Composites Science and Technology 107 (2015) 169–176

Contents lists available at ScienceDirect

Composites Science and Technology

journal homepage: www.elsevier.com/locate/compscitech

Experiments and FE simulation for twin screw mixing of nanocomposite of polypropylene/multi-walled carbon nanotubes

H. Djoudi [1], J.-C. Gelin [2], T. Barrière *

FEMTO-ST Institute, Applied Mechanic Laboratory, 24 rue de l'épitaphe, Besançon, France

ARTICLE INFO

Article history:
Received 1 November 2014
Received in revised form 22 December 2014
Accepted 28 December 2014
Available online 2 January 2015

Keywords:
A. Carbon nanotubes
Particle-reinforced composites
B. Non-linear behavior
C. Modeling
Computational mechanics

ABSTRACT

Elaboration of the feedstock in polymer/carbon nanotubes is introduced in both aspects of the experiments by a twin screw mixer, and the corresponded simulation of mixing process. The viscous behaviors are calibrated by a capillary rheometer under different temperature. The achieved data are used to determine the parameters in the suitable viscous laws with the satisfied verification. Front cover of the mixer is replaced by a transparent one, which permits the direct observation of mixing process, and the record of temperature evolution by an infrared camera with high performance IR imaging. The simulations of mixing processes in a twin screw mixer, with the determined viscous law and the temperature conditions justified by experiments, are realized by software COMSOL© with its performance in coupling of the multi-physics. Different referential frames and ALE formulation are used for the counter rotations of two screws in mixing chamber. The mechanism of nanocomposite mixing in twin screw chamber is investigated with the achieved results. Comparison between the results of simulation and experiments proved the proposed methods and the prepared feedstock for nanocomposite.

1. Introduction

The development and improvement of nanocomposites will be a key advance in the future. Since the discovery of carbon nanotubes in 1991 by Iijima [1], the research in nanomaterial became one of the most promising fields. Carbon nanotubes (CNTs) are allotropes of carbon with cylindrical nanostructure. Their diameters range from one to ten nanometers, and lengths in the micrometer range as reported by Pötschke [2]. The analyses and experiments revealed that carbon nanotubes have excellent mechanical properties, with Young's modulus of individual CNT in TPa order and a tensile strength as high as 200 GPa as reported [3–5]. The measurement on multi-walled carbon nanotube (MWCNT) by Demczyk et al. [6] estimated that the elastic modulus is about 0.8 TPa with error eq. 1.14%. Due to the super mechanical properties and large potentials, important researches have been carried to develop various nanocomposites of polymer/carbon nanotubes by injection molding process. Shaffer et al. [7] set up the first reinforcement of nanotubes in polymeric based composites in 1999. Prashanthaa et al. assessed the rheological and mechanical properties of polypropylene/MWCNT composites [8]. Lee et al. investigated their rheological and electrical properties [9].

The application of carbon nanotube based nanocomposites is not yet exhibited in practice, because of the uncertain load transfer between the matrix and the fibers. The homogeneous particles distribution and proper interfacial adhesion are crucial for successful preparation of the nanocomposites, as shown by Zhan et al. [10]. Thiébaud et al. [11] reported that the shear mixing by melt processing is currently one of the most used methods. They realized the simulation of mixing process in a twin screw mixer restricted in 2D model. Actually most studies are related to experiments, this work is aimed to provide a numerical counterpart. 3D Simulation of loaded polymer flows in a twin screw-mixer has been realized by finite elements method.

The elaboration of PP/MWCNT nanocomposites by a twin screw mixer is first introduced. Rheological characterization of PP/MWCNT elaborated samples at 4 selected temperatures (180, 200, 220 and 240 °C) has been conducted by a capillary rheometer, as reported [12]. These rheological properties are used to identify the viscosity parameters of flow models. Cross et al. [13] and Bourgeat et al. [14] suggested the way to fit the flow properties of polymers melt or a polymer solution by non-Newtonian flow model, i.e. to modify Newton's model by a variable viscosity that depends on the share rate and temperature. In order to characterize the flow of polymer/carbon nanotubes, the Carreau–Yasuda law has been adopted.

* Corresponding author. Tel.: +33 3 81 66 60 75; fax: +33 3 81 66 67 00.
E-mail addresses: hamza.djoudi@hotmail.fr (H. Djoudi), jean-claude.gelin@univ-fcomte.fr (J.-C. Gelin), thierry.barriere@univ-fcomte.fr (T. Barrière).
[1] Tel.: +33 3 81 66 60 10.
[2] Tel.: +33 3 81 66 60 35.

http://dx.doi.org/10.1016/j.compscitech.2014.12.017
0266-3538/

The second part involves the analysis and simulation of loaded polymer flows in mixing process by finite element method. COMSOL© multiphysics modeling software was used because of its efficiency for coupling of the multiphysics, the availability to add solution of the partial differential equations, and the convenience to cooperate with Matlab®. Evolution of the velocity fields, related shear rates, temperature fields and mixing torques during the mixing stage have been analyzed.

2. Experimental process

2.1. Elaboration of the nanocomposites

Nanocomposites of polypropylene (PP)/multi walled carbon nanotubes (MWCNTs) have been elaborated by mixing PP pellets [PP_Sabic® PHC 31_81, melt flow index 12.6 kg/10 min at 200 °C] with the masterbatch MWCNT produced by Chemical Vapor Deposition [NC7000 supplied by Nanocyl©, Belgium]. In order to properly disperse MWCNT in the polypropylene matrix, the chlorosulfonic acid and ultrasonication were used to get the carbon nanotubes dispersed properly. The acid adds positive charges to the surface of the nanotubes without damaging them, and causes the nanotubes to spontaneously separate from each other in their natural bundled form.

The mixing process was realized in a co-rotating twin screw mixer [Plastograph 50 W EHT] shown in Fig. 1. This equipment represents the upper capacity for velocity 150 rpm, temperature 500 °C, torque 200 N m, and volume 40 cm^3. The Choice of optimal mixing condition follows the research realized in laboratory [15]. Duration of the mixing is 100 min, with a chamber temperature 200 °C, and a screw speed 60 rpm. The speed ratio between two screws is about 2:3. The left screw rotates clockwise while the right one rotates counterclockwise. Counter-rotation at different speeds provides proper compounding and mixing of the nanocomposites. Evolutions of the mixing torque and temperature have been measured by sensors in real time. A transparent front wall is used for the twin screw mixer in experiments. Evolution of the temperature in cavity of the twin-screw mixer can be measured by an infrared camera of high thermal imaging quality, see Fig. 1.

The thermoplastic polymer matrix is polypropylene (Sabic PHC 31_81), with melting temperature 152 °C, density 0.905 g cm^{-3}, specific heat 1800 J/kg K and thermal conductivity 0.2 W/m K. The reinforcement by MWCNT (NC 7000) is Nanocyl® 7000, with diameter 9.5 nm, length 1.5 μm, thermal conductivity 3000 W/m K, purity 90%.

4 nanocomposite samples of different MWCNT loadings in polypropylene matrix, 10%, 20%, 30% and 40% in weight, have been elaborated. For a reference, pure PP matrix has been squeezed with the same mixing parameters. Main challenge in elaboration of the nanocomposites is to get uniform and homogenous dispersion of the MWCNT in PP matrix. The mixing torque depends on shear viscosity of the loaded polymers as reported by Kong et al. [16]. Evolutions of the mixing torque for PP/MWCNT samples are measured, and compared later with the simulation results in Fig. 10. When PP/MWCNT compounds are fed into the mixer, very high torque values are measured due to their solid state. After 10 min, the PP gets into molten state then it results in decrease of the mixing torque. More MWCNTs amount in mixture results in larger mixing torque, because of the effect of long carbon nanotubes and the formation of MWNT network in polymer matrix.

2.2. Rheological characterization of PP/MWCNT nanocomposites

Rheological tests are the essential ways to identify a suitable viscosity model. The shear viscosity of PP/MWCNT nanocomposite has been measured for different MWCNT ratios, at different temperatures and different shear rates. A capillary rheometer provided by Malvern©, with a capillary die of diameter 1 mm and length 16 mm, has been used in the range of shear rate 10–10^4 s^{-1}. The relationship between shear viscosity and shear rate for PP/MWCNT samples are obtained, and then used for calibration of the behavior model as shown in Fig. 5. The effect of temperature on shear viscosity is shown in Fig. 2.

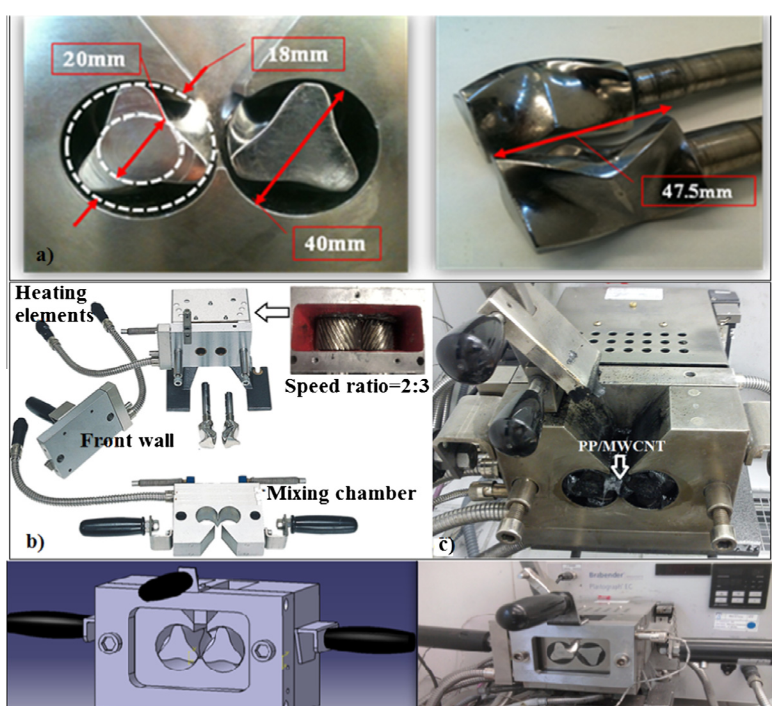

Fig. 1. Twin screw mixer Brabender® EC W50EHT: (a) mixing chamber and screws, (b) installation of the twin-screw mixer, (c) mixing process of the PP/MWCNT feedstock and (d) replacement of the front wall by a transparent one.