1
Advances in Polymer Composites: Biocomposites – State of the Art, New Challenges, and Opportunities

Koichi Goda, Meyyarappallil Sadasivan Sreekala, Sant Kumar Malhotra, Kuruvilla Joseph, and Sabu Thomas

1.1
Introduction

Environmental compatibility of polymer composites has become an important characteristic as the need to reduce environmental hazards is increasing worldwide. Many incidents taking place around the world are enough to bring us around to this point of view. A catastrophic earthquake and *tsunami* devastated the Pacific coast of north-eastern Japan on 11 March 2011. The earthquake, which was the most powerful earthquake ever measured in Japan, was of magnitude 9.0 on the Richter scale. About 19 000 were dead and missing. Three prefectures in the Tohoku (north-eastern) region of Japan, Miyagi, Iwate, and Fukushima, were most severely damaged. Reconstruction is yet to take place in many of the affected cities and towns. The area around the Fukushima Daiichi Nuclear Power Plant was evacuated owing to radioactive contamination. It is said that complete restoration will take more than 30 years, because the influence of the Chernobyl nuclear power plant disaster, which happened more than 25 years ago, continues to be felt. In Fukushima prefecture, many residents are still forced to lead lives as long-term refugees, and the residents in certain areas outside the refuge zone continue to live under threat of radiation that is much higher than is normal. The damage caused by radioactivity has also been considerable: it has already affected the soil of schoolyards, tapwater, grass, agricultural products, marine products, and so on, in large areas within the Fukushima prefecture. It is not clear how much of this damage is due to sea pollution and how long its effects will last in the future.

Against such a background, a planned conversion to renewable natural power sources as recommended by the energy policy, depending on nuclear power generation, attracts attention. For instance, it has been decided to abolish nuclear power generation systems in Germany; they propose to convert from 16% of total energy generation from the natural power sources at present to 35% by 2020 and to 80% by 2050 [1]. In the report ''The Green New Deal'' published in 2009 [2], promotion of use and development of alternative and renewable energy, improvement in energy efficiency, greenhouse gas reduction, and so on, have also been proposed. Today,

Polymer Composite: Volume 3, First Edition.
Edited by Sabu Thomas, Kuruvilla Joseph, S. K. Malhotra, Koichi Goda and M. S. Sreekala.

technologies for various natural power sources, such as solar power, hydraulic power, woody biomass, and wind force power generation, are already in practical use. The authors believe that many people in the world desire realization of a sustainable society that uses such renewable energy power generation technologies.

To realize a sustainable society, various supplies around our life also need to be made from renewable materials. Biomass-derived materials are one of the most sustainable materials, which can also be used as industrial materials. On the other hand, most engineering plastic products are petroleum-derived products. As is well-known, the use of fossil resources causes difficulties in recycling and induces the problem of waste plastic and petroleum products, of which the incineration also causes an increase in carbon dioxide linking to global warming. In addition, fossil resources are an exhaustible resource. To maintain a sustainable society, we are of the opinion that biomass resources may be suitably exploited socially/ecologically as much as possible, by their replacing fossil resources. Since the arrival of such a society will result in a carbon-neutral system, this would also greatly contribute to global environmental protection. It is said that biodegradable plastics, for example, polylactic acid (*PLA*) and polyhydroxyalkanoic acid (*PHA*), are among the leading biomass-derived materials, which are finally decomposed by microorganisms into water and carbon dioxide. Therefore, there are only a few impacts on natural environment compared with those of conventional petroleum-derived plastics. Such biomass-derived materials are expected to be more widely applicable for the commodities used by us on a daily basis, for industrial products, and so on.

The main drawbacks of biodegradable resins are low strength and stiffness, and therefore, it is not appropriate to apply resins directly for structural components. Plastics are often reinforced with inorganic fibers such as glass or carbon, as described in Volume I of this series. Carbon fiber-reinforced plastic matrix composites (*CFRP*), in particular, have been recently used for primary structural components in airplanes and automobiles as well as sport goods and construction materials, because of their excellent mechanical properties. Biodegradable resin may also be reinforced with such fibers, similarly to the conventional petroleum-derived plastics. However, let us recall here how we should construct a sustainable society. If the final products do not really require high strength and durability, do we need to use strong artificial fiber-reinforced composites? Cellulosic materials, namely, plant-based natural fibers such as flax, hemp, bamboo, and wood, have low densities, are biodegradable, and inexpensive, and they have relatively high stiffness and less wear/abrasion to material partners. If such cellulosic materials are used as reinforcements of biomass-derived plastics, this material would be a quite suitable for building a sustainable society. We call such a biomass-based composite material a *biocomposite*. This idea of using natural fibers had already been adopted in the experimentally developed automotive body in 1940s by Henry Ford [3]. Fifty years later, Mercedes-Benz applied composites produced from natural fibers and polypropylene to their car interior parts in the 1990s. Although the matrix used in the cars was petroleum-derived thermoplastic resin, this business should be evaluated as an advanced measure in terms of practical and large-scale production. The use of natural fiber-reinforced composites using biomass-based biodegradable

resin began in the 2000s. Toyota Motors first applied these composites to their spare tire covers in 2003, the constituents of which were kenaf fibers and PLA resin [4]. NEC and UNITIKA collaborated to develop a biocomposite of the same system, applicable for the body of mobile phones in 2005 [5]. Today, novel biocomposites are further being developed in research institutes and industries.

Technological innovation, which replaces all petroleum-based materials by biomass-based ones would be the task imposed on scientists and engineers of the twenty-first century, because it is anticipated that fossil resources will disappear in the near future. We consider that this innovation also includes the improvement and development of biomass-based fibers and resins, of which the mechanical properties are comparable to those of artificial fibers and petroleum-based resins, respectively. In this sense, the study of biocomposites must not end with just their being ''environmental-friendly,'' but must be advanced in the future in the quest toward establishing a sustainable society.

1.2
Development of Biocomposite Engineering

Biocomposites (the title of Volume III), are often interpreted as either biomass-based or biomedical materials. The former have a wider meaning than the latter, because they are available for various industrial purposes. A biomass-based composite consists of biomass and/or biomass-derived substance. On the other hand, a biomedical composite is a specified material because it is limited merely to biomedical use. In this use, the constituents are not necessarily biomass-based or biodegradable, but should be biocompatible. In the present volume, as stated earlier, by biocomposites, we mean biomass-based composites.

In this volume, the application of biocomposites is premised on structural use rather than functional one. From this point of view, we need to know exactly the mechanical properties, such as tensile strength and Young's modulus, of natural fibers and wood flours, similarly to the case of artificial reinforcing materials such as carbon and glass fibers. Tensile properties of natural fibers such as cotton, flax, wool, and silk have been examined in detail in the field of textile engineering. According to the book titled ''Physical Properties of Textile Fibers'' [6] published in 1962, the strength of fibers is mainly evaluated as maximum load divided by fiber specimen weight, denoted by the tenacity (g tex^{-1}) or the specific strength (g $denier^{-1}$). In textile engineering, a continuous filament called a *spun yarn* is a basic configuration [7]. Spun yarns are produced by spinning short fibers using a spinning machine or wheel, because most natural fibers are finite in length. Some of the spun yarns are further processed into twisted or blended yarns. To evaluate the various and complicated configurations, the concept of normalization by ''load per weight'' may be convenient to understand. The relation between the basic structure of spun yarns and their mechanical properties had already been clarified in the 1970s. The field of such study, called *yarn mechanics*, was extended to regenerated and chemical fibers, as well as natural fibers [8]. In the 1980s,

natural fibers began to attract attention as a sustainable material, in addition to textile use, which is deeply related to the solution of environmental and energy problems. India, especially, played a pioneering role regarding production and application of several materials containing jute fibers, as shown in many papers [9–13] and review articles [14, 15]. During this period, jute fiber composites using thermosetting resins had been the main targets of research; thus the idea of hybrid composites with glass fibers was proposed [9, 16, 17]. Application of plant-based natural fibers into cement concrete had also been reported by several Indian institutes [18, 19].

Meanwhile, the project known as *Poverty and Environment Amazonia* (*POEMA*) in Brazil, established by Daimler-Chrysler, also started in 1981 [20]. This organization contracted with the residents of the Amazon valley, and encouraged them to apply natural resources such as coconut fibers to car interior parts. In the 1980s, however, the natural fiber composites were not biodegradable, because the resins applied were petroleum based. In the 1990s, a new type of fibrous composites was reported, in which the reinforcement and matrix of the composites were both biodegradable; the constituents were respectively natural fibers and polyvinyl alcohol (*PVA*) [21]. Netravali *et al*. [22] also developed in 1998 the composite system of natural fibers and biomass-derived resin, and these were termed as *fully green composites*. Since then, green composites have been recognized as one of the representative biodegradable materials reinforced with natural fibers. Various production methods and properties of green composites have been studied, and they are applied for several industries, as mentioned above. In the studies of green composites, most of researchers treat the tensile strength of natural fibers as ''load per cross-sectional area.'' To estimate the exact strength, even the morphology of the fiber cross-section has often been investigated, because it is quite complicated and different from the circular cross-section seen in many artificial fibers [23–28]. In relation to such mechanics or strength estimation, several researchers have further extended it to the numerical [29–31] or stochastic [24, 32] viewpoint. Not only such academic points of view of natural fibers but also the mechanics of composites reinforced with textile yarns such as spun or twisted yarn is on the rise [33, 34] (see, also Chapter 10.1).

The study on the above-mentioned natural fiber strength is one example concerning the progress of biocomposite engineering, in which natural fibers are evaluated as a structural material. Meanwhile, studies on the improvement of interface between wood fibers/flour (*WF/F*) and polymeric resin have also been progressing, of which the material is known by the name of *wood–plastic composite*s (*WPC*s) [35] (see, also Chapter 5.2). This material is an in-between field that needs knowledge of both polymer chemistry and wood science. We also consider that WPC is in a category of biocomposites. The compatibility between WF/F and polymeric resin is quite poor, which leads to nonuniform dispersion of WF/F and low mechanical properties. The relation between wood and plastic is similar to that between oil and water – they do not mix so easily. Thermoplastic resins often used as a matrix material are hydrophobic, while WF/F is hydrophilic. These two contrary properties result in poor interfacial strength. As in the development of the

silane-coupling agent linking glass fibers to polymeric resin, studies on the effect of various chemical treatments on the interfacial strength, such as cross-liking and acetylation of cellulose, grafting, use of coupling agent, have been conducted since the 1970s. It has been reported in many papers [36–38] (see Chapter 5.2) that, for example, WPC is improved in strength and impact properties by addition of a compatibilizer such as maleic anhydride-*grafted*-polypropylene (*MAPP*). WPCs were first introduced into the decking market in the early 1990s, in which 50% wood flours and 50% low density polyethylene (LDPE) were combined. Today, the WPC industry has grown into one of the greatest in the various fields of biocomposites. Although surface treatments on inorganic filler or reinforcement have been developed in conventional composite engineering, the above polar and nonpolar interface improving technology, a common subject to natural fibers, has also been creating biocomposite engineering (see, Chapter 4). In the 2000s, such chemical treatment in the WPC production process has been extended to achieve compatibility with biodegradable resins such as PLA [39]. This research progress quite matches the idea of as-mentioned fully green composites. WPC is further progressing through a technology fibrillating WF/F into the nanoscale [40].

As in the above, hitherto unknown issues inherent in biocomposites are being solved and meanwhile the appropriate evaluation methods are also being built up. We believe that development of various researches and technologies, such as the following would lead to an unwavering future for biocomposite engineering:

- Structure–property relationships in biopolymers (Chapter 2)
- Basic and applied researches of cellulose (Chapter 3)
- Interface improvement technology (Chapter 4)
- Production technology for thermoplastic–resin matrix biocomposites (Chapter 5)
- Production technology for thermoset–resin matrix biocomposites (Chapter 6)
- Biofiber reinforcement in thermoplastics (Chapter 7)
- Biofiber reinforcement in natural rubber (Chapter 8)
- Cellulose containing effect for improvement of interfacial strength (Chapter 9)
- Yarn optimization for natural fiber composites (Chapter 10)
- Bionanocomposites (Chapter 11)
- Fully biodegradable green composites (Chapter 12)
- Applications and future scope of natural fiber composites (Chapter 13)
- Biomedical applications of polymer composites (Chapter 14)
- Environmental effects, biodegradability, and life cycle analysis of biocomposites (Chapter 15).

1.3 Classification of Biocomposites

In this section, we try to clarify where biocomposites are positioned among the whole composite materials. In the previous section, the importance of green composite studies was described, and related to the biocomposite-engineering field. The combination of natural fibers and biomass-derived biodegradable resin is

common to both biocomposites and green composites. What is the difference between biocomposites and green composites? PLA containing hydroxyapatite, a representative bioabsorbable biomedical composite, is expected to be applied widely as a bone-connecting material. Hydroxyapatite is a mineral-derived natural resource, but it is neither biodegradable nor biomass-based. Therefore, this composite cannot be denoted as a fully green composite, though it is partially biodegradable and biomass-based. Meanwhile, carbon fiber had originally been made from a biomass, as represented in a carbonized bamboo fiber filament developed by Thomas Edison. Although most of them are made from petroleum-derived acrylic fibers in the present technology, even now, some carbon fibers are made from pulp-originated rayon fibers. When we apply such a carbonized bamboo fiber to reinforcement, this may be called a *fully biomass-based composite* by combining it with biomass-derived resin even if the resin used is nonbiodegradable. Wood ceramics [41] is also a carbonized composite material composed of wood flour and phenolic resin, which is produced by sintering its precursor at high temperatures under inert atmosphere. (The precursor of ceramics is often called *green body*.) This material is expected to be applied to electromagnetism shields, tribological components, and heat-resistant and corrosion-resistant materials, because of their excellent properties. Wood ceramics is not green, but the great part of this material is biomass-based. Such carbonized biomass materials also attract attention from the viewpoint of carbon fixation technology, called *biochar*. It seems from the aforementioned that biocomposites can be defined as a biomass-based composite occupying a larger category than green composites.

On the other hand, we must not forget that ''green'' often means ''environment-friendly'' as well as biodegradable. Many unnecessary textiles and discarded composite products are often treated as industrial waste, but we understand that these are recyclable. For example, when waste uniform clothes composed of polyester and wool fibers are combined with PVA, they can be used as an agricultural material [42]. Growth of plants is promoted more effectively through this application. In this case, this could be termed *green* composite material. Discarded glass fiber-reinforced plastic (GFRP) or CFRP products are decomposed thermally and/or chemically, and can be used as a recycle glass or carbon fiber [43, 44]. If such fibers are used again as reinforcement of composites, then we could also call these *green* composites. Green chemistry means chemical technology aiming at lower environmental impact, in which one of the purposes is to improve life cycle efficiency for petroleum-based plastics. Another purpose is furthermore directed to refining of bioethanol from biomass resources and even polyolefin materials production using this ethanol. Composites made from such improved petroleum-based plastics or biopolyolefins may also be called *green* composites, despite the fact that they are not biodegradable. Thus, we should know that green composites are not necessarily a subset of biocomposites, but consist of the intersection of biocomposites and a disjoint part.

From such a point of view, we have classified biocomposites and green composites, as shown in Figure 1.1. This classification is based on the various matrix and

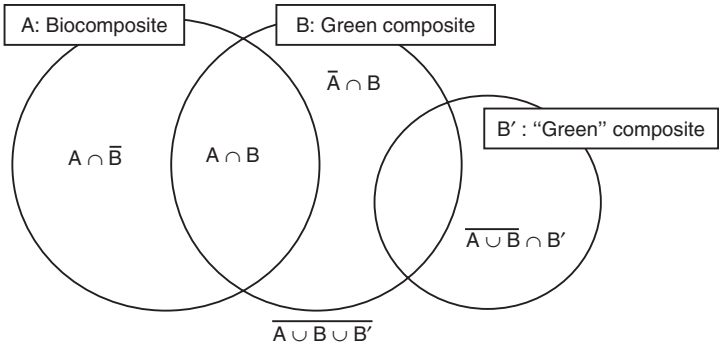

Figure 1.1 Classification of biocomposites and green composites.

Table 1.1 Various matrix materials and reinforcements (filler included).

(a) Matrix

	Biomass derived	**Petroleum derived**
Biodegradability	(Group BM1) PLA, PHA	(Group PM1) PCL, PVA
Nonbiodegradability	(Group BM2) cellulose ester, bioethylene, biopolypropylene	(Group PM2) ethylene, polypropylene

(b) Reinforcement or filler

	Biomass derived (including inorganic)	**Petroleum derived or inorganic**
Biodegradability	(Group BF1) natural fibers, wood flour, spider silk	(Group PF1) fibers made from PM1
Nonbiodegradability	(Group BF2) rayon-based carbon, carbonized wood flour, carbonized cellulose	(Group PF2) chemical fibers, glass, PAN-based carbon, hydroxyl-apatite, nanoclay, and crashed shell

PCL, poly(ε-caprolactone); PAN, polyacrylonitrile.

reinforcement (or filler) properties as shown in Table 1.1a,b, and the meaning of green is defined as biodegradability.

$A \cap B$ is the intersection of biocomposites and green composites. In this category, the materials of matrix and/or reinforcement consist of a biomass-based and biodegradable substances. The group combinations are given as BM1/BF1, BM1/PF1, PM1/BF1, BM1/BF2, BM1/PF2, PM1/BF2, BM2/BF1, BM2/PF1, and PM2/BF1. The first combination, i.e., BM1/BF1, leads to a fully biomass-based and biodegradable composite material.

$A \cap \overline{B}$ is the intersection of biocomposites and nongreen composites. In this category, the materials of matrix and reinforcement consist of a biomass-based and

nonbiodegradable substance. In addition, in case the material of either matrix or reinforcement satisfies this substance, its counterpart must not be biomass-based or biodegradable. The group combinations are given as BM2/BF2, BM2/PF2, and PM2/BF2.

$\overline{A} \cap B$ is the intersection of nonbiocomposites and green composites. In this category, the materials of matrix and/or reinforcement consist of a petroleum-derived (or inorganic) and biodegradable substance. However, the materials of matrix and reinforcement must not be petroleum derived (or inorganic) and nonbiodegradable. The group combinations are given as PM1/PF1, PM1/PF2, and PM2/PF1.

$\overline{A \cup B}$ is the compliment of biocomposites or green composites. In this category, the materials of matrix and reinforcement are both petroleum derived (or inorganic) and nonbiodegradable. The group combination is given as PM2/PF2. However, in case the material of reinforcement consists of recycled textiles or fibers or that of the matrix consists of recycled resin, it can be called a *green* composite. If the matrix material is an excellent life cycle resin in conformity with the concept of green chemistry, this material is also accepted as ''green'' composite even if it is petroleum derived and nondegradable. This category is presented as $\overline{A \cup B} \cap B'$.

Meanwhile, biocomposite research needs many tasks to identify. Identification and effective utilization of renewable resource materials are needed for biocomposite preparation. Finding out effective and economic processing methods is necessary for the separation of starting biomaterials into their pure forms for the production of biocomposites. Performance of the biocomposites is dependent on the inherent properties of the matrix and reinforcement and their interface characteristics. We can tailor the properties of the biocomposites by optimizing processing parameters and by employing suitable physical or chemical modifications to improve the interface. Identifying the thrust areas for the application of biocomposites and manufacture of prototypes and fabrication of useful products has an important role in biocomposite research. The biocomposites will play a major role in replacing nonbiodegradable synthetic materials in the near future.

References

1. Law for the Priority of Renewable Energies (Renewable-Energy-Law) – EEG, *http://www.gesetze-im-internet.de/bundesrecht/eeg_2009/gesamt.pdf* (accessed 1 April 2013).
2. UNEP Global Green New Deal, *http://www.unep.org/pdf/G20_policy_brief_Final.pdf* (accessed 1 April 2013).
3. Popular Mechanics Magazine (Dec. 1941), Vol. 76, No. 6.
4. Inoh, T., Industrial products of plant origin material – effective use of plant origin plastics for recycling society, *J. Jpn. Soc. Mech. Eng.*, **109**, 51–52 (2006) (in Japanese).
5. Iji M., Serizawa, S. and Inoue, K., Development of polylactic acid with kenaf and its application to electronic products, *Seikei-Kakou (J. JSPP)*, **15**, 602–604 (2003) (in Japanese).
6. Morton, W.E. and Hearle, J.W.S., *Physical Properties of Textile Fibers*, (1962) The Textile Institute & Butterworths, Manchester and London.
7. Hearle, J.W.S., Grosberg, P. and Backer, S., *Structural Mechanics of Fibers, Yarns*

and Fabrics, Vol. I (1969) John Wiley & Sons, Inc., New York.
8. Hearle, J.W.S. and Konopasek, M., On united approaches to twisted yarn mechanics, *Appl. Polym. Symp.*, **27** 253–273 (1975).
9. Shah, A. N. and Lakkad, S. C., Mechanical properties of jute-reinforced plastics, *Fibre Sci. Technol.*, **15**, 41–46 (1981).
10. Mukherjea, R.N., Pal, S.K., Sanyal, S.K., Studies on jute fiber composites with polyesteramide polyols as interfacial agent, *J. Appl. Polym. Sci.*, **28**, 3029–3040 (1983).
11. Pal, P.K., Jute reinforced plastics: a low cost composite material, *Plast. Rubber Process. Appl.*, **4**, 215–219 (1984).
12. Sanadi, A.R., Prasad, S.V., Rohatgi, P.K., Natural fibers and agro-wastes as fillers and reinforcements in polymer composites, *J. Sci. Ind. Res.*, **44**, 437–442 (1985).
13. Prashant, K., Mechanical behavior of jute fibers and their composites, *Indian J. Technol.*, **24**, 29–32 (1986).
14. Chand, N., Tiwary, R.K., Rohatgi, P.K., Bibliography resource structure properties of natural cellulosic fibres – an annotated bibliography, *Mater. Sci.*, **23**, 381–387 (1988).
15. Mohanty, A.K. and Misra, M., Studies on jute composites – a literature review, *Polym. Plast. Technol. Eng.*, **4**, 729–792 (1995).
16. Mohan, R., Kishore, A., Shridhar, M.K., and Rao, R.M.V.G.K., Compressive strength of jute-glass hybrid fibre composites, *J. Mater. Sci. Lett.*, **2**, 99–102 (1983).
17. Varma, I.K., Anantha Krishnan, S.R. and Krishnamoorthy, S., Composites of glass/modified jute fabric and unsaturated polyester resin, *Composites*, **20**, 383–388 (1989).
18. Parameswaran, V.S., Krishnamoorthy, T.S. and Balasubramanian, K., Current research and applications of fiber reinforced concrete composites in India, *Transp. Res. Rec.*, **1226**, 1–6 (1989).
19. Sethunarayanan, R., Chockalingam, S., Ramanathan, R, Natural fiber reinforced concrete, *Transp. Res. Rec.*, **1226**, 57–60 (1989).
20. Tomari, M. and Harago, Y., *Amazon no Hatake de Toreru Mercedes-Benz* (1997) Tsukiji-Shokan, Tokyo (in Japanese).
21. Herrmann, A.S., Nickel, J. and Riedel, U., Construction materials based upon biologically renewable resources, *Polym. Degrad. Stab.*, **59**, 251–261 (1998).
22. Luo, S. and Netravali, A.N., Interfacial and mechanical properties of environment-friendly 'green' composites made from pineapple fibers and poly (hydroxybutyrate-co-valerate) resin, *J. Mater. Sci.*, **34**, 3709–3719 (1999).
23. Suzuki, K., Kimpara, I., Saito, H. and Funami, K., Cross-sectional area measurement and monofilament strength test of kenaf bast fibers, *J. Soc. Mater. Sci. Jpn.*, **54**, 887–894 (2005) (in Japanese).
24. Tanabe, K., Matsuo, T., Gomes, A., Goda, K. and Ohgi, J., Strength evaluation of curaua fibers with variation in cross-sectional area, *J. Soc. Mater. Sci. Jpn.*, **57**, 454–460 (2008) (in Japanese).
25. Silva, F. A., Chawla, N., Filho, R. D. T., Tensile behavior of high performance natural (sisal) fibers, *Compos. Sci. Technol.*, **68**, 3438–3443 (2008).
26. (a) Virk, A.S., Hall, W. and Summerscales, J., Multiple data set (MDS) weak-link scaling analysis of jute fibres, *Composites Part A*, **40**, 1764–1771 (2009); (b) Virk, A. S., Hall, W. and Summerscales, J., Tensile properties of jute fibres, *Mater. Sci. Technol.*, **25**, 1289–1295 (2009).
27. Xu, X.W. and Jayaraman, K., An image-processing system for the measurement of the dimensions of natural fiber cross-section, *J. Comput. Appl. Technol.*, **34** (2), 115–121 (2009).
28. Goda, K., Current status and future prospects of biocomposites II: strength evaluation of plant-based natural fibers for green composites, *J. Soc. Mater. Sci. Jpn.*, **59** (12), 977–983 (2010) (in Japanese).
29. Gassan, J., Chate, A. and Bledzki, A.K., Calculation of elastic properties of natural fibers, *J. Mater. Sci.*, **36**, 3715–3720 (2001).
30. Baley, C., Analysis of the flax fibres tensile behaviour and analysis of the tensile

stiffness increase, *Composites Part A* **33** 939–948 (2002).
31. Xu, P. and Liu, H., Models of microfibril elastic modulus parallel to the cell axis, *Wood Sci. Technol.*, **38**, 363–374 (2004).
32. Andersons, J., Porike, E. and Sparnins, E., The effect of mechanical defects on the strength distribution of elementary flax fibres, *Compos. Sci. Technol.*, **69**, 2152–2157 (2009).
33. Shioya, M., Itoh, T., Kunugi, T. and Takaku, A., Variation of longitudinal modulus with twist for yarns composed of high modulus fibers, *Text. Res. J.*, **71** (10), 928–936 (2001).
34. Yoshida, K., Kurose, T., Nakamura, R., Noda, J., and Goda, K., Effect of yarn structure on mechanical properties of natural fiber twisted yarns and green composites reinforced with the twisted yarn, *J. Soc. Mater. Sci. Jpn.*, **61** (2), 111–118 (2012) (in Japanese).
35. Ashori, A., Wood–plastic composites as promising green-composites for automotive industries! (Review Paper) *Bioresour. Technol.*, **99**, 4661–4667 (2008).
36. Felix, J.M. and Gatenholm, P., Nature of adhesion in composites of modified cellulose fibers and polypropylene, *J. Appl. Polym. Sci.*, **42**, 609–620 (1991).
37. Kazayawoko, M., Balatinecz, J. and Matuana, L.M., Surface modification and adhesion mechanisms in woodfiber-polypropylene composites, *J. Mater. Sci.*, **34**, 6189–6199 (1999).
38. La Mantia, F.P. and Morreale, M., Green composites: a brief review, *Composites Part A*, **42**, 579–588 (2011).
39. Takatani, M. and Okamoto, T., Wood/plastic composite of high filler content, *Mol. Cryst. Liq. Cryst.*, **483**, 326–338 (2008).
40. Abe, K., Iwamoto, S. and Yano, H., Obtaining cellulose nanofibers with a uniform width of 15 nm from wood, *Biomacromolecules*, **8**, 3276–3278 (2007).
41. Okabe, T., *et al.*, *Wood Ceramics*, (1996) Uchida Rokakuho, Tokyo (in Japanese).
42. Sekkuden, M., Yamamura, T., Okazawa, T., Sano, T., Tanaka, K., Goda, K., Ogawa, K., and Okabe, T. (2012) Eco-friendly utilization of uniform cloths waste – composites with PVA and anti-fungal biomass oil. Proceedings of the 10th International Conference on Ecomaterials (ICEM-10), pp. 203–206.
43. Shima, H., Takahashi, H., and Mizuguchi, J., Recovery of glass fibers from fiber reinforced plastics, *Mater. Trans.*, **52**, 1327–1329 (2011).
44. Liu, Y., Liu, J., Jiang, Z. and Tang, T., Chemical recycling of carbon fibre reinforced epoxy resin composites in subcritical water: synergistic effect of phenol and KOH on the decomposition efficiency, *Polym. Degrad. Stab.*, **97** (3), 214–220 (2012).