

論文 / 著書情報
Article / Book Information

題目(和文)	メルトブロー過程によるナノ繊維不織布の製造
Title(English)	Melt blowing process for production of non-woven nano-fiber web
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出典(和文)	学位:博士(学術), 学位授与機関:東京工業大学, 報告番号:甲第10247号, 授与年月日:2016年3月26日, 学位の種別:課程博士, 審査員:鞠谷 雄士,扇澤 敏明,浅井 茂雄,塩谷 正俊,松本 英俊
Citation(English)	Degree:Doctor (Academic), Conferring organization: Tokyo Institute of Technology, Report number:甲第10247号, Conferred date:2016/3/26, Degree Type:Course doctor, Examiner:,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	要約
Type(English)	Outline

Thesis Outline

The following list describes the outline of this thesis;

Chapter 1 - “Introduction”

In this chapter, melt blowing process, a single step process for producing non-woven fabrics, is introduced. It is explained that the advantage of this process is the capability of producing fiber web from various types of polymer with high production rate, and therefore the technology is widely applied in the industrial field. It is also pointed out that further improvement of the melt blowing process to reduce fiber diameter to nanometer range attracts much attention these days because such improvement introduces some favorable properties to the web for applications such as filtration of fine particles. Accordingly, the objective of this research was set to develop technology for producing non-woven web of nano-fibers as well as to investigate the fiber formation behavior in the melt blowing process. Polymethylpentene (PMP) and polypropylene (PP) of high melt flow rate (MFR) were selected as the starting materials considering the different characteristics of the two polymers such as glass transition temperature and crystallization behavior.

Chapter 2 - “Fundamental Fiber Formation Behavior of Polymethylpentene and Polypropylene in Melt Blowing Process”

The fundamental of melt blowing process was investigated by study the effect of the spinning conditions in the melt blowing process, i.e. air flow rate, throughput rate and die-to-collector distance (DCD), on the fiber diameter and the structure of fibers comparing the results for PP and PMP. PP fibers exhibited finer diameters than PMP fibers under similar spinning conditions because of the lower viscosity of PP. Fiber diameter decreased with the decrease of throughput rate and increase of air flow rate in the cases of both PP and PMP. The two polymers showed different behavior for the change of DCD. In case of PP, fiber diameter increased when the DCD was increased, whereas fiber diameter was not affected by DCD in case of PMP. Structure analyses of

PP and PMP fibers were carried out in order to elucidate the origin of such difference between the two polymers. Through the wide-angle X-ray diffraction (WAXD) analysis, crystallized structure with no preferred orientation was observed for PMP fibers even though the glass transition temperature of PMP is higher than the room temperature, whereas mesomorphic structure was observed for PP. These results suggested that the PMP fiber was crystallized and solidified in the spinning process. On the other hand, PP fiber was still in a molten state when the fibers reached the collector. In other words, by the change of DCD thinning behavior of PMP was not affected whereas that of PP was affected. It was concluded that such difference in the structure formation behavior is the origin for the different effect of DCD on fiber diameter for PP and PMP.

Chapter 3 - "Fine Filament Formation Behavior of Polymethylpentene and Polypropylene near Spinneret in Melt Blowing Process"

The formation behavior of fine fibers of PMP and PP in the melt blowing process was investigated through the observation of the spin-line in the vicinity of the spinning nozzle using a high speed camera. Reduction of the throughput rate of polymer and increase of the air flow rate were necessary to achieve the fine diameter fibers, however these conditions generally cause the instability in the spinning process. Observation of the spin-line at the melt blowing die revealed the periodic accumulation of polymer flow near the spinning nozzle followed by the quick pulling down of the accumulated polymer by the air flow. This behavior caused the periodic fluctuation of fiber diameter as well as the intermittent breakage of the spin-line under extreme conditions. Because of higher extrusion viscosity, PMP showed more stable spinning behavior than PP. Frequency of diameter fluctuation became higher with the increases of air flow rate and throughput rate, and the maximum frequency of about 60 Hz was observed for spinning of PP with the throughput rate of 18 mg/min/hole. Diameter distribution of the fibers in the prepared web was also analyzed to compare with the spinning behavior. Fiber diameter distributions were narrow and symmetric under stable spinning conditions, whereas skewed diameter profiles with a maximum at a low diameter value and a long tail to the larger diameter region were observed under

unstable conditions. Intermittent spin-line breakage caused flaws of “shot” and/or “fly”, and the skewed fiber diameter distribution with the presence of a small number of fibers of extremely large diameter.

Chapter 4 - “Effect of the Reduction of Nozzle Diameter for Production of Fine Filaments in Melt Blowing Process”

In order to improve the process for better production of fine fibers, the effect of the utilization of the spinning nozzle of extremely small diameter was investigated. From the view point of spin-line dynamics, reduction of the nozzle diameter leads to the reduction of the final fiber diameter under the steady-state condition as well as the stabilization of the spinning process with periodic fluctuation of fiber diameter under the high draw-down conditions. When nozzle diameter was reduced from 0.12 to 0.06 mm, average fiber diameter of around 1 μm or below was achieved. Influence of the variation of spinning conditions on the fiber diameter was similar to those obtained using the nozzles of larger diameter. In terms of fiber structure development, the results from differential scanning calorimetry (DSC) and WAXD analyses for PMP exhibited that the structure of fibers prepared under various spinning conditions showed insignificant difference except for the influence of nozzle size. When nozzle size of 0.06 mm was used, WAXD result exhibited crystalline structure with orientation of c-axis along the fiber length whereas the fibers prepared using large diameter nozzle only presented crystalline structure with no preferred orientation. In case of PP, almost all fibers prepared under various spinning conditions exhibited a mesomorphic structure whereas only the fiber prepared with small DCD conditions exhibited the development of α -form crystals. It was considered that this result is due to the higher temperature of the web on the collector. Increase of web temperature with the reduction of DCD was confirmed through the temperature measurement of web. Considering the positive effect of the reduction of nozzle diameter, use of the nozzle of extremely small diameter of 0.03 mm was attempted. Through the observation of fiber formation behavior in the vicinity of spinning head using a high speed camera, stabilization of the overall spinning process was confirmed when using smaller diameter nozzles for both polymers.

In addition, reduction of mean and minimum diameters were confirmed for both PP and PMP even though CV% was increased for PP.

Chapter 5 - "Modification of Melt Blowing Process for Production of Fine Filaments"

In this chapter, the modification of the process to further decrease fiber diameters was presented by applying two external methods. One is the application of electric field to reduce the effect of surface tension of fiber. The other one is the addition of low molecular weight wax to the polymer for reduction of melt viscosity of polymer. In case of electric field, the results were different from expectation that fiber diameters were only slightly reduced when using polymer throughput rate of 30 and 54 mg/min/hole even after increasing the voltage to 45 or 30 kV, respectively. Considering that the electric charge on the fiber surface acts not only for the generation of elongational force to the spinning line, but also for the stabilization of the process through the reduction of surface tension which causes the fiber breakage into droplet, effect of applying voltage of 45 kV on fiber diameter distribution was investigated under the combined conditions of the use of a spinning nozzle of a small diameter of 0.03 mm and adopting the extremely low throughput rate of 9 mg/min/hole. Even though there was only a slight effect of electric field for the reduction of mean fiber diameter, minimum fiber diameter was reduced from 120 nm to 60 nm indicating that the electric charge is effective exclusively for the fibers of smaller diameter.

Effect of the blending of PP wax to PP on the spinning behavior and the diameter of produced fibers were investigated. It was found that the blending of the PP wax of up to 50 wt% caused the significant reductions of both mean and minimum diameters. At the throughput rate of 15 mg/min/hole, the mean fiber diameter of less than 500 nm and the minimum fiber diameter of 60 nm were achieved. It was also noticed that the reduction of nozzle diameter did not affect the fiber diameter. Observation of the spinning process in the vicinity of spinning nozzle revealed that there was a formation of polymer pool on the surface of the spinneret. This thread was continuously formed from this polymer pool with gradual increase of the volume of

polymer melt. The accumulated polymer tended to form spherical shape and intermittently fell down as a droplet. Therefore, a certain amount of spherical drops of small volume could be observed on the fine fiber web when blending PP with wax.

Chapter 6 - “Effect of the Interaction of Two Components on Fiber Formation in Bi-component Melt Blowing Process”

Bi-component melt blowing process was applied for the production of web consisting of bi-component fibers of PP and polylactic acid (PLA) with side-by-side cross-sectional configuration. Changes in the spinning conditions on the characteristics of resultant fibers exhibited similar trend in comparison with those for ordinary single-component melt blowing process. If the fibers obtained through the single- and bi-component melt blowing process of similar spinning conditions were compared, the fiber diameter of single-component PP was smaller than that of single-component PLA, whereas the PP/PLA fibers exhibited similar or even lower diameters in comparison with those for PP. It was speculated that the different thermal properties of PP and PLA such as glass transition temperature and crystallizability are the origin of such unusual behavior through the variations of thinning profiles and solidification behavior. The diameter distribution profiles of bi-component fibers also exhibited narrower range of distribution. These results indicated that bi-component fibers were more uniform than single-component fibers. Consequently, the better quality of fiber web for filter application was confirmed from the results of air permeability measurement. Web of bi-component fibers also exhibited an improvement of thermal characteristics with lower percentage of shrinkage in comparison with the web of single-component PLA which exhibits high percentage of shrinkage. This indicates the better coefficient of the web when using bi-component side by side melt blown.

Chapter 7 - “Concluding Remarks”

This final chapter of this thesis describes the summary results obtained through this research and the future prospects in this field are discussed.