

## Conductive and electroactive composite paper reinforced by coating of polyaniline on lignocelluloses fibers

Aamir Razaq,<sup>1</sup> M. H. Asif,<sup>1</sup> Riffat Kalsoom,<sup>1</sup> Ather Farooq Khan,<sup>2</sup> M. S. Awan,<sup>3</sup> S. Ishrat,<sup>1</sup> Shahid M. Ramay<sup>4</sup>

<sup>1</sup>Department of Physics, COMSATS Institute of Information Technology, Lahore 54000, Pakistan

<sup>2</sup>Interdisciplinary Research Centre in Biomedical Materials, COMSATS Institute of Information Technology, Lahore 54000, Pakistan

<sup>3</sup>SIT, H-11/4, Islamabad, Pakistan

<sup>4</sup>College of Science, Physics and Astronomy Department, King Saud University, P.O. Box 800, 11421 Riyadh, Saudi Arabia

Correspondence to: A. Razaq (E-mail: aamirrazaq@ciitlahore.edu.pk)

**ABSTRACT:** Direct use of lignocelluloses fibers as substrate for fabrication of conductive, electroactive, biodegradable, and low-cost electrode materials are in demand for high-tech applications of ion-exchange and energy storage devices. This article presents the preparation and characterizations of conductive and electroactive lignocelluloses-polyaniline (cellulose/PANI) composite paper. Lignocelluloses fibers were directly collected from the stem of self-growing plant, *Typha Angustifolia*, and subsequently coated with the conductive and electroactive layer of PANI through chemical synthesis. Individual PANI-coated lignocelluloses fibers were converted into sheet and further characterized with Scanning Electron Microscopy, Fourier Transform Infrared, Thermogravimetric Analysis, electronic conductivity, and Cyclic Voltammetry. Cellulose/PANI composite paper revealed superior thermal characteristics and used as a working electrode in three different electrolytes for ion-exchange properties. Conductive composite paper (CCP) showed the charge storage capacity of  $\sim 52$  C/g at scan rate of 5 mV/s in 2M HCl solution. © 2015 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 2015, 132, 42293.

**KEYWORDS:** coatings; composites; conducting polymers; electrochemistry; fibers

Received 15 January 2015; accepted 31 March 2015

DOI: 10.1002/app.42293

### INTRODUCTION

In recent years conductive composite papers (CCPs) have received significant attention for high-tech applications because of flexibility, light-weight, and environment friendly characteristics. CCPs are prepared mainly by two main approaches: (i) by coating of metallic nanorods,<sup>1,2</sup> (ii) by coating of conducting polymer.<sup>3–7</sup> Coatings of natural cellulose fibrils with conducting polymers are successively investigated for versatile applications in energy storage devices because of facile synthesis and the entire organic constituents.<sup>8–10</sup> Conducting polymers-based CCPs are also attractive because of environmentally friendly and relatively light-weight in comparison of other metallic based CCPs. In last few decades, conducting and electroactive polymers family received much attention because of display of their metal-like electric conductivity and reversible switching between redox states.

Polyaniline (PANI) is a well-known conducting polymer with attractive properties i.e. good environmental stability, high electrical conductivity, and facile synthesizes.<sup>11</sup> PANI has been

extensively investigated in different applications such as sensors, biomedical field, electronics, electrochromic devices, energy storage devices, and printed circuit boards.<sup>12–18</sup> However, applications of PANI for bulk-level are restricted because of poor mechanical properties. Flexible polymers like cellulose, rubber, plastic, and textile fibers are excessively investigated as substrates for mechanically reinforce matrix of electrically conductive polymers.<sup>19–32</sup>

Lignocelluloses is one of the most abundant, renewable, and eco-friendly material on earth. Current environmental issues show a pressing need for innovative, sustainable, and recyclable materials in high-tech applications. Self-growing and pollutant (because of excessive growth) plants are highly feasible for extraction of natural fibers as substrate materials for further coating of conducting polymers and metallic layers. *Typha Angustifolia*, narrow-leaved cattail, are found in wet or saturated soils and aquatic sediments in marshes, wet meadows, lake-shores, pond margins, seacoast estuaries, roadside ditches, bogs, and fens.<sup>33</sup> The stem of *T. Angustifolia* plant encloses the



**Figure 1.** Collection of lignocellulose fibers from stem of *T. Angustifolia* plant. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

lignocellulose fibrils like jute and used in several applications e.g. bio fuels,<sup>34</sup> to examine water quality,<sup>35</sup> to analyze gas flow pathway,<sup>36</sup> treatment of waste water,<sup>37</sup> powder coating in furniture,<sup>38</sup> and pulp & paper industry.<sup>39–41</sup> The chemical composition of lignocellulose fibers consist of cellulose [(C<sub>6</sub>H<sub>10</sub>O<sub>5</sub>)<sub>n</sub>] fibers within matrix of aromatic lignin [(C<sub>31</sub>H<sub>34</sub>O<sub>11</sub>)<sub>n</sub>] and hemicellulose [(C<sub>6</sub>H<sub>10</sub>O<sub>5</sub>)<sub>n</sub>].<sup>42</sup>

In era of modern disposable electronic technology, development of thin, flexible, lightweight, and environmentally friendly electrode materials are highly feasible.<sup>43</sup> This study presents the preparation and characterization of metal like cellulose/PANI conductive composite paper as a working electrode material for proposed applications of ion-exchange and energy storage devices.

## MATERIAL AND METHOD

### Chemicals and Reagents

Raw lignocellulose fibers were directly collected from the stem of self-growing plant, *T. Angustifolia* as shown in Figure 1. Collected fibers were dried in air under sunlight for four days for removal of moisture. Aniline, ammonium persulfate (APS), sodium hypochlorite (NaOCl), hydrochloric acid (HCl), and sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) were supplied by Merck. Aniline was freshly distilled before use.

### Synthesis of Paper Sheets

**Preparation of Lignocellulose Sheet.** About 1.5 g of dried (as described above) raw lignocellulose fibers were dispersed in 200 mL of 20% NaOCl solution at temperature of 50°C for 4 h. The bleached fibers were filtered on Buchner funnel (90 mm-d) and pressed overnight to achieve surface smoothness of sheet. The yield of fibers was approximately 70% after bleaching process in comparison of dry weights.

**Preparation of Cellulose/PANI Composite Sheet.** About 0.5 g of bleached lignocellulose fibers (as described above) were dispersed in 50 mL of distilled water for 30 min. APS (1.3 g) and

aniline (0.5 mL) were dissolved in 50 and 68 mL of 1M HCl solutions, respectively.<sup>44</sup> Lignocellulose fiber and Aniline solutions were mixed together and stirred for 10 min, subsequently APS solution was added drop wise. The reaction was allowed to continue for 20 h in water ice bath for oxidative polymerization. The homogenous solution of PANI-coated lignocellulose fibers were filtered on Buchner funnel and converted into sheet by pressing overnight in air. The obtained black paper sheet was flexible which can be cut with help of scissors.

### Characterization

**Scanning Electron Microscopy.** Structural morphology and microscopic information were obtained by using FE-Scanning Electron Microscopy (SEM) (Quanta FEG-480) having E-SEM (environmental SEM) mode. The samples were mounted on aluminum stubs using double-sided adhesive black carbon tape.

**Fourier Transform Infrared (FTIR) Spectroscopy.** The structure and intermolecular interactions between components of the naked and coated paper sheets were investigated by Fourier transform infrared (FTIR) spectroscopy. FTIR spectra of samples were recorded with Thermo Scientific Nicolet 8700 FTIR spectrometer at 8 cm<sup>-1</sup> resolution in averaging of 256 scans. The spectra were collected over 4000–400 cm<sup>-1</sup> range.

**Thermogravimetric Analysis (TGA).** Simultaneous thermogravimetric (TG) and differential scanning microscopy (DSC) analyses of all sheets were carried out from ambient temperature to 800°C at a heating rate of 10°C/min under Ar dynamic flow by using Model Netsch 409C thermogravimetric analyzer.

**Electronic Conductivity.** The resistance of cellulose/PANI samples was measured at room temperature using a two probe method. The voltage *U*, which was applied from 1 to 10 V, and resulting dc current *I* was measured by. The electronic conductivity  $\sigma$  was then calculated as

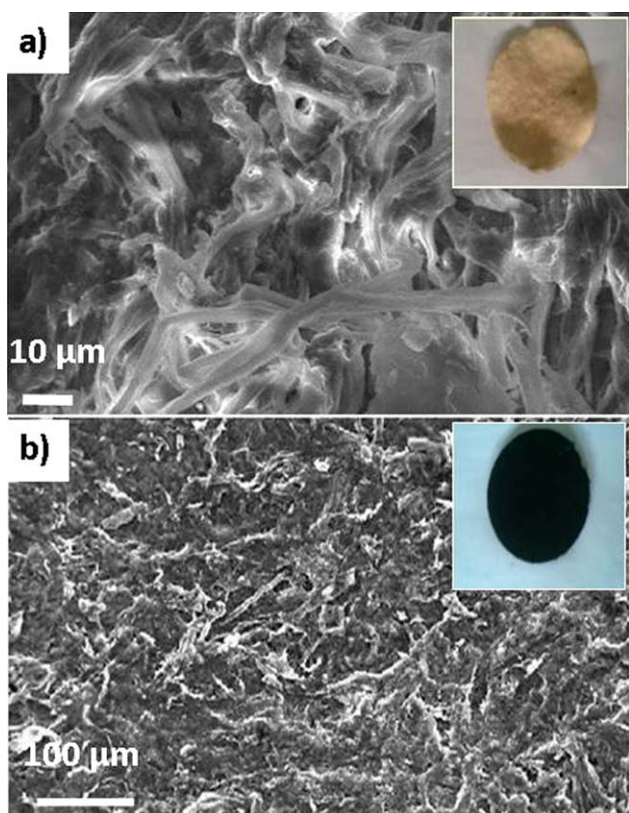
$$\sigma = (\Delta I / \Delta U) (L / wt)$$

where  $\Delta I / \Delta U$  denotes the conductance of the sample obtained from slope of the current versus voltage curve, *L* is the length, *w* is the width, and *t* is the thickness of the sample.

**Cyclic Voltammetry.** Cyclic voltammetric (CV) measurements were performed in a standard three-electrode electrochemical cell utilizing a Potentiostat/Galvanostat with GPES interface (ECO Chemie, The Netherlands). The cellulose/PANI sample was employed as a working electrode, Platinum (Pt) wire as a counter electrode and Ag/AgCl as a reference electrode. CV measurements were recorded in potential window of 0.2 to 0.7 V at scan rate of 5 mV/s in 2.0M solutions of sodium chloride (NaCl), HCl, and H<sub>2</sub>SO<sub>4</sub>, respectively. Fresh samples were used for each measurement. The weights of the samples were in the range of 3–8 mg and results were normalized with respect to mass.

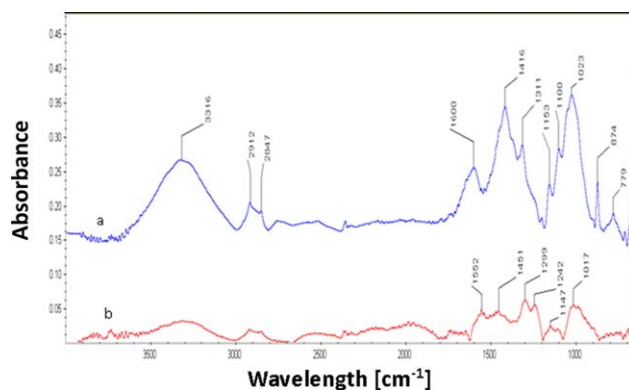
## RESULT AND DISCUSSION

The morphology of bleached lignocellulose fibers and coated cellulose/PANI sheet were investigated by SEM as shown in Figure 2. Figure 2(a) shows the dense interlocking fibril structure of uncovered sheet whereas embedded image reveals the



**Figure 2.** Scanning Electron Microscopy (SEM) images of (a) lignocelluloses fibers sheet and (b) cellulose/PANI sheet. Embedded images show the macroscopic view of lignocelluloses fibrils and cellulose/PANI sheets in Figures (a) and (b), respectively. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

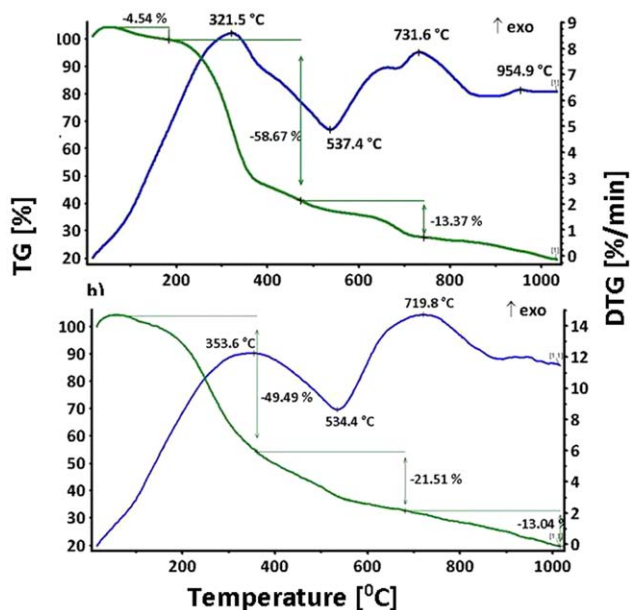
macroscopic view of compact structure of lignocelluloses fibrils sheet. Figure 2(b) displays the granular agglomerates morphology of cellulose/PANI sheet, which is subjected to conventional synthesis via oxidative polymerization.<sup>45</sup> The embedded image in Figure 2(b) depicts the appearance of cellulose/PANI composite as a black paper sheet and viable integrity of lignocelluloses fibers after coating of PANI. FTIR spectra of naked lignocelluloses and coated cellulose/PANI sheets are shown in Figure 3. Figure 3(a) shows the peak at  $3316\text{ cm}^{-1}$  (derived from hydroxyl groups stretching), peak at  $2912\text{ cm}^{-1}$  (C-H stretching of  $\text{CH}_2$ ),  $1416\text{ cm}^{-1}$  ( $\text{CH}_2$  symmetric bending), and  $1311\text{ cm}^{-1}$  (C-H bending) can be assigned for stretching and bending modes of hydrocarbons at the backbone of lignocellulose. Other peaks at 1153, 1023, and  $874\text{ cm}^{-1}$  are resulted because of asymmetric bridge C-O stretching, C-O-C stretching and vibration of anomeric C1, respectively.<sup>46</sup> Figure 3(b) represents spectrum of cellulose/PANI sheet, which predominantly reveals characteristics of PANI because of presence of peaks at 1552 and  $1451\text{ cm}^{-1}$  are characteristics peaks of C=C stretching vibration in quinone ring and benzene ring, respectively. The presence of C-N stretching peak at  $1299\text{ cm}^{-1}$  further indicates the presence of PANI. Lower intensity of hydroxyl group peak in cellulose/PANI composite at  $3316\text{ cm}^{-1}$  is because of disappearance of intermolecular hydrogen bonding in lignocelluloses fibers, in accordance with previous work from Hu *et al.* From



**Figure 3.** FTIR spectra of (a) lignocelluloses fibers sheet and (b) cellulose/PANI sheet. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

the discussion of FTIR spectrum it can be concluded that hydroxyl groups are successfully interacting between PANI and lignocelluloses fibers.<sup>47</sup>

Thermogravimetric analysis (TGA) was performed for lignocelluloses fibers sheet and cellulose/PANI sheet in order to investigate the effect of PANI coating on thermal degradation of lignocelluloses fibers as shown in Figure 4(a,b). In Figure 4(a), TGA curve of lignocelluloses fibers shows a first mass loss of 4.54% at  $100^\circ\text{C}$  which is because of vaporization of moisture and some volatile materials contained in the sample. The second mass loss was seen in the region  $210\text{--}450^\circ\text{C}$ , where thermal degradation of cellulose took place drastically. It can be seen in Figure 4(b) that thermal degradation of cellulose/PANI composite proceeded from  $180^\circ\text{C}$  in contrast with the behavior of lignocellulose, which showed thermal stability. A major weight loss of cellulose/PANI composite is recorded in the region  $210\text{--}$



**Figure 4.** Thermo gravimetric (TG) analysis of (a) lignocelluloses fibers sheet and (b) cellulose/PANI sheet. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]



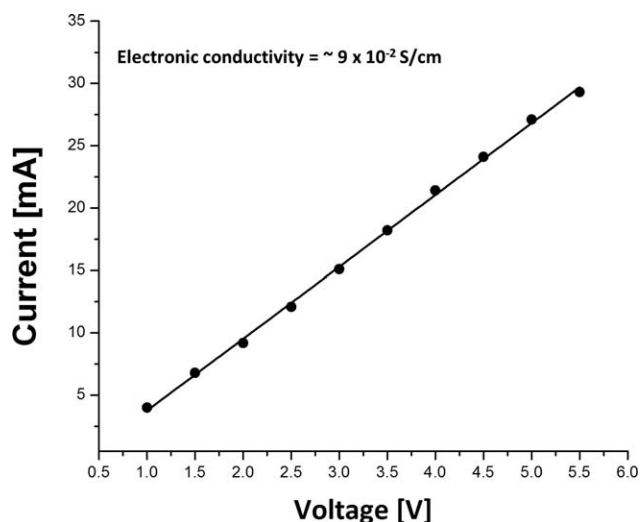


Figure 5.  $I$ - $V$  curve of cellulose/PANI composite paper sheet.

380°C because of the burning of lignocelluloses part in composite. A smaller weight loss is observed at high temperature range 550–950°C, which is because of thermal oxidative degradation of PANI. However, the thermal instability of lignocellulose was compensated by coating of PANI on lignocelluloses fibers. Figure 5 shows the bulk electrical conductivity measurement of cellulose/PANI composite sheet. It can be observed that PANI/Cellulose composite obeys Ohm's law and revealed the electronic conductivity of  $\sim 9 \times 10^{-2}$  S/cm whereas simple lignocelluloses sheet was insulating. According to literature survey, electronic conductivity of PANI depends on various factors i.e. degree of doping, particle morphology, crystallinity, and inter-chain interactions.<sup>48–51</sup> The electronic conductivity of presented cellulose/PANI is found to be less than pure PANI ( $\sim 12$  S/cm) prepared by oxidative polymerization shown by Olad *et al.*<sup>52</sup> However electronic conductivity of prepared cellulose/PANI is

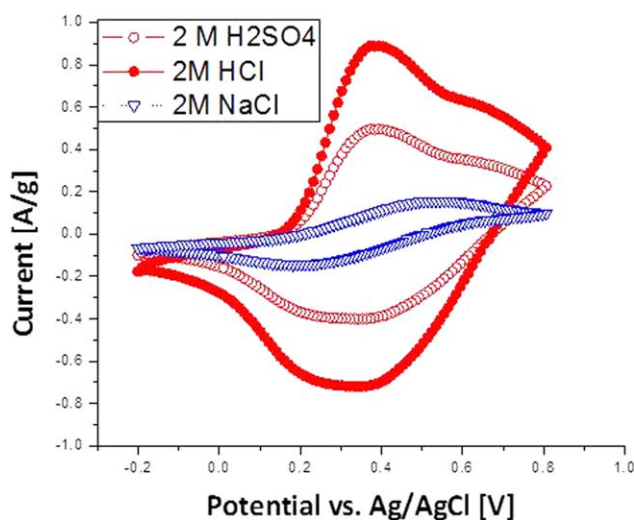


Figure 6. Cyclic voltammograms of cellulose/PANI composite paper sheet performed under three-electrode setup in (a) 2M H<sub>2</sub>SO<sub>4</sub>, (b) 2M HCl, and (c) 2M NaCl solutions. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

comparable with electronic conductivity of CCPs in literature.<sup>20,26,29,53</sup>

Figure 6 shows the redox properties of cellulose/PANI composite sheet by using as working electrode in CV measurements. Three different solutions of 2M NaCl, HCl, and H<sub>2</sub>SO<sub>4</sub> solutions and scan rate of 5 mV/s were maintained for CV measurements. To estimate the ion-exchange potential of cellulose/PANI composite, the charge capacities (shown in Figure 6) were calculated by integrating charge versus time curves from the anodic scan of respective voltammogram. Cellulose/PANI composite showed maximum charge capacity of 52.3 C/g in 2M HCl solution at scan rate of 5 mV/s. Oxidation peaks in Figure 6 are occurred around 0.4 V in 2M HCl and H<sub>2</sub>SO<sub>4</sub> solutions, whereas oxidation peaks are shifted towards more positive potential of 0.5 V in 2M NaCl solutions because of low ionic conductivity. The results clearly indicate the possibility to use the present cellulose/PANI composite as a working electrode material in ion-exchange and energy storage applications.

## CONCLUSIONS

This study presents the fabrication of cellulose/PANI composite reinforced by coating of PANI on lignocelluloses fibers. Oxidative polymerization technique was used for coating of PANI on directly collected lignocelluloses fibers from self-growing *T. Angustifolia* plant. Individual conductive microfibers acquired through synthesis were transformed into paper sheets of bulk thickness of  $\sim 1.0$  mm. The composite sheets were flexible that can cut with the help of scissor. For instance cellulose/PANI composite was found to be conductive ( $\sim 9 \times 10^{-2}$  S/cm) and electroactive (for Cl<sup>-</sup> and SO<sub>4</sub><sup>-</sup>) with maximum charge capacity of 52.3 C/g. The presented outcomes consequently give rise to new possibilities for production of low-cost, light weight, environment friendly, and conductive electrode material from self-growing plant for ion-exchange and energy storage applications.

## ACKNOWLEDGMENTS

The financial support from CRGP CIIT (16–46/CRGP/CIIT/LHR13), HEC Pakistan startup research grant (No. PD/IPFP/HRD/HEC/2013/1922) and Deanship of Scientific Research at King Saud University funding (No. RG 1435-004) are greatly acknowledged.

## REFERENCES

- Hu, L.; Choi, J. W.; Yang, Y.; Jeong, S.; Mantia, L. F.; Cui, L. F.; Cui, Y. *PNAS* **2009**, *106*, 21490.
- Hu, L.; Cui, Y. *Energy Environ. Sci.* **2012**, *5*, 6423.
- Yuan, L. Y.; Yao, B.; Hu, B.; Huo, K. F.; Chen, W.; Zhou, J. *Energy Environ. Sci.* **2013**, *6*, 470.
- Mihiranyan, A.; Nyholm, L.; Bennett, A. E. G.; Stromme, M. *J. Phys. Chem.* **2008**, *112*, 12249.
- Johnston, J. H.; Kelly, F. M.; Moraes, J.; Borrmann, T.; Flynn, D. *Curr. Appl. Phys.* **2006**, *6*, 587.
- Strømme, M.; Frenning, G.; Razaq, A.; Gelin, K.; Nyholm, L.; Mihiranyan, A. *J. Phys. Chem. B* **2009**, *14*, 4582.

7. Nyström, G.; Mihranyan, A.; Razaq, A.; Lindström, T.; Nyholm, L.; Strømme, M. *J. Phys. Chem. B* **2010**, *114*, 4178.
8. Nyholm, L.; Nyström, G.; Mihranyan, G.; Strømme, M. *Adv. Mater.* **2011**, *23*, 3751.
9. Snook, G. A.; Kao, P.; Best, A. S. *J. Power Sources* **2011**, *196*, 1.
10. Ramya, R.; Sivasubramanian, R.; Sangaranarayanan, M. V. *Electrochim. Acta* **2013**, *101*, 109.
11. Qaiser, A. A.; Hyland, M. M.; Patterson, D. A. *J. Phys. Chem. B* **2009**, *113*, 14986.
12. Katz, H. E.; Searson, P. C.; Poehler, T. O. *J. Mater. Res.* **2010**, *25*, 1561.
13. Guimard, N. K.; Gomez, N.; Schmidt, C. E. *Prog. Polym. Sci.* **2007**, *32*, 876.
14. Bai, H.; Chen, Q.; Li, C.; Lu, C. H.; Shi, G. Q. *Polymer* **2007**, *48*, 4015.
15. Hong, S. F.; Hwang, S. C.; Chen, L. C. *Electrochem. Acta* **2008**, *53*, 6215.
16. Amrithesh, M.; Aravind, S.; Jayalekshmi, S.; Jayasree, R. S. *J. Alloy. Compd.* **2008**, *449*, 176.
17. Prabhakar, N.; Arora, K.; Singh, H.; Malhotra, B. D. *J. Phys. Chem. B* **2008**, *112*, 4808.
18. Lange, U.; Roznyatouskaya, N. V.; Mirsky, V. M. *Anal. Chim. Acta* **2008**, *614*, 1.
19. Mo, Z. I.; Zhao, Z. I.; Chen, H.; Niu, G. P.; Shi, H. F. *Carbohydr. Polym.* **2009**, *75*, 660.
20. Hu, W.; Chen, S.; Yang, Z.; Liu, L.; Wang, W. *J. Phys. Chem. B* **2011**, *115*, 8453.
21. Cerqueira, D. A.; Valente, A. J. M.; Filho, G. R.; Burrows, H. D. *Carbohydr. Polym.* **2009**, *78*, 402.
22. Chandran, A. S.; Narayanankutty, S. K. *Eur. Polym. J.* **2008**, *44*, 2418.
23. Lakshmi, K.; John, H.; Mathew, K. T.; Joseph, R.; George, K. E. *Acta Mater.* **2009**, *57*, 371.
24. Jeevananda, T.; Siddaramaiah, *Eur. Polym. J.* **2003**, *39*, 569.
25. Razaq, A.; Mihranyan, A.; Welch, K.; Nyholm, L.; Strømme, M. *J. Phys. Chem. B* **2009**, *113*, 426.
26. Stejskal, J.; Trchová, M.; Kovářová, J.; Prokeš, J.; Omastová, M. *Chem. Pap.* **2008**, *62*, 181.
27. Stejskal, J.; Trchová, M.; Sapurina, I. *J. Appl. Polym. Sci.* **2005**, *98*, 2347.
28. Stejskal, J.; Trchová, M.; Brodinová, J.; Sapurina, I. *J. Appl. Polym. Sci.* **2006**, *103*, 24.
29. Guilherme, M. O.; Barra, Débora, P.; Schmitz, Sílvia, D. A. S.; Ramôa, Silveira, A.; Araujo, T. M.; Pegoretti, A. *Polym. Test.* **2014**, *38*, 18.
30. Araujo, J. R.; Adamo, C. B.; Marco, A. D. P. *Chem. Eng. J.* **2011**, *74*, 425.
31. Razak, S. I. A.; Rahman, W. A. W. A.; Hashim, S.; Yahya, M. Y. *Compos. Interface* **2012**, *19*, 365.
32. Souza, F. G.; Oliveira, G. E.; Rodrigues, C. H. M.; Soares, B. G.; Nele, M.; Pinto, J. C. *Macromol. Mater. Eng.* **2009**, *294*, 484.
33. Grace, J. B.; Harrison, J. S. *Can. J. Anal. Sci.* **1986**, *66*, 361.
34. Gomber, C.; Parihar, S. *Int. J. Environ. Eng. Manag.* **2013**, *4*, 25.
35. Jinadasa, K. B. S. N.; Tanaka, N.; Sasikala, S.; Werellagama, D. R. I. B.; Mowjood, M. I. M.; Ng, W. J. *J. Environ. Sci. Health* **2008**, *43*, 664.
36. Tornberg, T.; Bendix, M.; Brix, H. *Aquat. Bot.* **1994**, *49*, 91.
37. Kantawanichkul, S.; Kladprasert, S.; Brix, H. *Ecol. Eng.* **2009**, *35*, 238.
38. Wuzella, G.; Mahendran, A. R.; Bätge, T.; Jury, S.; Kandelbauer, K. *Ind. Crop. Prod.* **2011**, *33*, 683.
39. Bajpai, P. *Biotechnol. Prog.* **1999**, *15*, 147.
40. John, M. J.; Thomas, S. *Carbohydr. Polym.* **2008**, *71*, 343.
41. Zheng, Z. G.; McDonald, J.; Killan, R.; Su, Y.; Shutava, T.; Grozdits, G.; Lvov, Y. M. *J. Nanosci. Nanotechnol.* **2006**, *6*, 624.
42. Fengel, D.; Wegener, G. *J. Polym. Sci.* **1985**, *23*, 601.
43. Nishide, H.; Oyaizu, K. *Surface* **2008**, *319*, 737.
44. Zhang, D.; Zhang, L.; Wang, B.; Piao, G. *J. Mater.* **2013**, *2013*, 1.
45. Huang, J. *Pure Appl. Chem.* **2006**, *78*, 15.
46. Pavlovic, M. M.; Cosvic, V.; Pavlovic, M. G.; Talijan, N.; Bojanic, V. *Int. J. Electrochem. Sci.* **2011**, *6*, 3812.
47. Zhang, L. X.; Zhang, L. J.; Wan, M. X.; Wei, Y. *Synt. Met.* **2006**, *156*, 454.
48. Zhang, Z.; Wei, Z.; Wan, M. *Macromolecules* **2002**, *35*, 5937.
49. Abdiryim, T.; Jamal, R.; Nurulla, I. *J. Appl. Polym. Sci.* **2007**, *105*, 576.
50. Stafström, S.; Brédas, J. L.; Epstein, A. J.; Woo, H. S.; Tanner, D. B.; Huang, W. S.; MacDiarmid, A. *J. Phys. Rev. Lett.* **1987**, *59*, 1464.
51. Lee, B. H.; Kim, H. J.; Yang, H. S. *Curr. Appl. Phys.* **2012**, *12*, 75.
52. Olad, A.; Khatamian, M.; Naseri, B. *Int. J. Nanosci. Nanotechnol.* **2010**, *6*, 43.
53. Camacho, D. H.; Gerongay, S. R. C.; Macalinao, J. P. C. *Cellulose Chem. Technol.* **2013**, *47*, 125.