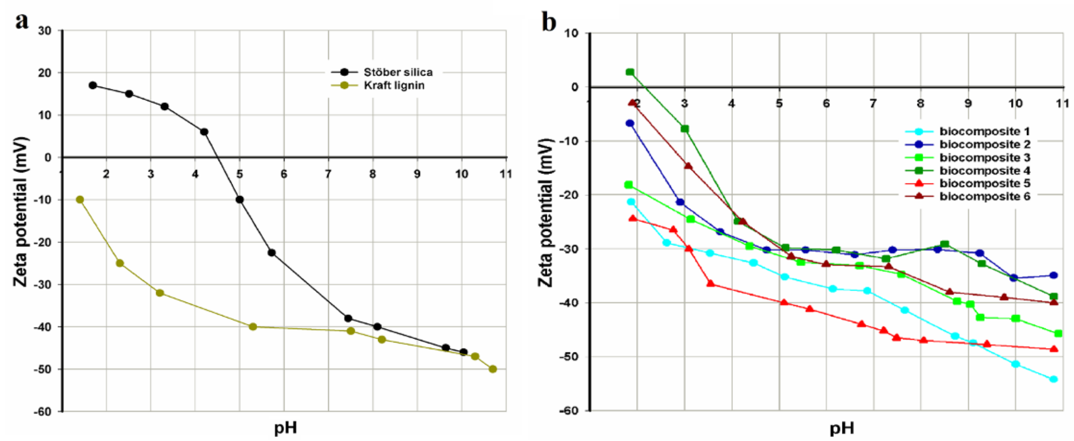

Figure 6. Zeta potential *versus* pH of (a) silica and Kraft lignin, and (b) silica/lignin biocomposites.

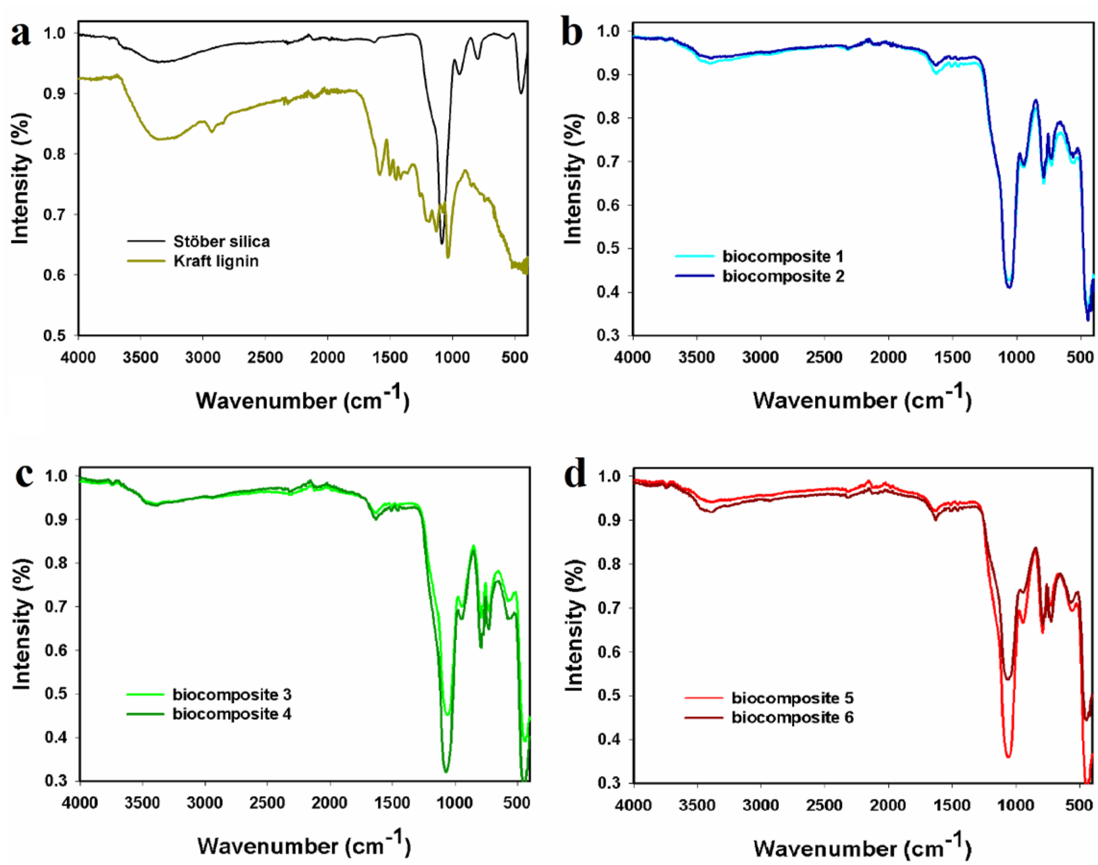

Figure 7. FT-IR spectra of (a) precursors of biocomposites, (b) SiO_2/lignin biocomposites obtained by method I, (c) method II, and (d) method III.

a band attributed to the stretching vibrations of these appeared at 3600–3200 cm^{-1} – and ether groups – a band attributed to the stretching vibrations of these appears at 1045 cm^{-1}. The detailed analysis of the spectrum of lignin was consistent with the data published in [38,39]. The FT-IR spectra of SiO_2/lignin biocomposites (Figs. 7b-7d) verified the successful and controlled production of composites by the three proposed methods. The bands characterizing the precursors overlapped the corresponding bands in the spectra of the final products. A detailed analysis of silica/lignin biocomposites based on X-ray photoelectron spectroscopy (XPS), proving the effective bonding of the precursors, is presetned in [21]. The bands with the highest intensity were recorded for the biocomposite obtained by method II at the higher temperature of the medium.

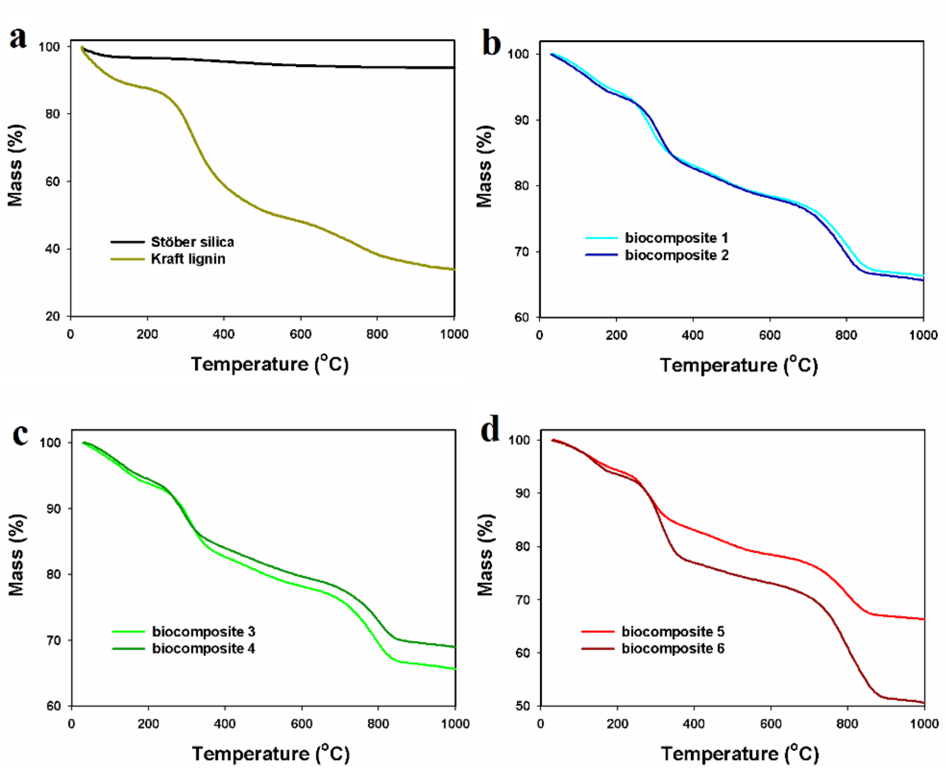

Figure 8. Thermal analysis of (a) silica and Kraft lignin, (b) silica/lignin biocomposites obtained by method I, (c) method II, and (d) method III.

3.2.4. Thermal analysis

The samples of silica, lignin and their biocomposites were subjected to thermogravimetric analysis to determine their thermal stability, which is of profound importance when considering the application of these materials as advanced polymer fillers. The results are illustrated in Fig. 8.

The TG curve recorded for SiO_2 (Fig. 8a) indicates a small mass loss of about 5%, confirming the high thermal stability of the silica. The TG curve recorded for lignin indicates a significant mass loss of about 65% relative to the initial mass of the sample [40-42]. The first small mass loss of about 10% taking place up to 200°C is mainly a result of the removal of water bonded on the lignin surface. The second, larger mass loss of about 35% taking place in the range 200–600°C is related to the complex thermal decomposition of lignin, involving the formation of new bonds as a consequence of crosslinking reactions. The third mass loss of about 15% observed in the range 600–1000°C is interpreted as a consequence of the partial elimination of hydrogen and other carbon fragments caused by fragmentation of the molecules in uncontrolled and undetermined reactions.

The silica/lignin biocomposites obtained by all three proposed methods are characterised by relatively high thermal stability. The only exception is composite 6 (Fig. 8d), which shows considerably poorer thermal properties and decreases in mass by 50% with respect to the initial mass. This result may indicate that method III is less effective in silica/lignin biocomposite formation than the other two methods (Figs. 8b, 8c). This assumption is supported by the weak intensity of bands in the FT-IR spectrum of biocomposite 6 (Fig. 7d). In general, the results characterizing the silica/lignin composites indicate that they can be successfully used as biodegradable and relatively cheap polymer fillers.

4. Conclusions

The experimental design has shown to be a useful tool in the studies of silica/lignin biocomposite formation. It considerably reduced the number of samples that were needed to be synthesized in order to obtain comprehensive information about the composites. Most importantly it helped to establish the influence of particular input data on the properties of the samples. The effectiveness of the proposed methods for the synthesis of silica/lignin biocomposites has been confirmed by the FT-IR analysis and the electrokinetic investigations. The best dispersive–morphological properties were demonstrated by the samples obtained by method II, which is described in detail in this paper. Results of electrokinetic measurements allows for confirmation of effectiveness of silica/lignin products preparation. High electrokinetic stability of the obtained biocomposites expands their application potential to medicine and pharmacy. Results from thermal analysis obtained at this stage point out that biocomposites, created based on silica and lignin, will be successfully applied as a novel polymers filler. Further areas of application of these biocomposites will be sought, including in electrochemistry as advanced electrode materials. Studies will be continued in the two above-mentioned areas.

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