



US010280294B2

(12) **United States Patent**  
**Gardner et al.**

(10) **Patent No.:** **US 10,280,294 B2**  
(45) **Date of Patent:** **May 7, 2019**

(54) **NANOFIBRIL-POLYMER COMPOSITES**

*C08J 2323/12* (2013.01); *C08J 2451/06*  
(2013.01); *C08L 2205/16* (2013.01); *C08L*  
*2310/00* (2013.01)

(71) Applicant: **University of Maine System Board of Trustees**, Bangor, ME (US)

(58) **Field of Classification Search**

CPC .. *C08L 23/10*; *C08L 1/00*; *C08L 51/06*; *C08L*  
*1/04*; *C08L 23/12*; *C08L 2205/16*; *C08L*  
*2310/00*; *B29K 2201/00*; *B29K 2073/00*;  
*B29K 2023/14*; *B29K 2023/08*; *B29C*  
*45/0001*; *B29C 43/003*; *B29C 64/165*;  
*B33Y 10/00*; *B33Y 70/00*; *C08J 3/226*;  
*C08J 5/005*; *C08J 2323/12*; *C08J 2451/06*

(72) Inventors: **Douglas J. Gardner**, Brewer, ME (US); **Yusoo Han**, Bangor, ME (US); **Yucheng Peng**, Evansville, IN (US)

USPC ..... 524/35  
See application file for complete search history.

(73) Assignee: **University of Maine System Board of Trustees**, Bangor, ME (US)

(\* ) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 361 days.

(21) Appl. No.: **14/905,349**

(56) **References Cited**

U.S. PATENT DOCUMENTS

(22) PCT Filed: **Jul. 17, 2014**

8,372,320 B2 2/2013 Gardner et al.

(86) PCT No.: **PCT/US2014/047100**

§ 371 (c)(1),

(2) Date: **Jan. 15, 2016**

OTHER PUBLICATIONS

(87) PCT Pub. No.: **WO2015/009972**

PCT Pub. Date: **Jan. 22, 2016**

Bahar, E. et al, Thermal and Mechanical Properties of Polypropylene Nanocomposite Materials Reinforced with Cellulose Nano Whiskers, *Journal of Applied Polymer Science*, 125(4): 2882-2889 (2012).

Gauthier, R. et al., Interfaces in polyolefin/cellulosic fiber composites: Chemical coupling, morphology, correlation with adhesion and aging in moisture, *Polymer Composites*, 19(3):287-300 (1998).

Gong, G. et al, Tensile behavior, morphology and viscoelastic analysis of cellulose nanofiber-reinforced (CNF) polyvinyl acetate (PVAc), *Composites Part A: Applied Science and Manufacturing*, 42(9): 1275-1282 (2011).

(65) **Prior Publication Data**

US 2016/0152811 A1 Jun. 2, 2016

(Continued)

**Related U.S. Application Data**

(60) Provisional application No. 61/847,751, filed on Jul. 18, 2013.

*Primary Examiner* — Angela C Scott

(74) *Attorney, Agent, or Firm* — Choate Hall & Stewart, LLP; Brian E. Reese; Meaghan E. Bychowski

(51) **Int. Cl.**

*C08J 3/22* (2006.01)  
*C08J 5/00* (2006.01)  
*C08L 1/00* (2006.01)  
*C08L 1/04* (2006.01)  
*B29C 43/00* (2006.01)  
*B29C 45/00* (2006.01)  
*B29K 23/00* (2006.01)  
*B29K 73/00* (2006.01)  
*B33Y 10/00* (2015.01)  
*B33Y 70/00* (2015.01)  
*C08L 23/10* (2006.01)  
*C08L 23/12* (2006.01)  
*C08L 51/06* (2006.01)  
*B29C 64/165* (2017.01)  
*B29K 201/00* (2006.01)

(57) **ABSTRACT**

The present invention provides, among other things, compositions comprising nanofibrils, at least one maleic-anhydride (MA) copolymer and at least one matrix polymer, and methods of making such compositions. The provided methods and compositions allow for the production of composites with unexpectedly superior properties including improved impact resistance, tensile modulus of elasticity, tensile strength, and flexural modulus of elasticity as compared to previously known composites. In some embodiments, the present invention provides methods including the steps of providing cellulose nanofibrils, associating the cellulose nanofibrils with a maleic-anhydride (MA) copolymer to form a nanofibril-MA copolymer blend, preparing the nanofibril-MA copolymer blend for addition to a matrix polymer, and forming a composite by associating the nanofibril-MA copolymer blend with the matrix polymer, wherein the amount of cellulose nanofibrils in the composite is between 3% and 50% by weight of the composite.

(52) **U.S. Cl.**

CPC ..... *C08L 23/12* (2013.01); *B29C 43/003* (2013.01); *B29C 45/0001* (2013.01); *B29C 64/165* (2017.08); *C08J 3/226* (2013.01); *C08J 5/005* (2013.01); *C08L 1/00* (2013.01); *C08L 1/04* (2013.01); *C08L 23/10* (2013.01); *C08L 51/06* (2013.01); *B29K 2023/08* (2013.01); *B29K 2023/14* (2013.01); *B29K 2073/00* (2013.01); *B29K 2201/00* (2013.01); *B33Y 10/00* (2014.12); *B33Y 70/00* (2014.12);