Handbook of Composites from Renewable Materials
Handbook of Composites from Renewable Materials
Edited by Vijay Kumar Thakur, Manju Kumari Thakur and Michael R. Kessler

Volume 1: Structure and Chemistry

Volume 2: Design and Manufacturing
ISBN: 978-1-119-22365-8

Volume 3: Physico-Chemical and Mechanical Characterization

Volume 4: Functionalization

Volume 5: Biodegradable Materials

Volume 6: Polymeric Composites
ISBN: 978-1-119-22380-1

Volume 7: Nanocomposites: Science and Fundamentals
ISBN: 978-1-119-22381-8

Volume 8: Nanocomposites: Advanced Applications

8-volume set
To my parents and teachers who helped me become what I am today.

Vijay Kumar Thakur
## Contents

### Preface

xix

### 1 Rice Husk and its Composites: Effects of Rice Husk Loading, Size, Coupling Agents, and Surface Treatment on Composites’ Mechanical, Physical, and Functional Properties

A. Bilal, R.J.T. Lin and K. Jayaraman

1.1 Introduction 1

1.2 Natural Fiber-Reinforced Polymer Composites 3

1.3 Rice Husk and its Composites 5
   1.3.1 Polymers Used in the Manufacturing of RH Composites 7
   1.3.2 Effects of RH Loading on the Properties of RH Composites 8
   1.3.3 Effects of RH Size on the Properties of Composites 11

1.4 Effects of Coupling Agents on the Properties of RH Composites 12
   1.4.1 Effects of Surface Treatment of RH on the Properties of RH Composites 14
   1.4.2 Potential Applications of RH Composites 15

1.5 Summary 15

References 16

### 2 Biodegradable Composites Based on Thermoplastic Starch and Talc Nanoparticles

Luciana A. Castillo, Olivia V. López, M. Alejandra García, Marcelo A. Villar and Silvia E. Barbosa

2.1 Introduction 23

2.2 Thermoplastic Starch-Talc Nanocomposites 27
   2.2.1 Effects of Talc Presence on TPS Structure 28
   2.2.2 Effects of Talc Presence on TPS Thermal Properties 34
   2.2.3 Effects of Talc Presence on TPS Dimensional and Thermal Stability 36
   2.2.4 Effects of Talc Presence on TPS Optical Properties 38

2.3 Use of Talc Samples with Different Morphologies 40
   2.3.1 Talc Morphology Influence on Composite Structure 40
   2.3.2 Talc Morphology Influence on Composite Thermal Properties 44
   2.3.3 Talc Morphology Influence on Composite Final Properties 45

2.4 Packaging Bags Based on TPS–Talc Nanocomposites Films 49
   2.4.1 Thermo-Sealing Capacity 49
3 Recent Progress in Biocomposite of Biodegradable Polymer

Vicente de Oliveira Sousa Neto and Ronaldo Ferreira do Nascimento

3.1 Introduction 61
3.2 Biodegradable Polymers: Natural Origin and Development 63
3.3 Polysaccharides 63
  3.3.1 Polysaccharides from Vegetal Sources: Development and Application 64
    3.3.1.1 Cellulose 64
    3.3.1.2 Chitosan 69
3.4 Chemical Synthesis Produced Polymer 77
  3.4.1 Polylactic Acid 77
    3.4.1.1 Polylactic Acid: Structure and Properties 77
    3.4.1.2 Poly(lactic acid): Monomer from the Biomass 77
    3.4.1.3 Application and Advantage of Productions of PLA 78
    3.4.1.4 Packaging Materials: PLA 79
    3.4.1.5 PLA Fibers: Environment-Friendly Materials 80
3.5 Polyesters Produced by Microorganism or by Plants 83
  3.5.1 Polyhydroxy-Alcanoates 83
    3.5.1.1 PHA Blended with Others Biopolymers and Eco-Composites 84
    3.5.1.2 PHA-Based Green Renewable Eco-Composites 84
    3.5.1.3 Poly-3-hydroxybutyrate: Antiadhesion Applications 86
3.6 Concluding Remarks 87
References 88

4 Microbial Polyesters: Production and Market

Neha Patni, Yug Saraswat and Shibu G. Pillai

4.1 Introduction 95
4.2 Polyhydroxy Alkanoates 96
  4.2.1 Production 96
  4.2.2 Applications 97
  4.2.3 Organisms 98
  4.2.4 Co-Culture Production Strategy 100
  4.2.5 Biocompatibility and Rate of Drug Release 100
4.3 Bacterial Cellulose 100
  4.3.1 Production 101
  4.3.2 Applications 101
4.4 Polylactic Acid or Polylactide 102
4.5 Polyglycolic Acid 102
4.6 Brief Overview of the Local and World Scenario of Bioplastics 103
4.7 Summary 103
References 104
5 Biodegradable and Bioabsorbable Materials for Osteosynthesis Applications: State-of-the-Art and Future Perspectives 109

Sandra Carolina Cifuentes, Rosario Benavente, Marcela Lieblich and José Luis González-Carrasco

5.1 Introduction 109

5.2 State-of-the-Art 111

5.2.1 Poly(α-Hydroxyacids) as Biodegradable Materials for Osteosynthesis Implants 111

5.2.2 Mechanical Properties of Polylactic Acid 113

5.2.3 Degradation of Polylactic Acid 114

5.2.4 Biocompatibility of Polylactic Acid 117

5.3 Future Perspectives 117

5.3.1 Biodegradable Metals 118

5.3.1.1 Magnesium as a Biodegradable Material for Osteosynthesis Implants 118

5.3.1.2 Mechanical Properties of Mg and its Alloys 119

5.3.1.3 Degradation of Mg and its Alloys 120

5.3.1.4 Biocompatibility of Mg and its Alloys 123

5.3.2 Polymer/Mg Composites 125

5.3.2.1 Mechanical Properties of Polymer/Mg Composites 128

5.3.2.2 Degradation of Polymer/Mg Composites 129

5.3.2.3 Biocompatibility of Polymer/Mg Composites 130

5.4 Conclusions 131

References 132

6 Biodegradable Polymers in Tissue Engineering 145

Silvia Ioan and Luminita Ioana Buruiana

6.1 Introduction 145

6.2 Biodegradable Materials for Bone Tissue Engineering 146

6.3 Biocompatibility and Biodegradation of Polymer Networks 147

6.3.1 Parameters Influencing the Host Response 152

6.3.2 Host Response to Biomaterials 152

6.3.3 Materials Selected for Implantable Devices 152

6.3.4 Implantable Medical Devices 153

6.4 Biomaterial Reaction to Foreign Bodies 153

6.5 Design of Immunomodulatory Biomaterials 154

6.6 Applications Potential of Polyurethanes in Engineering Tissues 154

6.6.1 Biodegradation of Poly(urethane)s 157

6.6.2 Biodegradable Polyurethane Scaffolds for Regeneration and Tissue Repair 158

6.6.3 Tissue In-growth After Implantation of the Polyurethane Scaffold 158

6.6.4 In Vivo Cytokine-Associated Responses to Biomaterials 158

6.6.5 Thermostable, Biodegradable, and Biocompatible Hyperbranched Polyurethane/Ag Nanocomposites 159

6.6.6 Polyurethane Composite Scaffolds Containing Bioglass 160
### 6.7 Application Potential of Polycarbonates
- **6.7.1 Biocompatible Polycarbonates**
- **6.7.2 Bone–Polycarbonate Implant Interface**
- **6.7.3 Polycarbonates for Tissue Scaffold**
- **6.7.4 Polycarbonate Biomaterials for Tissue and Organ Regeneration**

### 6.8 Poly(amido Amine)
- **6.8.1 Gene Transfer via Hydrolytic Cationic Ester Polymers**
- **6.8.2 Poly(amido Amine)-Based Multilayered Thin Films for Surface-Mediated Cell Transfection**
- **6.8.3 Diagnostic Imaging of Pathologic Tissue in Cerebral Ischemic Zones**
- **6.8.4 Amine-Modified Polyesters as Biodegradable Gene Delivery Systems**
- **6.8.5 Reduction-Sensitive Polymers and Bioconjugates for Biomedical Applications**
- **6.8.6 Macromolecular Vehicules for the Intracellular and Controlled Delivery of Bioactive Molecules**

### 6.9 Polyester Amine
- **6.9.1 Lactic Acid-Based Poly(ester Amide)**
- **6.9.2 Biodegradable Elastomeric Polymers**
- **6.9.3 Functionalized Poly(ester Amide)s**
- **6.9.4 Polymeric Micelle as Intelligent Vehicles for Magnetic Resonance Imaging**

### 6.10 Polypyrrole-Based Conducting Polymers
- **6.10.1 Polypyrrole Compounds as Conductive Nerve Conduits**
- **6.10.2 Polypyrrole for Neural Tissue Applications**
- **6.10.3 Electro-Conductive Conjugated Polymers in Neural Stem Cell Differentiation**
- **6.10.4 Electroactive Tissue Scaffolds for Biomimetic Tissue**
- **6.10.5 Modulation of Hemocompatibility and Inflammatory Responses**
- **6.10.6 Keratinocytes Culture on Polypyrrole Films**

### 6.11 Remarks and Future Directions

### Acknowledgment

### References
8 Biodegradable Composites: Properties and Uses

Daniel Belchior Rocha and Derval dos Santos Rosa

8.1 Introduction

8.2 Biodegradable Polymers Applied in Composites

8.3 Composites Using Matrices by Biomass Polymers

8.4 Composites Using Matrices by Biopolymers Synthesized from Monomers

8.5 Composites Using Matrices by Biopolymers Produced by Microorganism

8.6 Conclusion

Acknowledgments

References

9 Development of Membranes from Biobased Materials and Their Applications

K.C. Khulbe and T. Matsuura

9.1 Introduction

9.2 Membranes from Biopolymer or Biomaterials

9.2.1 Algic Acid (Algin or Alginate)

9.2.2 Chitin and Chitosan

9.2.3 Cellulose

9.2.4 Polyamide

9.2.5 Polyhydroxalkanoates

9.2.6 Polylactic Acid

9.2.7 Other Biomaterials

9.2.7.1 C60 (Fullerene)

9.2.7.2 Marine Algie

9.2.7.3 Ferulic Acid

9.2.7.4 Polyethylene

References
Contents

9.2.7.5 Lignin 272
9.2.7.6 Biodegradable Polyvinyl Alcohol/Biopolymer Blends 273

9.3 Summary 274
References 275

10 Green Biodegradable Composites Based on Natural Fibers 283

Magdalena Wróbel-Kwiatkowska, Mateusz Kropiwnicki and Waldemar Rymowicz

10.1 Introduction 283
10.2 Plant Fibers Composition 284
10.3 Fiber Modifications 285
10.4 Composites Based on Different Plant Fibers 289
   10.4.1 Composites Based on Stem Fibers 290
      10.4.1.1 Hemp 290
      10.4.1.2 Kenaf 290
      10.4.1.3 Flax 290
   10.4.2 Leaf Fibers as Reinforcement of Composites 292
   10.4.3 Composites Based on Seed Fibers 292
   10.4.4 Composites Reinforced with Fruit Fibers 293
10.5 Future and Perspectives of Composites 293
10.6 Conclusions 295
References 295

11 Fully Biodegradable All-Cellulose Composites 303

Fabrizio Sarasini

11.1 Introduction 303
11.2 Self-Reinforced Composites 305
11.3 All-Cellulose Composites 306
   11.3.1 Nonderivatized All-Cellulose Composites 306
   11.3.2 Derivatized All-Cellulose Composites 314
11.4 Conclusions and Future Challenges 315
References 316

12 Natural Fiber Composites with Bioderivative and/or Degradable Polymers 323

Kamila Salasinska and Joanna Ryszkowska

12.1 Introduction 323
12.2 Materials 325
12.3 Methods for the Manufacture of Composites 326
12.4 Research Methodology of Plant Component and Composites 328
   12.4.1 Plant Component 328
      12.4.1.1 Evaluation of Particles Geometry 328
      12.4.1.2 Analysis of Particle Size Distribution 328
   12.4.2 Research Methodology of Composites 329
      12.4.2.1 Determination of the Density of Composites 329
      12.4.2.2 Composite Microstructure Assessment Using Scanning Electron Microscopy 329
References 329
12.4.2.3 Analysis of Materials’ Microstructure via Computer Microtomography 329
12.4.2.4 Impact Testing of Composite Materials 329
12.4.2.5 Determination of Strength Characteristics in the Static Tensile Test 330
12.4.2.6 Description of Materials’ Structure Using Differential Scanning Calorimetry 330
12.4.2.7 Features of Processes of Filler and Composite Degradation Based on Thermogravimetric Analysis Results 331
12.4.2.8 Evaluation of Composites’ Properties to Swell and Absorb Water 331
12.4.2.9 Analysis of Composites’ Resistance to Humidity in Cyclical Conditions 331

12.5 Test Results 332

12.5.1 Plant Component 332

12.5.1.1 Geometry of Particle Size 332
12.5.1.2 Grain Size Distribution 333

12.5.2 Composite Materials 334

12.5.2.1 Density Test 334
12.5.2.2 Assessment of Composite Microstructure Using Scanning Electron Microscopy 335
12.5.2.3 Analysis of Material Structure via Computer Microtomography 336
12.5.2.4 Impact Strength Testing of Composite Materials 339
12.5.2.5 Determination of Strength Characteristics in the Static Tensile Test 340
12.5.2.6 Evaluation of Material Structure with the Use of Differential Scanning Calorimetry 342
12.5.2.7 Features of the Composites Degradation Processes Based on the Results of Thermogravimetric Analysis 344
12.5.2.8 Evaluation of the Composites’ Capacity to Swell and Absorb Water 347
12.5.2.9 Analysis of the Composites’ Resistance to Humidity Under Cyclical Conditions 348

12.6 Comparison of the Properties of Composites with Different Types of Polymer Matrices 350

12.7 Summary and Conclusive Statements 351
Acknowledgments 352
References 352

13 Synthetic Biodegradable Polymers for Bone Tissue Engineering 355
Jiuhong Zhang, Zhiqiang Xie, Juan Yan and Jian Zhong 355

13.1 Introduction 355
13.2 Synthetic Biodegradable Polymers 356

13.2.1 Saturated Aliphatic Polyesters 357
13.2.2 Polypropylene Fumarate 359  
13.2.3 Polyanhydrides 360  
13.2.4 Poly(orthoesters) 360  
13.2.5 Poly(phosphazene) 361  
13.2.6 Copolymer, Polymer Blend/Composite 362

13.3 Physico-Chemical Characterizations of Polymeric Scaffolds 363  
13.3.1 Surface Property 363  
13.3.2 Porosity and Pore Size 364  
13.3.3 Biocompatibility 364  
13.3.4 Biodegradability 364  
13.3.5 Mechanical Properties 365  
13.3.6 Osteoinductivity 365

13.4 Definition and Clinical Needs of Bone Tissue Engineering 365  
13.4.1 Tissue Engineering 365  
13.4.2 Clinical Needs in Bone Tissue Engineering 366

13.5 Application of Synthetic Biodegradable Polymers in Bone Tissue Engineering 367

13.6 Summary 369  
Acknowledgments 370  
References 370

14 Polysaccharides as Green Biodegradable Platforms for Building-up Electroactive Composite Materials: An Overview 377  
Fernanda F. Simas-Tosin, Aline Grein-Iankovski, Marcio Vidotti  
and Izabel C. Riegel-Vidotti

14.1 Introduction 377

14.2 Main Chemical and Physical Chemical Properties of the Polysaccharides Used in the Synthesis of Electroactive Composites 379  
14.2.1 Structure 380  
14.2.1.1 Algal Polysaccharides: Alginate, Agar, and Carrageenan 380  
14.2.2.2 Polysaccharides from Exoskeletons of Arthropods: Chitin and Chitosan 384  
14.2.2.3 Glycosaminoglycans from Mammalian Cells: Hyaluronic Acid, Chondroitin Sulfate, and Heparin 385  
14.2.2.4 Some Plant Polysaccharides: Starch, Cellulose, and Gum Arabic 387

14.2.2 Relevant Physical–Chemical Properties 391

14.3 Electroactive Materials 394  
14.3.1 Basic Concepts 397  
14.3.2 Conducting Polymers 398  
14.3.3 Colloidal Synthesis of Conducting Nanoparticles in Aqueous Media 400

14.4 Spectroscopic Characterization of Colloidal Gum Arabic/Polyaniline and Gum Arabic/Poly(3,4-Ethylenedioxythiophene) 401

14.5 Polysaccharides/Conducting Polymer: Final overview 406

References 409
15  Biodegradable Polymer Blends and Composites from Seaweeds 419
Yolanda Freile-Pelegrín and Tomás J. Madera-Santana
15.1 Introduction 419
15.2 Seaweed Resources: World Scenario 420
  15.2.1 Classification of Seaweed 420
  15.2.2 Commercial Seaweeds: Uses and Applications 420
  15.2.3 Wild and Cultured Seaweed as Feedstock for Biodegradable Polymers 422
15.3 Seaweed Polymers with Potential Materials Applications 422
  15.3.1 Sulfated Galactans Derived from Red Seaweeds: Agar and Carrageenans 422
  15.3.2 Polymer Derived from Brown Seaweeds: Alginates and Fucoidan 424
  15.3.3 Polymers Derived from Green Seaweeds: Ulvan 425
15.4 Potential Biopolymer Blends and Composites from Seaweeds 426
  15.4.1 Biopolymer Blends Based on Agar and Others Phycocolloids 426
  15.4.2 Biopolymer Composites Based on Agar and Others Phycocolloids 430
  15.4.3 Perspectives and Novel Applications of Biopolymers from Seaweeds 433
References 433

16  Biocomposite Scaffolds Derived from Renewable Resources for Bone Tissue Repair 439
S. Dhivyaa and N. Selvamurugan
16.1 Introduction 439
16.2 Polysaccharide-Based Polymers 440
  16.2.1 Chitosan 440
  16.2.2 Alginate 442
  16.2.3 Bacterial Cellulose 444
  16.2.4 Lignocellulose 448
  16.2.5 Hyaluronic Acid 449
  16.2.6 Fucoidan 452
  16.2.7 Ulvan 453
  16.2.8 Pullulan 454
  16.2.9 Acemannan 455
16.3 Glycosaminoglycans 455
16.4 Protein-Based Polymers 459
  16.4.1 Silk 459
  16.4.2 Collagen and Gelatin 460
16.5 Polyesters 463
  16.5.1 Polyurethanes 463
16.6 Polyhydroxyalkanoates 465
  16.6.1 Poly(3-Hydroxybutyric Acid-co-3-hydroxyvaleric Acid), Polyhydroxyl Butyrate and Poly(3-Hydroxybutyrate-co-3-hydroxyhexanoate) 465
16.7 Others 466
16.8 Conclusions and Future Direction 467
Acknowledgment 468
Abbreviations 468
References 470

17 Pectin-Based Composites 487
Veronika Bátori, Dan Åkeson, Akram Zamani
and Mohammad J. Taherzadeh
17.1 Introduction 487
17.2 Pectin 488
  17.2.1 Properties and Structure of Pectin 489
    17.2.1.1 Structure of Pectin 489
    17.2.1.2 Properties of Pectin 491
    17.2.1.3 Bioactivity of Pectin 493
17.3 Biosynthesis of Pectin Polymers during Cell Differentiation 495
17.4 Production of Pectin 495
  17.4.1 Extraction of Pectin 496
  17.4.2 Designer Pectins 498
  17.4.3 Applications and Markets 499
17.5 Pectin-Based Biocomposites 499
  17.5.1 Definition 499
    17.5.1.1 Biodegradability and Durability of Composite Materials 500
  17.5.2 Pectin-Based Biocomposites 501
  17.5.3 Applications, Applied Materials, and Methods in Pectin-Based Composites 504
    17.5.3.1 Bone Regeneration and Tissue Engineering 504
    17.5.3.2 Wound Healing 508
    17.5.3.3 Application to Bone Hemorrhage 508
    17.5.3.4 Drug Delivery Applications 509
    17.5.3.5 Copper Removal from Aqueous Solutions 509
    17.5.3.6 Environmental Remediation 510
    17.5.3.7 Cationic Dye Adsorption 510
    17.5.3.8 Active Packaging 511
    17.5.3.9 Biobased Membranes and Films with Improved Mechanical and Thermal Properties 511
    17.5.3.10 Casing Film for Sausages 512
    17.5.3.11 Sensors for Determining Different Substances (Electrochemical Method) 512
17.6 Conclusions 513
References 513

18 Recent Advances in Conductive Composites Based on Biodegradable Polymers for Regenerative Medicine Applications 519
Ilaria Armentano, Elena Fortunati, Luigi Torre and José Maria Kenny
18.1 Introduction 519
18.2 Regenerative Medicine 520
18.3 Biodegradable Polymers 521
18.4 Conductive Nanostructures 524
  18.4.1 Carbon Nanotubes 525
  18.4.2 Graphene 525
18.5 Polymer Nanocomposite Approach 526
  18.5.1 Processing Technology 527
  18.5.2 Techniques Used in Estimating Conductive Properties 528
    18.5.2.1 Electrical Properties 528
    18.5.2.2 Dielectrical Properties 531
  18.5.3 Biomedical Applications 533
18.6 Conclusions and Future Perspectives 535
References 536

19 Biosynthesis of PHAs and Their Biomedical Applications 543
K.-S. Heng, Y.-F. Lee, L. Thinagaran, J.-Y. Chee,
P. Murugan and K. Sudesh
19.1 Introduction 543
19.2 Genetic and Metabolic Pathway of PHA Production 545
19.3 PHA Production from Sugars 548
19.4 PHA Production from Oils 554
19.5 Exploration and Application of PHAs as Biomaterials 566
  19.5.1 PHA-Based Monofilament Sutures/Fibers 569
  19.5.2 PHA-Based Scaffolds 570
  19.5.3 PHA Microspheres and Nanoparticles 571
19.6 Future Perspectives 573
Acknowledgments 574
References 574

20 Biodegradable Soy Protein Isolate/Poly(Vinyl Alcohol) Packaging Films 587
Jun-Feng Su
20.1 Introduction 587
20.2 Experimental 589
  20.2.1 Materials 589
  20.2.2 Preparation of SPI/PVA Blend Films 589
  20.2.3 Characterizations 590
    20.2.3.1 Surface Morphologies 590
    20.2.3.2 X-ray Diffraction Analysis 590
    20.2.3.3 Fourier Transform Infrared Spectra Analysis 590
    20.2.3.4 Mechanical Properties 590
    20.2.3.5 Differential Scanning Calorimetry 591
    20.2.3.6 Thermogravimetric Analysis 591
    20.2.3.7 Water Solubility 591
    20.2.3.8 Heat-Sealing Process 591
    20.2.3.9 Peel Strength Tests 592
    20.2.3.10 Tensile Tests of Heat Sealing 592
    20.2.3.11 Moisture Sorption 593
References 594
20.2.3.12 Contact Angle Determination 594
20.2.3.13 Water Vapor Permeability 595
20.2.3.14 Aerobic Biodegradation Tests 596
20.2.3.15 Weight Loss and Surface Morphologies Characterization 597
20.2.3.16 Statistical Analysis 597

20.3 Results and Discussion

20.3.1 Surface Morphologies of Films 597
20.3.2 Compatibility of SPI/PVA Blends 598
20.3.3 FTIR Analysis 601
20.3.4 Mechanical Properties of Films 603
20.3.5 Thermal Stability of Films 604
20.3.6 Effect of Heat-Sealing Temperature on Peel Strength 606
20.3.7 Effect of Heat-Sealing Temperature on Tensile Strength 608
20.3.8 Water Sorption Isotherms 609
20.3.9 Guggenheim–Anderson–DeBoer Model Fitting 610
20.3.10 Contact Angle Measurements 614
20.3.11 WVP of Films 617
20.3.12 Aerobic Biodegradation of SPI/PVA Films 618
20.3.13 Weight Loss and Surface Morphologies of Films during Biodegradation 619

20.4 Conclusion

References 620

21 Biodegradability of Biobased Polymeric Materials in Natural Environments 625
Sudhakar Muniyasamy and Maya Jacob John

21.1 Introduction 625
21.2 Biobased Polymers from Renewable Resources 629
21.2.1 Extraction of Biopolymers from Biomass 630
21.2.2 Biobased Polymers Derived from Biobased Monomer 631
21.2.3 Biobased Polymers from Bacterial Synthesis 632
21.3 Biodegradable and Compostable Polymeric Materials from Renewable Resources 632
21.3.1 Definition and Concept 633
21.3.2 Biodegradation Standard Test Methods 635
21.4 Overview of Biodegradation Studies of Biobased Polymers in Different Environmental Conditions 640
21.5 Biodegradation Mechanisms of Biobased Polymeric Materials 645
21.6 Concluding Remarks 648

References 649

Index 655
The concept of green chemistry and sustainable development policy impose on industry and technology to switch raw material base from the petroleum to renewable resources. Remarkable attention has been paid to the environmental-friendly, green, and sustainable materials for a number of applications during the past few years. Indeed, the rapidly diminishing global petroleum resources, along with awareness of global environmental problems, have promoted the way to switch toward renewable resources-based materials. In this regard, biobased renewable materials can form the basis for a variety of eco-efficient, sustainable products that can capture and compete markets presently dominated by products based solely on petroleum-based raw materials. The nature provides a wide range of the raw materials that can be converted into a polymeric matrix/adhesive/reinforcement applicable in composites formulation. Different kinds of polymers (renewable/nonrenewable) and polymer composite materials have been emerging rapidly as the prospective substitute to the ceramic or metal materials, due to their advantages over conventional materials. In brief, polymers are macromolecular groups collectively recognized as polymers due to the presence of repeating blocks of covalently linked atomic arrangement in the formation of these molecules. The repetitive atomic arrangements forming the macromolecules by forming covalent links are the building block or constituent monomers. As the covalent bond formation between monomer units is the essence of polymer formation, polymers are organic or carbon compounds of either biological or synthetic origin. The phenomenon or process of polymerization enables to create diverse forms of macromolecules with varied structural and functional properties and applications. On the other hand, composite materials, or composites, are one of the main improvements in material technology in recent years. In the materials science field, a composite is a multiphase material consisting of two or more physically distinct components, a matrix (or a continuous phase) and at least one dispersed (filler or reinforcement) phase. The dispersed phase, responsible for enhancing one or more properties of matrix, can be categorized according to particle dimensions that comprise platelet, ellipsoids, spheres, and fibers. These particles can be inorganic or organic origin and possess rigid or flexible properties.

The most important resources for renewable raw materials originate from nature such as wood, starch, proteins, and oils from plants. Therefore, renewable raw materials lead to the benefit of processing in industries owing to the short period of replenishment cycle resulting in the continuous-flow production. Moreover, the production cost can be reduced by using natural raw materials instead of chemical raw materials. The waste and residues from agriculture and industry have also been used as an alternative renewable resources for producing energy and raw materials such as chemicals,
cellulose, carbon, and silica. For polymer composites applications, an intensifying focus
has been directed toward the use of renewable materials. Biobased polymers are one of
the most attractive candidates in renewable raw materials for use as organic-reinforcing
fillers such as flex, hemp, pine needles, coir, jute, kenaf, sisal, rice husk, ramie, palm,
and banana fibers, which exhibited excellence enhancement in mechanical and thermal
properties. For green polymer composites composed of inorganic-reinforcing fillers,
renewable resources-based polymers have been used as matrix materials.

Significant research efforts all around the globe are continuing to explore and
improve the properties of renewable polymers-based materials. Researchers are col-
lectively focusing their efforts to use the inherent advantages of renewable polymers for
miscellaneous applications. To ensure a sustainable future, the use of biobased materi-
als containing a high content of derivatives from renewable biomass is the best solution.

This volume of the book series 'Handbook of Composites from Renewable Materials'
is solely focused on the 'Biodegradable Materials'. Some of the important topics include
but not limited to rice husk and its composites; biodegradable composites based on
thermoplastic starch and talc nanoparticles; recent progress in biocomposites of bio-
degradable polymer; microbial polyesters: production and market; biodegradable and
bioabsorbable materials for osteosynthesis applications; biodegradable polymers in tis-
ue engineering; composites based on hydroxyapatite and biodegradable polylactide;
biodegradable composites; development of membranes from biobased materials and
their applications; green biodegradable composites based on natural fibers; fully bio-
degradable all-cellulose composites; natural fiber composites with bioderivative and/
or degradable polymers; synthetic biodegradable polymers for bone tissue engineer-
ing; polysaccharides as green biodegradable platforms for building up electroactive
composite materials; biodegradable polymer blends and composites from seaweeds;
biocomposites scaffolds derived from renewable resources for bone tissue repair; pect-
in-based composites; recent advances in conductive composites based on biodegrad-
able polymers for regenerative medicine applications; biosynthesis of PHAs and their
biomedical applications; biodegradable soy protein isolate/poly(vinyl alcohol) packag-
ing films; and biodegradability of biobased polymeric materials in natural environment.

Several critical issues and suggestions for future work are comprehensively discussed
in this volume with the hope that the book will provide a deep insight into the state
of the art of ‘Biodegradation’ of the renewable materials. We would like to thank the
Publisher and Martin Scrivener for the invaluable help in the organization of the edit-
ing process. Finally, we would like to thank our parents for their continuous encoura-
gement and support.

Vijay Kumar Thakur, Ph.D.
University of Cranfield, U.K.

Manju Kumari Thakur, M.Sc., M.Phil., Ph.D.
Himachal Pradesh University, Shimla, India

Michael R. Kessler, Ph.D., P.E.
Washington State University, U.S.A.
Rice Husk and its Composites: Effects of Rice Husk Loading, Size, Coupling Agents, and Surface Treatment on Composites’ Mechanical, Physical, and Functional Properties

A. Bilal, R.J.T. Lin* and K. Jayaraman

Centre for Advanced Composite Materials, Department of Mechanical Engineering, University of Auckland, Auckland, New Zealand

Abstract

Among the many natural fibers used as reinforcements/fillers in the manufacture of natural fiber composite materials, rice husk (RH) has not been attracting the deserved attention despite its significant annual yield of tens of million tons due to the huge worldwide rice-consuming population. This chapter presents an introduction to natural fibers and their composites with an emphasis on RH and its use in the manufacture of composite materials. A thorough review has been carried out on the manufacturing of RH composites with various polymers and manufacturing processes. The effects of RH loading, size, surface treatment, and the use of coupling agents on mechanical, physical, and functional properties of RH composites have been discussed in detail. Although RH has also been used in the form of ash in manufacturing different composites, this chapter only focuses on RH used in its natural form and its resulting composites.

Keywords: Rice husk, coupling agents, surface treatment, composites manufacturing, mechanical, physical and functional properties

1.1 Introduction

By definition, natural fibers are fibers which are not artificial or manmade (Ticoalu et al., 2010). Natural fibers can be plant based such as wood, sisal, flax, hemp, jute, kenaf, and ramie or animal based, e.g., wool, avian feather, and silk or mineral based such as basalt and asbestos. They have been used as reinforcements with a variety of materials for over 3000 years (Taj et al., 2007) and have demonstrated immense potential to replace synthetic fibers, such as glass and carbon fibers, because of their eco-friendly and biodegradable characteristics.
There is a large variation in the properties of natural fibers, which is affected by several factors such as fiber’s place of growth, cultivation conditions, growth time, moisture content, and form (yarn, woven, twine, chopped, and felt) (O’Donnell et al., 2004; Ochi, 2008; Pickering et al., 2007). Table 1.1 shows various plant-based natural fibers and their regions or countries of origin.

The mechanical and physical properties of natural fibers are greatly affected by their chemical composition and structure (Taj et al., 2007). The majority of plant-based natural fibers have cellulose, hemicellulose, and lignin as their main constituents along with pectin and waxes (John & Thomas, 2008). The reinforcing ability of natural fibers depends on cellulose and its crystallinity (Bledzki & Gassan, 1999, John & Thomas, 2008), whereas biodegradation, micro-absorption, and thermal degradation of natural fibers depend on hemicelluloses (Taj et al., 2007), which is hydrophilic in nature (John & Thomas, 2008). On the other hand, lignin which is hydrophobic in nature plays a critical role in protecting the cellulose/hemicellulose from severe environmental conditions such as water (Thakur & Thakur, 2014), and is thermally stable but prone to UV degradation (Olesen & Plackett, 1999); pectin gives plants flexibility, while waxes consist of various types of alcohols (John & Thomas, 2008). Each of these constituents of natural fibers plays an important role in determining the overall properties of natural fibrous materials (Thakur et al., 2014b).

These fibers are chemically active and decompose thermo-chemically between 150 °C and 500 °C (cellulose between 275 °C and 350 °C; hemicellulose mainly between 150 °C and 350 °C; and lignin between 250 and 500 °C) (Kim et al., 2004).

The relative percentages of cellulose, hemicellulose, and lignin vary for different fibers (John & Thomas, 2008). Table 1.2 shows the chemical composition of some natural fibers.

| **Table 1.1** Fibers and their origin (Taj et al., 2007; Kim et al., 2007). |
|-----------------|---------------------------------------------------------------|
| **Fibers**      | **Regions/countries of origin**                              |
| Flax            | Borneo                                                        |
| Hemp            | Former Yugoslavia, China                                     |
| Sun hemp        | Nigeria, Guyana, Sierra Leone, India                         |
| Ramie           | Honduras, Mauritius                                           |
| Jute            | India, Egypt, Guyana, Jamaica, Ghana, Malawi, Sudan, Tanzania|
| Kenaf           | Iraq, Tanzania, Jamaica, South Africa, Cuba, Togo             |
| Roselle         | Borneo, Guyana, Malaysia, Sri Lanka, Togo, Indonesia, Tanzania|
| Sisal           | East Africa, Bahamas, Antigua, Kenya, Tanzania, India         |
| Abaca           | Malaysia, Uganda, Philippines, Bolivia                       |
| Coir            | India, Sri Lanka, Philippines, Malaysia                       |
| Rice husk       | Asia, Pacific rim, North America                             |
Generally, an increase in the cellulose content increases tensile strength and Young’s modulus of fibers, whereas stiffness also depends on the micro-fibrillar angle. Fibers are rigid, inflexible, and have high tensile strength if the micro-fibrils have an orientation parallel to the fiber axis. If the micro-fibrils are oriented in a direction spiral to the fiber axis, the fibers are more ductile (John & Thomas, 2008). This variation of material properties does cause some concerns about the use of such materials in the more advanced and critical applications such as composite components for automobiles, infrastructure, aeronautical, and aerospace industries.

Agricultural wastes such as RH, wheat straw, rice straw, and corn stalks also come under the category of natural fibers. Researchers are now increasingly looking toward these by-products for manufacturing composite materials (Panthapulakkal et al., 2005b; Nourbakhsh & Ashori, 2010; Ghofrani et al., 2012). The use of these agricultural by-products provides great opportunity to start a natural fiber industry in those countries which have little or no wood resources (Ashori & Nourbakhsh, 2009). The chemical components and contents of these materials are similar to those of wood and they can be used in the form of fibers or particles (Yang et al., 2004; Yang et al., 2006b). With the comparatively large quantity of agro-wastes from annual crops, Table 1.3, there is a potential that wood can be substituted by these alternative materials (Ashori & Nourbakhsh, 2009). These agro-residues are normally used as animal feed or household fuel and a large proportion is burned for disposal, which adds to environmental pollution (Ashori & Nourbakhsh, 2009). These agricultural waste fibers can be formed into chips or particles similar to wood (Yang et al., 2003), and their exploration and utilization will contribute to rural agricultural-based economies in a positive way (Sain & Panthapulakkal, 2006).

### Table 1.2 Chemical composition of some natural fibers (Malkapuram et al., 2009).

<table>
<thead>
<tr>
<th>Fiber</th>
<th>Cellulose (wt%)</th>
<th>Hemicellulose (wt%)</th>
<th>Lignin (wt%)</th>
<th>Pectin (wt%)</th>
<th>Wax (wt%)</th>
<th>Moisture content (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wood</td>
<td>40–50</td>
<td>15–25</td>
<td>15–30</td>
<td>–</td>
<td>–</td>
<td>8–16</td>
</tr>
<tr>
<td>Jute</td>
<td>61–71.5</td>
<td>13.6–20.4</td>
<td>12–13</td>
<td>0.4</td>
<td>0.5</td>
<td>12.6</td>
</tr>
<tr>
<td>Hemp</td>
<td>70.2–74.4</td>
<td>17.9–22.4</td>
<td>3.7–5.7</td>
<td>0.9</td>
<td>0.8</td>
<td>10</td>
</tr>
<tr>
<td>Kenaf</td>
<td>31–39</td>
<td>21.5</td>
<td>15–19</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Flax</td>
<td>71</td>
<td>18.6–20.6</td>
<td>2.2</td>
<td>2.3</td>
<td>1.7</td>
<td>10</td>
</tr>
<tr>
<td>Sisal</td>
<td>67–78</td>
<td>10–14.2</td>
<td>8–11</td>
<td>10</td>
<td>2.0</td>
<td>11</td>
</tr>
<tr>
<td>Coir</td>
<td>36–43</td>
<td>10–20</td>
<td>41–45</td>
<td>3–4</td>
<td>–</td>
<td>8</td>
</tr>
<tr>
<td>Bamboo</td>
<td>26–49</td>
<td>15–27.7</td>
<td>21–31</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

1.2 Natural Fiber-Reinforced Polymer Composites

Composite materials consist of two or more ingredients in which one component acts as the matrix material and the other as the reinforcement (Pappu et al., 2015) and their
overall properties depend on the individual characteristics of the polymer matrix and the reinforcement (Thakur et al., 2014a).

Although research on natural fiber-reinforced polymer composites (NFRCs) began in 1908 (John & Thomas, 2008), it has not received much attention until from about three decades ago (Westman et al., 2010). Nowadays, both the academic and industrial sectors are showing a significantly increased interest in the use of NFRCs due to their low cost, environmental friendliness, lightweight, biodegradable, and nonabrasive nature (Rozman et al., 2000). Moreover, they have high electrical resistance, good acoustic insulating properties, low energy consumption, less dermal and respiratory irritation, good chemical and corrosion resistance, and are safe to handle (Ticoalu et al., 2010; Taj et al., 2007; John & Thomas, 2008; Malkapuram et al., 2009; Ashori, 2008; Mavani et al., 2007).

With the reported advantages of NFRCs and the growing awareness on the depletion of petroleum-based resources as well as global environmental issues, the demand of NFRCs has predicted to grow 15–20% annually with a growth rate of 15–20% in automotive applications (Malkapuram et al., 2009), and 50% or more in building and construction applications. North America is known as the leading region of NFRC applications in the building and construction sectors with mainly wood fiber-based composites, whereas Europe is the leading region of NFRC applications in the automotive industries with mostly nonwood fiber-based composites (Lucintel, 2011). Of course, there are other NFRC applications emerging in the other regions of the globe. The earlier forecast for the NFRC market was with a compound annual growth rate

<table>
<thead>
<tr>
<th>Fiber source</th>
<th>World production 10^3 tons</th>
<th>Origin</th>
<th>Fiber source</th>
<th>World production 10^3 tons</th>
<th>Origin</th>
</tr>
</thead>
<tbody>
<tr>
<td>Abaca</td>
<td>70</td>
<td>Leaf</td>
<td>Nettles</td>
<td>Abundant</td>
<td>Stem</td>
</tr>
<tr>
<td>Bamboo</td>
<td>10,000</td>
<td>Stem</td>
<td>Oil palm fruit</td>
<td>Abundant</td>
<td>Fruit</td>
</tr>
<tr>
<td>Banana</td>
<td>200</td>
<td>Stem</td>
<td>Palm rah</td>
<td>--</td>
<td>Stem</td>
</tr>
<tr>
<td>Broom</td>
<td>Abundant</td>
<td>Stem</td>
<td>Ramie</td>
<td>100</td>
<td>Stem</td>
</tr>
<tr>
<td>Coir</td>
<td>100</td>
<td>Fruit</td>
<td>Roselle</td>
<td>250</td>
<td>Stem</td>
</tr>
<tr>
<td>Cotton lint</td>
<td>18,500</td>
<td>Stem</td>
<td>Rice husk</td>
<td>Abundant</td>
<td>Fruit/grain</td>
</tr>
<tr>
<td>Elephant grass</td>
<td>Abundant</td>
<td>Stem</td>
<td>Rice straw</td>
<td>Abundant</td>
<td>Stem</td>
</tr>
<tr>
<td>Flax</td>
<td>810</td>
<td>Stem</td>
<td>Sisal</td>
<td>380</td>
<td>Stem</td>
</tr>
<tr>
<td>Hemp</td>
<td>215</td>
<td>Stem</td>
<td>Sun hemp</td>
<td>70</td>
<td>Stem</td>
</tr>
<tr>
<td>Jute</td>
<td>2,500</td>
<td>Stem</td>
<td>Wheat straw</td>
<td>Abundant</td>
<td></td>
</tr>
<tr>
<td>Kenaf</td>
<td>770</td>
<td>Wood</td>
<td></td>
<td>1,75,000</td>
<td></td>
</tr>
<tr>
<td>Linseed</td>
<td>Abundant</td>
<td>Fruit</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 1.3 Annual production of natural fibers and sources (Taj et al., 2007).
Rice Husk and its Composites

(RH) of 10% to reach $3.8B by 2016 (Lucintel, 2011); interestingly, it has shown that RH is emerging as an alternative for wood fibers in the applications of the building and construction sectors.

Despite the promising forecast, NFRCs do have some inherent issues which need to be addressed properly before their full potential can be realized for widespread industrial applications in various sectors. Along with the nature of the fibers, the properties of the resulting composites are also influenced by the type of polymer matrix used and the amount and dimensions of the fiber. One of the critical issues is the weak adhesion and poor interfacial bond strength between natural fibers and the matrix (Lee et al., 2004; Hristov et al., 2004), and formation of aggregates during their processing (Taj et al., 2007; Ashori, 2008), leading to inferior mechanical properties. Natural fibers are polar and hydrophilic in nature and polymer matrix is nonpolar and hydrophobic, which form the heterogeneous systems for NFRCs. Surface tension as well as polarity of matrix and the fibers should be the same in order to have good interfacial adhesion in NFRCs (Mwaikambo & Ansell, 2002), and a suitable adhesion is required between the matrix and the filling material in order to improve mechanical properties of the composites (Yang et al., 2004).

Due to the hydrophilicity of natural fibers, NFRCs could absorb water when used in moist conditions which leads to the poor compatibility between fibers and hydrophobic polymer matrices (Yang et al., 2006a). The water absorption (WA) is due to the hydrogen bond developed between the hydroxyl groups (OH) in the natural fibers and water molecules present in the environment. Therefore, it is essential to prevent such moisture infiltration so that swelling and/or permanent damage can be avoided for effective usage of these cellulosic composites in wet conditions (Ishak et al., 2001).

In order to enhance the performance of NFRCs, the compatibility between hydrophilic fibers and hydrophobic polymers can also be improved by using coupling agents and/or surface modification of fibers. These measures can create efficient interfacial bond strength between the fibers and the polymer matrices so that the effective load transfer can be achieved when NFRCs are subjected to loading during applications.

Coupling agents, who have both the hydrophilic and hydrophobic properties necessary to bond well with the fiber and the polymer matrix, make polymers more reactive toward the surface of the natural fiber (Panthapulakkal et al., 2005b; Ershad-Langroudi et al., 2008; Ahmad Fuad et al., 1993; Stark & Rowlands, 2003; Toro et al., 2005; Park et al., 2004; Sombatsompop et al., 2005). They chemically link with the hydrophilic fiber on one side and the hydrophobic polymer chain on the other to facilitate the wetting of polymer surrounding the fibers. The interfacial region between the fiber and the matrix has two types of interaction, i.e., primary and secondary bonding represented by covalent bonding and hydrogen bonding, respectively (Rozman et al., 2005a; Rozman et al., 2003).

1.3 Rice Husk and its Composites

Rice is a source of primary food for the majority of the population worldwide. Around 20 wt% of paddy received is husk which is separated from the rice grain during milling process (Chand et al., 2010); therefore, rice husk (RH) is abundantly available
in significant quantity. The annual production of rice in 2012 was approximately 718 million tons according to the Food and Agriculture Organization of the United Nations (FAO, 2012). RH is biodegradable, abundant, cost effective, lightweight, easily available, reduces the density of the finished product, has no residues or toxic by-products, is environmentally friendly, low density, and recyclable (Yang et al., 2004; Yang et al., 2006a; Ibrahim and Kuek, 2011; Rahman et al., 2010a; Premalal et al., 2002).

RH is mainly used as fuel, fertilizer in agriculture, landfill, and animal bedding (Kim & Eom, 2001; Park et al., 2003; Mano, 2002), but the majority of RH is burnt for disposal because of its resistance to decomposition in the ground, and its difficulty to digest and low nutritional value for animals (Piva et al., 2004). In the past few years, researchers have looked into the possibility of using RH, which is mostly an unwanted material, for making composite materials (Razavi-Nouri et al., 2006).

Similar to other natural fibers, RH has cellulose, hemicelluloses and lignin as its main constituents (George and Ghose, 1983), noticeably it also contains significant amount of silica (20 wt%), which is present on its outer surface in the form of silicon–cellulose membrane (Yoshida, 1962). RH has a cellulose content (35 wt%) similar to that of wood (Martí-Ferrer et al., 2006; Rosa et al., 2009b) but has lower contents of lignin (20 wt%) and hemicellulose (25 wt%) than those found in most other natural fibers including wood. Since the thermal degradation of RH occurs due to the degradation of hemicellulose and lignin (Kim et al., 2004), a similar mechanism to that of wood, the lower content of lignin and hemicellulose allows RH-filled polymers to be processed at higher temperatures as compared to wood polymer composites. While wood has thermal stability issues at temperatures over just 200 °C, RH degrades and decomposes at temperatures around 250 °C which enables the manufacturing of RH composites to be performed at higher temperatures up to 250 °C (Martí-Ferrer et al., 2006) without concern of losing material properties.

RH as reinforcement/filler in polymer-based composite materials has proven to be a good option, provided there is good compatibility between RH and base polymer matrix (Chand et al., 2010). Like other plant-based natural fibers, RH is hydrophilic and its use with hydrophobic thermoplastics results in poor compatibility and adhesion between the counter parts (Panthapulakkal et al., 2005a; Dhakal et al., 2007; Sain & Kokta, 1993; Lai et al., 2003; Kazayawoko et al., 1999; Sain et al., 1993; Li & Matuana, 2003). One of the reasons for poor adhesion is the presence of silica, which is present in the form of a silicon–cellulose membrane on the outer surface of RH (Vasisht, 1974). Removal of silica and other surface impurities can result in a better adhesion between the fiber and the matrix and in turn improve properties of composites (Sain & Panthapulakkal, 2006). Fiber matrix adhesion can also be improved by introducing coupling agents (Panthapulakkal et al., 2005a; Dhakal et al., 2007; Kazayawoko et al., 1999, Lai et al., 2003; Sain et al., 1993; Sain & Kokta, 1993).

RH is also more resistant to WA and fungal decomposition because it contains 20 wt% amorphous silica in combination with 30 wt% of a phenyl propanoid structural polymer called lignin (Rahman et al., 2010b). As mentioned earlier, common NFRCs have a major disadvantage of WA mainly due to diffusion or infiltration (Czél & Kanyok, 2007). In the case of RH, the percentage of cellulose is very low and the waxes contained also make it comparatively less prone to water uptake.
Composites made from RH have better dimensional stability under moist conditions, good termite resistance, and high resistance to biological attack as compared to wood-based materials (Kim et al., 2007). These RH composites have reasonable strength and stiffness, no residues or toxic by-products when burnt, are recyclable, and low CO$_2$ emissions when compared with inorganic-filler-reinforced polymer composites (Kim et al., 2007; Yang et al., 2006a; Razavi-Nouri et al., 2006; Kim et al., 2005).

Flammability is another problem faced by natural fiber composites. Synthetic polymers are petroleum based and are highly flammable. Various flame-retardant materials such as halogen and phosphorus-based compounds can be used with polymers to improve flame retardancy, but these flame retardants have a negative impact on the environment and raise health concerns as well (Zhao et al., 2009). RH could prove to be a good flame-retardant material in composites as it contains silica as one of the main constituents. Silica is mainly responsible for the improved flame retardancy by providing thermal shielding and diffusion barrier effects during the combustion process (Zhao et al., 2009; Arora et al., 2012).

RH has been used both in thermoplastics and thermosets. The following subsections discuss a wide range of research undertaken in the area of RH composites. The main focus of discussion is the type of polymers and manufacturing processes involved in the manufacture of RH composites. The effects of RH loading and coupling agents on mechanical, physical and functional properties of RH composites are also discussed.

### 1.3.1 Polymers Used in the Manufacturing of RH Composites

Over the past two decades, although both thermoplastics and thermosets have been used as matrices in manufacturing of RH composites, yet thermoplastic polymers have been the primary candidate for RH composites. Among the commonly available thermoplastic resins, PE and PP of different densities (i.e., low, medium, and high) have been used the most. On one hand, PE is primarily used as an exterior building component. Low-density polyethylene (LDPE) has properties such as fluidity, flexibility, transparency, and a glossy surface and has been used mainly as a food packing material in the forms of sheet and film; whereas, high-density polyethylene (HDPE) has toughness, stiffness, solvent resistance, and electrical insulation and is mainly used as an insulating material for electric wire and for producing various types of containers (Yang et al., 2007b). The manufacturing of composites with RH as reinforcement and PE (low, medium, and high densities) as polymer matrix has been carried out by quite a number of researchers (Yang et al., 2007b; Kim et al., 2004; Panthapulakkal et al., 2005b; Ghofrani et al., 2012; Yang et al., 2006a; Rahman et al., 2010a; Panthapulakkal et al., 2005a; Rahman et al., 2010b; Zhao et al., 2009; Khalf & Ward, 2010; Najafi & Khademi-Eslam, 2011; Fávaro et al., 2010; Syafri et al., 2011; Rahman et al., 2011; Bilal et al., 2014a-c).

On the other hand, PP is one of the most widely used packaging materials (George et al., 2007). It is also commonly used in the automotive industry and recently has been studied for use as building profiles (Razavi-Nouri et al., 2006). Similar to PE, composites manufactured with PP (low, medium, and high densities) and RH has also been widely researched (Kim et al., 2007; Kim et al., 2004; Ashori & Nourbakhsh, 2009; Yang et al., 2004; Yang et al., 2006a,b; Ishak et al., 2001; Ershad-Langroudi et al., 2008;
Handbook of Composites from Renewable Materials-Volume 5

Premalal et al., 2002; Razavi-Nouri et al., 2006; Rosa et al., 2009a,b; Czél & Kanyok, 2007; Santiagoo et al., 2011; El Sayed et al., 2012; Aminullah et al., 2010; Yang et al., 2007a; He et al., 2011; Yussuf et al., 2010, de Carvalho et al., 2011; Nourbakhsh et al., 2014.

Apart from PE and PP, phenol formaldehyde (PF) (Bhatnagar, 1994; Ndazi et al., 2007), polyurethane (PU) (Sheriff et al., 2009; Rozman et al., 2003), polyester (Rozman et al., 2005a; Ahmad et al., 2007, Rozman et al., 2005b), polymer lactic acid (PLA) (Yussuf et al., 2010, Hua et al., 2011), polyvinylchloride (PVC) (Chand et al., 2010), polyvinyl alcohol (PVA) (Arora et al., 2012), polystyrene (Rozman et al., 2000), urea formaldehyde (UF) (Bakar & Muhammed, 2011), and epoxy (Ibrahim & Kuek, 2011) have also been used to manufacture composites with RH.

Injection molding, compression molding, extrusion, and hot press are the most commonly used techniques to manufacture RH-reinforced composite materials. The manufacturing of composites with different manufacturing processes using RH is shown in Table 1.4.

### Table 1.4 Manufacturing processes used in the manufacture of RH composites.

<table>
<thead>
<tr>
<th>Manufacturing process</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Injection molding</td>
<td>Kim et al., 2007; Ashori &amp; Nourbakhsh, 2009; Yang et al., 2004;</td>
</tr>
<tr>
<td></td>
<td>Yang et al., 2006b; Yang et al., 2006a; Ishak et al., 2001; Ershad-</td>
</tr>
<tr>
<td></td>
<td>Langroudi et al., 2008; Rahman et al., 2010a; Razavi-Nouri et al.,</td>
</tr>
<tr>
<td></td>
<td>2006; Rahman et al., 2010b; Czél &amp; Kanyok, 2007; Yang et al., 2007b;</td>
</tr>
<tr>
<td></td>
<td>Fávaro et al., 2010; Rahman et al., 2011; Bilal et al., 2014b; Bilal et,</td>
</tr>
<tr>
<td></td>
<td>2014c; Bilal et al., 2014a; Yang et al., 2007a; He et al., 2011; Yussuf</td>
</tr>
<tr>
<td></td>
<td>et al., 2010, de Carvalho et al., 2011; Nourbakhsh et al., 2014</td>
</tr>
<tr>
<td>Compression molding</td>
<td>Chand et al., 2010; Premalal et al., 2002; Rosa et al., 2009b; Zhao et,</td>
</tr>
<tr>
<td></td>
<td>2009; Syafri et al., 2011; Rosa et al., 2009a; Santiagoo et al., 2011</td>
</tr>
<tr>
<td>Extrusion</td>
<td>Aminullah et al., 2010; Wang et al., 2014; Panthapulakkal et al., 2005a;</td>
</tr>
<tr>
<td></td>
<td>Panthapulakkal et al., 2005b</td>
</tr>
<tr>
<td>Hot press</td>
<td>Ghofrani et al., 2012; Rozman et al., 2000; Rozman et al., 2005a;</td>
</tr>
<tr>
<td></td>
<td>Rozman et al., 2003; Khalif &amp; Ward, 2010; Najafi &amp; Khademi-Eslam,</td>
</tr>
<tr>
<td></td>
<td>2011; Nordyana et al., 2013; Zuhaira et al., 2013; Zuhina et al., 2004;</td>
</tr>
<tr>
<td></td>
<td>El Sayed et al., 2012; Ndazi et al., 2007; Rozman et al., 2005b; Bakar</td>
</tr>
<tr>
<td></td>
<td>&amp; Muhammed, 2011</td>
</tr>
<tr>
<td>Others</td>
<td>Arora et al., 2012; Sheriff et al., 2009; Ahmad et al., 2007; Hua et,</td>
</tr>
<tr>
<td></td>
<td>2011; Sharma &amp; Chand, 2013</td>
</tr>
</tbody>
</table>

Premalal et al., 2002; Razavi-Nouri et al., 2006; Rosa et al., 2009a,b; Czél & Kanyok, 2007; Santiagoo et al., 2011; El Sayed et al., 2012; Aminullah et al., 2010; Yang et al., 2007a; He et al., 2011.

Apart from PE and PP, phenol formaldehyde (PF) (Bhatnagar, 1994; Ndazi et al., 2007), polyurethane (PU) (Sheriff et al., 2009; Rozman et al., 2003), polyester (Rozman et al., 2005a; Ahmad et al., 2007, Rozman et al., 2005b), polymer lactic acid (PLA) (Yussuf et al., 2010, Hua et al., 2011), polyvinylchloride (PVC) (Chand et al., 2010), polyvinyl alcohol (PVA) (Arora et al., 2012), polystyrene (Rozman et al., 2000), urea formaldehyde (UF) (Bakar & Muhammed, 2011), and epoxy (Ibrahim & Kuek, 2011) have also been used to manufacture composites with RH.

Injection molding, compression molding, extrusion, and hot press are the most commonly used techniques to manufacture RH-reinforced composite materials. The manufacturing of composites with different manufacturing processes using RH is shown in Table 1.4.

### 1.3.2 Effects of RH Loading on the Properties of RH Composites

RH has been used with different percentages for the manufacturing of composites, as shown in Table 1.5. The effect of RH loading on mechanical, physical, and functional properties has been widely investigated.