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Original Article

Effect of Polymer Grade and Plasticizer Molecular Weights on Viscoelastic Behavior of Coating Solutions

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Abstract

Film coating solutions containing different grades of HPMC (E5, E15 and E50) with and without polyethylene glycol (with various molecular weights) were examined by an oscillatory method, using Haake CV 100 rheometer. Fundamental rheological parameters (η, G´ and G″) were measured over the frequency range from 0.04 to 2 Hz. The variation with frequency of the loss tangent (a ratio of viscous to elastic contribution) provides a useful parameter for the characterization of pharmaceutical products. Magnitudes of the loss tangent are discussed with relation to coating solutions made by different grades of polymers and plasticizers.

Keywords: Viscoelastic properties; Rheological parameters; Oscillatory method; Film coating.

Introduction

Aqueous film coating techniques are of current interest in pharmaceutical industry. This technology has precedents in both paints and adhesives technology. It is a field of applied science such as polymer, surface, mechanics, and rheological sciences. The quality of coating depends on film coating materials. Therefore, much effort have been devoted to study the solubility, permeability, mechanical and rheological properties of films, made of different film coating materials. Studies on pharmaceutical film coatings have often examined mechanical properties of free films prepared by casting or spraying techniques. The rheological properties of coating solutions are important in film coating process because of their effects on spraying, atomization, spreading, and penetration stage (4). Aulton and coworkers have studied the elastic, plastic and viscoelastic properties of HPMC films by indentation method (1). The effects of film preparation methods (cast and sprayed films) were studied by Obara and coworkers (2). The water vapor transmission and mechanical properties (puncture strength and % elongation) of the films were investigated as a function of the polymer type and viscosity, plasticizer type and concentration (3).

The purpose of this investigation was to examine the effects of polymer grades and plasticizer molecular weight on viscoelastic behavior of coating solutions.

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Experimental

Materials

Methocel E5, E15, E50, (HPMC) were all purchased from Colorcon, Ltd. Polyethylene glycol (PEG) 300, 600, 1500, 4000 were purchased from Merck &Co., Inc. PEG400 was purchased from Sigma Chemical Co.

Method

The desired amount of different grades of HPMC, with and without PEG were weighed and mixed with approximately half of the calculated hot distilled water, and the solutions were left overnight.

Instrumentation

Haake C V 100 rheometer and sensor system Z B 15, were used at four temperatures (30, 40, 50, and 60°C) and six frequencies (0.04, 0.08, 0.2, 0.4, 1, and 2 Hz).

Calculated viscoelastic parameters

Values for dynamic viscosity (η), storage modulus (G') and loss modulus (G'') were calculated from the raw data. A useful dimensionless parameter, tan.δ (loss tangent) was also calculated. It is a measure of the ratio of energy loss to energy stored in a cyclic deformation (G''/G'). This provides a practically interesting parameter that merges both the elastic and viscous contributions.

Results and Discussion

Effect of HPMC grades

Loss tangent of different grades of HPMC (E5, E15 and E50) was plotted against ω. These results show that the loss tangent increases to ω = 6.25 (viscous properties) and then decreases at high frequency for HPMC E50. HPMC E5 shows that loss tangent decreases in this frequency apparently due to its less viscosity in all temperatures except 60°C (Figure 1). This temperature is more than HPMC thermal gelling point (=52°C), therefore precipitation occurs and the system shows higher viscosity and higher loss tangent. The difference between behavior of 15% (w/v) solutions of E5 and E15 is less than that can be observed in E5 and E50 solutions, because of relatively equal molecular weights (Figure 2). By using a mechanical model made up from a combination of spring (elastic elements) and dashpots (viscous elements), the behavior of coating solutions under oscillation could be best understood. At high frequency, the springs can elongate and contract under imposed shear but the dashpots have very little time to move (5). The system, therefore, behaves essentially as an elastic solid of modulus G. At low frequency, the springs can also extend but in this case the dashpots have ample time to move and their extension greatly exceed that for the springs. The system therefore behaves essentially as a viscous fluid of viscosity η.

Effect of HPMC concentration

According to the rheological data and closeness to real condition in film coating process, T = 40°C, ω = 6.25 and f = 1 Hz were chosen for investigation of HPMC concentration and plasticizer molecular weights on loss tangent. The results showed that loss tangent increases as the frequency increased in all cases when the polymer concentration changed from 10% to 20% w/v. A loss tangent increase of 0.004278, 0.006923 and 0.009028 were found for 10, 15, 20% w/v HPMC E5 solutions, respectively. This could be related to more entanglement point of polymer solution network as the polymer concentration increased. Therefore, polymer solution shows higher storage modulus, loss tangent and viscous properties.

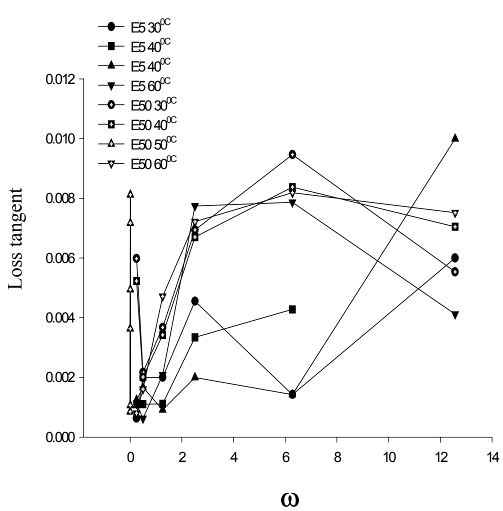

Figure 1. Loss tangent against ω for HPMC E5 and E15 15% solutions at different temperatures (30, 40, 50, and 60°C).

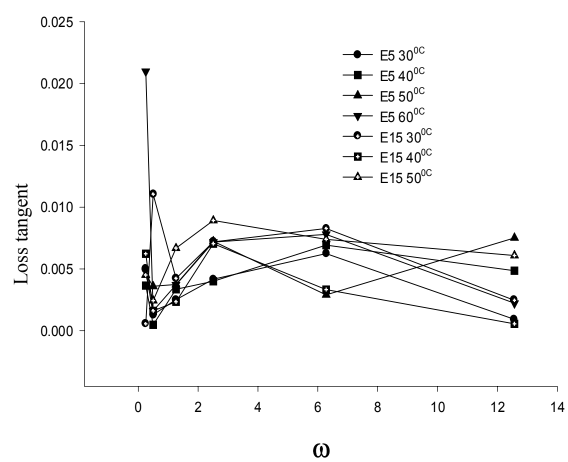

Figure 2. Loss tangent against ω for HPMC E5 and E50 at different temperatures (30, 40, 50 and 60°C).

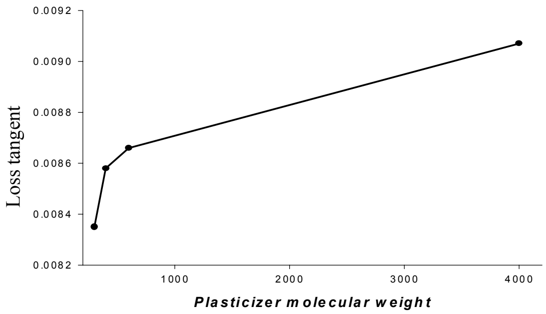

Figure 3. The effect of plasticizer (10% w/w) molecular weights on loss tangent.

Effect of plasticizer molecular weight

Plotting of loss tangent against different plasticizer molecular weights in previous condition showed an increase in the loss tangent as the plasticizer molecular weight increased (Figure 3). As we have already reported; there is a more profound decrease in both mechanical and thermomechanical properties of films by using plasticizers with lower molecular weights (6). According to gel theory, active center forces joint polymer molecules to each other in solution. These bonds are in a dynamic equilibrium in solution form. Water molecules and plasticizers are in competition for the same sites and reduce the number of active centers and the number of polymer-polymer contacts, thus they decrease the rigidity of the three dimensional structure formed on drying and change mechanical and thermomechanical properties of film. In the solution, stage viscosity plays the main role and plasticizers with higher molecular weights increase the solution viscosity and as expected decrease modulus and loss tangent.

The interrelationship between G and η is the relaxation time τ (= η/G). If $1/\omega \gg \tau$, the dashpots can keep-up with the oscillation and the energy stored in each cycle is almost completely dissipated. But if $1/\omega \ll \tau$, almost no energy is dissipated. It is important to know how rheological behavior changes as a function of time, in order to optimize the process parameters and the film coating procedure. Such data gives useful predications concerning the changing of rheological properties of coating solutions on the surface of particles during the coating process Plasticizers especially with lower molecular weights show lower loss tangent and more energy loss (G'') and plasticity.

Acknowledgements

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