



The comparative studies of charge storage stabilities among three PP/porous PTFE/PP electret

L.L. Cui^a, M.H. Song^b, Y.X. Kong^a, L. Cheng^b, D. Wang^a, Y.H. Xiao^a, J. Jiang^{b,*}

^a Department of Inorganic Chemistry, School of Pharmacy, Second Military Medical University, Shanghai 200433, China

^b Department of Physics and Mathematics, Faculty of Basic Medical Sciences, Second Military Medical University, Shanghai 200433, China

ARTICLE INFO

Article history:

Received 11 September 2008

Received in revised form

3 December 2008

Accepted 13 January 2009

Available online 2 February 2009

Keywords:

Electret

Transdermal drug delivery

Charge storage stability

Porous polytetrafluoroethylene

Polypropylene

ABSTRACT

In this paper, PP/porous PTFE/PP electret, pressure sensitive adhesive PP/porous PTFE/PP electret and PP/porous PTFE/PP electret drug transdermal patch were prepared. By means of the surface potential decaying measurement and thermal discharge method, the charged storage stability was compared among them. The results indicated that (1) negative PP/porous PTFE/PP electret could still hold 93.4% of its initial surface potential when it was kept at RT for 16 days and exhibited better charge storage stability than positive electret. (2) The humidity almost had no influence in the charge storage stability of the electret. (3) The positive and negative PP/porous PTFE/PP adhesive electret could respectively hold 87.5% and 75.6% of their initial surface potential when being kept at room temperature for 13 days. The adhesive had a little influence in the charge storage stability of the electret. (4) Both the negative and positive PP/porous PTFE/PP electret drug transdermal patch had excellent charge storage stability.

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1. Introduction

Due to its excellent dielectric properties, outstanding charge storage stability at high temperature and good ventilation and biocompatibility, porous polytetrafluoroethylene (porous PTFE) is widely used in rehabilitation of wound and damaged nerve, study of transdermal drug delivery system (TDD system), and so on [1,2].

TDD system is the delivery of drugs across the skin and into systemic circulation. Because of the barrier function of skin, the number of drugs that can be delivered in this manner is limited. Therefore, some strategies have been investigated and developed, such as the use of chemical enhancers and physical methods that facilitate the diffusion of drugs through the human skin. The physical strategies include iontophoresis, electroporation, ultrasound, and so on [3]. However, with the development of the application of electret in medical science, many researchers engaged in the branches of biomedical engineering, pharmaceuticals and functional materials have focused their studies on the preparation of electret TDD system and the understanding of the enhancement mechanism of the system on drug percutaneous permeation [4].

In order to get a qualified electret TDD system, the charge storage stability of the system under high humidity and/or matrix containing conditions is the most important. As compared with PTFE, porous PTFE has a much larger surface to contact with atmosphere and matrix as well as drug, which makes it easier to lose the surface charge, therefore decreasing the charge storage stability and limiting drug transdermal delivery. However, polypropylene (PP) not only bears the similar dielectric properties and chemical stabilities to PTFE, but also holds particular excellent hydrophobicity and lower ventilation, being fit for use at high humidity circumstance. In addition, polyethylene (PE) can be used as the adhesive of space charge electret and composite electret for its better charge storage stability and lower softening temperature.

In this study, we used PE as the adhesive to prepare PP/porous PTFE/PP composite film. On the basis of this composite film, three kinds of electrets, PP/porous PTFE/PP electret (composite electret), pressure sensitive adhesive (PSA) containing PP/porous PTFE/PP electret (PSA electret) and PP/porous PTFE/PP electret drug transdermal patch (electret transdermal patch), were prepared to study the influence of humidity, matrix and drug in the charge storage stability. All the electrets were prepared by means of heat melting and constant corona charging methods. The thermally stimulated discharge (TSD) and isothermal surface potential were investigated.

* Corresponding author. Tel./fax: +86 21 2507 0282.

E-mail address: jjiang@smmu.edu.cn (J. Jiang).

2. Materials and methods

2.1. Preparation of composite electret

The biaxially oriented porous PTFE (1 ~ 5 μm aperture, 50 μm thickness and 70% porous, Shanghai Plastic Institute, China) was used. The PP (Toray Fan Bo, Japan) thickness was 13 μm . PE is the product sold. All the materials were from the same rolls to keep the same morphometrics.

The bare porous PTFE film was covered on the PE and mono-aluminium-plated (100 nm thickness) PP to prepare the charge storage layer by means of corona charging (CORONATROL, Model 152A, Monroe Electronics Co., USA) for 5 min. The point and grid voltages were ± 8 kV and ± 1 kV, respectively. Then the charged film was covered by PE and bare PP, pressed and heat melted at 120 $^{\circ}\text{C}$ to get the PP/porous PTFE/PP composite electret after cooling to a constant temperature. The equivalent surface potential of the charged film and composite electret film was measured by a compensation method with the application of surface potentiometer (SD8303, Shanghai Electromotive Facility Institute, China). The open TSD current spectrum was measured from room temperature (RT) to 300 $^{\circ}\text{C}$ in a rate of 3 $^{\circ}\text{C}/\text{min}$ with the plated side of the sample attached to a lower electrode and bare side to an upper electrode in a program controlled stove (Heraeus, T5042K, Germany).

2.2. Preparation of PSA electret

After been dissolved in acetone, Eudragit E100 (Röh, Germany), a pressure sensitive adhesive, was added to tributyl citrate to form homogeneous solution. Then the solution was coated onto the bare side of the PP/porous PTFE/PP electret film with an area of 8×16 cm^2 . After removing of the residual solvent, PSA electret was prepared.

2.3. Preparation of electret transdermal patch

Meloxicam was selected as a model drug and was added to the mixtures of acetone solution of Eudragit E100 and tributyl citrate to form a homogeneous solution. Then the same method as PSA electret preparation was used to obtain the electret transdermal patch.

3. Results and discussion

3.1. Charge storage and decay of composite electret

Due to the surface defects caused by chemical impurity and oxides, or chain broken and the difference of short-range order between surface and body, there exists charge capture center (trap) on the polymer surface of 0 ~ 1 μm that is called the surface trap. The different structure of the materials, such as defects of monomer, deformation of crystal and irregular molecular chain accounts for the charge trap center in the bulk [5]. The electret field of corona charging leads to air ionization (paschen). Because energy of ion is only about 2 ~ 3 eV, large amounts of ions (or charge carriers) are trapped by surface traps and shallow traps to form space charge electrets with basically no dipole effect [6]. Charges are located between the energy traps of the band gap of conduction band and valence band of the material.

As it has been known, thermally stimulated discharge (TSD) could provide a lot of information on the thermal stability of space charges in electret [7,8] and the peaks of TSD at higher temperature corresponds to the surface and bulk traps [9]. PTFE, PP and PE are

space charge electret polymers that could store space charge near the surface or in the bulk for long periods of time [10].

Fig. 1 is the open TSD current profiles of negative composite electret, negative porous PTFE electret and negative PP electret corona charged at RT respectively. It indicated that two space charge peaks appeared around 180 $^{\circ}\text{C}$ and 280 $^{\circ}\text{C}$ respectively for composite electret. The larger peak area at 180 $^{\circ}\text{C}$ than that at 280 $^{\circ}\text{C}$ indicates that the abundance of charge carriers were captured at surface traps, whereas only a small part of carriers were captured at deep traps. The results were further supported by the larger peak area at 270 $^{\circ}\text{C}$ for porous PTFE electret than the peak area at 280 $^{\circ}\text{C}$ for composite electret in Fig. 1. Because the injected ion energy was only 2 ~ 3 eV stimulated by corona field during the RT and constant voltage corona charging process, charges were deposited at the surface or shallow traps of the sample. Since porous PTFE possesses open through hole structure and forms larger surface and much more defects when being stretched at high temperature, part of the charges could inject into the surface and each bulk energy trap of porous PTFE as well as the surface trap of PE via the open holes during the corona charging process. Besides, the heat melting process (or heat ageing) at 120 $^{\circ}\text{C}$ led to the filling up of the melted PE into some holes of the porous PTFE at the surface or in the bulk which resulted in the structure change of porous PTFE, and changed the ratio of energy trap. On the other hand, heat stimulation made the detrapping of charge carriers captured in the shallow traps of porous PTFE and in the surface traps of PE. Detrapped charges might be recaptured by deep traps of porous PTFE or PE during the directional migration under the action of internal electric field of composite electret. Therefore, a space charge peak formed at 180 $^{\circ}\text{C}$ for composite electret caused by thermal stimulating of captured charge during TSD process.

Comparing the TSD profile of composite electret with that of PP electret in Fig. 1, it also indicated that composite electret had larger peak height at 180 $^{\circ}\text{C}$ than PP electret had at 146 $^{\circ}\text{C}$, suggesting that more charge carriers were captured at each trap of porous PTFE with the energy level higher than PP. Therefore, heat melting effectively changed the ratio of deep trap to shallow trap in composite electret, which was good for improving the charge storage stability of composite electret. It further indicated in Fig. 1 that composite electret had higher charge storage density as compared with porous PTFE. Therefore, the preparation technics for composite electret could be a good help to obtain the electret of higher charge density.

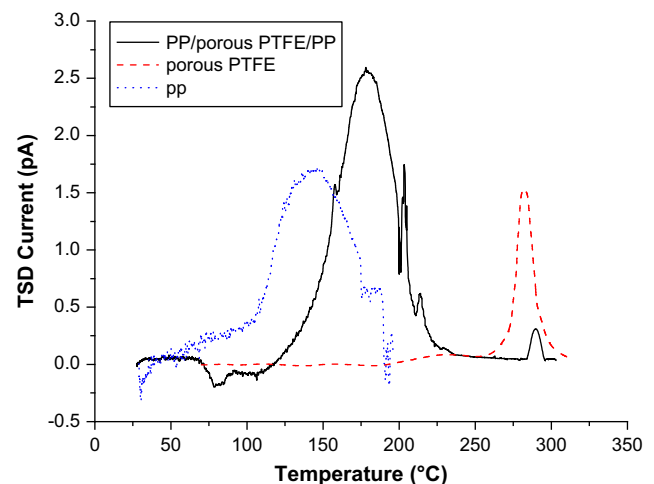


Fig. 1. Open TSD current spectrum of the electret.

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