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Tensile Properties of Free Films Cast from Aqueous Ethylcellulose Dispersions

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Free films of two commercially available formulations of aqueous ethylcellulose dispersion differing only in plasticizer content (Surelease/E-7-7050 without silica and E-7-7060 containing dibutyl sebacate and glyceryl tricaprylate/caprate as plasticizers, respectively) were cast and coalesced at temperatures ranging between 30 and 70°C. Mechanical properties of these films were measured using tensile stress analysis. Three mechanical parameters, namely, tensile strength, work of failure, and elastic modulus, were computed from the load-time profiles of these films. The results showed that the tensile strength and elastic modulus values of the films cast from both formulations increased with the corresponding increase in coalescence temperature up to 60°C, beyond which no significant differences were observed. In the case of work of failure, however, the difference between the two formulations was observed above 60°C. The films cast from Surelease/E-7-7050 formulation without silica (dibutyl sebacate as the plasticizer) were relatively softer than those from Surelease/E-7-7060 formulation (glyceryl tricaprylate/caprate as the plasticizer). At coalescence temperatures above 50°C, the films cast from both formulations exhibited temperature-dependent plastic deformation.

KEY WORDS: ethylcellulose; Surelease; dibutyl sebacate; glyceryl tricaprylate/caprate; cast films; mechanical properties; tensile strength; work of failure; elastic modulus.

INTRODUCTION

Polymeric films are finding an increasing range of applications in pharmaceutical research and dosage form design. Films of water-insoluble polymers (e.g., ethylcellulose) are frequently used in the fabrication of membrane-controlled oral drug delivery systems. Traditionally, water-insoluble polymers for controlled release coatings have been applied as organic solvent-based solutions. However, because of environmental concerns aqueous-based polymeric dispersions have gained popularity for film coating applications. At present, on a commercial scale, the use of aqueous-based controlled-release coating systems has been limited to latexes and pseudolatexes of either methacrylate copolymers or ethylcellulose (1). The mechanisms involved in the formation of a continuous film from an aqueous dispersion are different from those involved in film formation from an organic solution (2). In the case of coating from an aqueous polymeric dispersion, the selection of the process conditions and the choice of a plasticizer and its level have a decisive impact on the characteristics of the film.

Ethylcellulose is widely used as a dissolution rate controlling polymer for the coated controlled release dosage forms (3). At present, there are two commercially available aqueous dispersions of ethylcellulose, namely, Surelease (Colorcon) and Aquacoat (FMC Corp.). Since the glass transition temperature of ethylcellulose is relatively high (T_g = 135°C) (4), it does not form flexible films under normal coating conditions. In order to provide flexibility and elasticity to the polymeric films which are essential for their integrity and durability, aqueous dispersions of ethylcellulose need to be plasticized. In the case of Surelease, the plasticizer is incorporated in the dispersion during the manufacturing process itself, whereas a plasticizer must be added to Aquacoat prior to its application as coating. Two formulations of Surelease are commercially available, one containing dibutyl sebacate and the other containing glyceryl tricaprylate/caprate as the plasticizer. The glass transition temperature, T_g, of both these formulations was observed to be about 35°C when determined using differential scanning calorimetric analysis (5).

The release characteristics of film-coated extended release formulations are strongly dependent on the properties of the film, e.g., film permeability and its mechanical strength (6,7). Traditionally, stress–strain testing in the tensile mode has been a popular and widely used mechanical test for the polymeric films. The tensile test is practical, and analysis of its data is relatively straightforward. The tensile test gives an indication not only of the elasticity and strength, but also of the toughness of the film. However, polymers are viscoelastic, and their mechanical behavior is dependent upon many factors. In the development of a film coating system, the evaluation of free films has been established as a valuable tool, since it can be readily used to characterize and evaluate the fundamental properties of the coating (8).

The objective of the present study was to evaluate the mechanical properties, using load-time profiles, of free films cast from two commercially available formulations of a plasticized aqueous dispersion of ethylcellulose differing only in plasticizer content (Surelease/E-7-7050 without silica and E-7-7060 containing dibutyl sebacate and glyceryl tricaprylate/caprate as plasticizers, respectively), and to study the effect of coalescence temperature on the mechanical properties of these films. Such an evaluation would help in characterizing the performance of plasticizers in the polymeric dispersions and, also, in assessing the fundamental mechanical behavior of plasticized polymeric films subjected to elevated temperatures during the coating process and, as a result, possibly on their *in situ* performance.

MATERIALS AND METHODS

Materials

Surelease/E-7-7050 without silica plasticized with dibutyl sebacate (DBS) and Surelease/E-7-7060 plasticized with glyceryl tricaprylate/caprate (GTC) were kindly provided by Colorcon, West Point, PA. (Commercially available Sure-

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lease/E-7-7050 formulation contains silica as an anti-tack agent, whereas Surelease/E-7-7060 formulation does not contain silica or any other anti-tack agent.) Pressure-sensitive protective overlay of polytetrafluoroethylene (Teflon) and anhydrous calcium sulfate (Drierite) were purchased from Fisher Scientific, Springfield, NJ.

Preparation of Free Films

Free films of both Surelease formulations were prepared using a draw-knife casting technique. This approach has been reported to be useful in obtaining reliable data regarding the fundamental properties of polymeric coating formulations (9). Glass plates (100 × 150 mm) were laminated with a pressure-sensitive protective overlay of Teflon and barriers of precise height (220 μm) were mounted on the edges of the plates. The polymeric dispersion was carefully poured onto the leveled plate avoiding bubble formation and spread uniformly over the entire surface with the help of a Gardner casting knife (Pacific Scientific/Gardner-Neotec Instrument Division, Silverspring, MD). The height of the barriers mounted on the edges of the plate and the knife setting were kept constant to control the thickness of the wet film in order to obtain the desired film thickness after drying. The wet film was dried in a leveled forced-air drying oven (Thelco Model 28, Precision Scientific, Chicago, IL), preset at a desired coalescence temperature, for 24 hr. While the dried film was still warm, it was peeled off from the plate with the help of a sharp knife, allowed to equilibrate to room temperature, sealed in a double plastic bag, and stored in a desiccator containing Drierite at 25°C prior to testing. Depending on the coalescence temperature employed, the mean thickness of the dried films was in the range of 50–75 μm.

Preparation of Film Specimens

All tests were conducted under controlled environmental conditions of 20 ± 1°C and 60 ± 2% RH. Before the measurement of mechanical properties, film specimens (10 × 100 mm) were cut using a sharp blade and examined visually for physical defects. Special care was exercised during cutting to avoid jagged edges which could otherwise produce stress concentrations during mounting. The thickness of each film specimen was measured at various points using a micrometer, and the mean thickness was used for the computation of mechanical properties. The film specimens coalesced at a specific temperature with more than 10% variation in thickness or physical deformities were discarded.

Measurement of Mechanical Properties

The mechanical properties of the films were measured from their load-time profiles. According to the ASTM guidelines (10), the data for tensile properties may be acquired in the form of a load-time (elapsed) profile instead of a typical load–displacement or stress–strain profile. The data collected to a load-time profile can be converted in the form of a load–displacement profile with the help of applied strain rate.

A tensile testing instrument (Model T5002, MTS Systems Corp., Minneapolis, MN) mounted with a 500-N capacity load cell was used for the measurements. Depending on the breaking load of the specimen, either a 0.01× (5-N) or a 0.04× (20-N) scaling amplification was used. The film specimen was clamped using an upper and a lower flat-faced metal grip laminated with cardboard to prevent film damage. The distance between the grips, and therefore the effective length of the film under stress, was kept constant at 50 mm. Proper alignment of the film specimen between the upper and the lower grip was checked before initiation of the test. A cross-head speed (strain rate) of 5 mm/min and a chart paper/cross-head speed ratio of 1:1 were used for all measurements. The load-time profiles were recorded electronically, and a hard copy was printed using an *X–Y* plotter. Figure 1 shows a typical load-time profile for a free film subjected to tensile testing. The load-time profiles of only those film specimens were considered which ruptured in the middle of the stressed film. An average of at least six measurements was taken for each film.

Three mechanical properties, namely, tensile strength, work of failure, and elastic modulus, were computed from the load-time profile, cross-head speed, and film dimensions. From preliminary studies, it was observed that the mechanical properties of the films were independent of their thickness within the range (50–75 μm) used in the present study. The theory behind the computation of these parameters is well documented (9,11) and therefore is not repeated here. However, it is necessary to present the final equations that define each of these parameters.

Tensile Strength. Tensile strength is the maximum stress applied to a point at which the film specimen breaks (Fig. 1). Tensile strength can be computed from the applied load at rupture and the cross-sectional area of fractured film as described in Eq. (1):

$$\text{Tensile strength} = \frac{\text{Load at failure}}{\text{Film thickness} \times \text{Film width}} \qquad (1)$$

The determination of tensile strength alone is not very useful in predicting mechanical performance of the films; however, higher values of tensile strength of the films are desirable for abrasion resistance.

Work of Failure. Work of failure is a function of work done in breaking the film specimen and is representative of

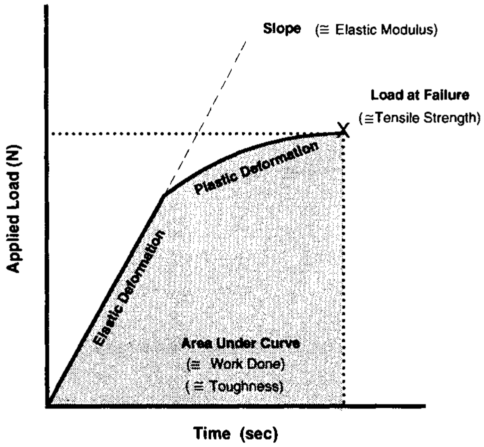

Fig. 1. A typical load-time profile observed in the tensile testing of free films.

the film toughness. It can be calculated from the area under curve of the load-time profile, cross-head speed, and the film dimensions as described in Eq. (2):

$$\text{Work of failure} = \frac{\text{Area under curve} \times \text{Cross-head speed}}{\text{Film thickness} \times \text{Film width}} \quad (2)$$

Elastic Modulus. Elastic modulus is the most basic and structurally important of all the mechanical properties and is a measure of stiffness of the film. It is the ratio of applied stress and corresponding strain in the region of approximately linear elastic deformation. It can be computed from the slope of the linear portion of elastic deformation on the load-time profile, cross-head speed, and film dimensions using Eq. (3):

$$\text{Elastic modulus} = \frac{\text{Slope}}{\text{Film thickness} \times \text{Film width} \times \text{Cross-head speed}} \quad (3)$$

RESULTS AND DISCUSSION

Characteristic features of the load-time or typical stress–strain profiles from tensile tests are routinely used to characterize polymer properties (12). For example, a soft and weak polymer is characterized by a low elastic modulus, low tensile strength, and low strain at break; a hard and brittle polymer is characterized by a high elastic modulus, moderate tensile strength, and low strain at break; a soft and tough polymer is characterized by a low elastic modulus, moderate tensile strength, and high strain at break; and a hard and tough polymer is characterized by a high elastic modulus, high tensile strength, and high strain at break. Toughness is directly proportional to the area under a load-time or load-displacement curve which is quantitated as energy. The higher the amount of energy a polymeric film can absorb prior to break under load, the higher is its toughness. Elastic modulus is an indicator of the elasticity of the film, with lower values corresponding to higher elasticity. On the other hand, tensile strength is an indicator of the film strength, with larger values corresponding to the stronger films. Ideally a film coating must be both strong and elastic and, therefore, must yield a higher tensile strength-to-elastic modulus ratio. This ratio has been used as an indicator of the overall mechanical quality of the films (13).

The commercially available DBS plasticized Surelease formulation contains silica as an anti-tack agent. During preliminary studies, it was observed that the films cast from the polymeric dispersion exhibited several microcracks upon drying and therefore were unsuitable for tensile testing. The microcracks in the dried films might have been due to the high magnitude of internal stress induced by the presence of insoluble silica particles. This hypothesis was substantiated by the absence of microcracks in the films cast from DBS plasticized Surelease without silica. Therefore, a DBS plasticized Surelease formulation without silica was used in the present study. In addition, since GTC plasticized Surelease does not contain silica, the use of DBS plasticized Surelease without silica allowed a true comparison of the effect of two different plasticizers in the polymeric formulations on the tensile properties of the films.

Figure 2 depicts the plot of tensile strength as a function of coalescence temperature for the films cast from both Surelease formulations. The tensile strength of the films increased with a corresponding increase in coalescence temperature and did not reach a plateau within the temperature range used in this investigation. In addition, the films cast from GTC plasticized Surelease were relatively stronger than those cast from DBS plasticized Surelease up to a coalescence temperature of 60°C. At temperatures above 60°C, no appreciable differences were observed in the tensile strengths of the films cast from both formulations. This could possibly be due to evaporation of the plasticizers from the films at elevated temperatures. Similar results from the tensile strength tests have been reported for the films of DBS plasticized Surelease (7). The data of the present study indicate that, within the normal range of drying temperature (40–60°C), films cast from GTC plasticized Surelease would be more resistant to stress than those from DBS plasticized Surelease. This could be an important criterion for the controlled release coating applications in which the physical integrity of the coated dosage form is very important in providing and maintaining desired and reproducible drug release rates.

Figure 3 compares the values of work of failure plotted as a function of coalescence temperature for the films of both Surelease formulations. No significant difference was observed in the work of failure for the films cast from either formulation of the polymeric dispersion coalesced at temperatures up to 50°C. At higher coalescence temperatures, however, films cast from GTC plasticized Surelease absorbed significantly higher energy, as evident from higher values of work of failure, compared to the films cast from DBS plasticized Surelease. This may possibly be due to the ability of GTC molecules to penetrate into the polymeric network of ethylcellulose more rapidly and completely compared to the DBS molecules at higher coalescence temperatures. From the load-time profiles, it was observed that the films of both formulations of Surelease coalesced at and above 50°C exhibited distinct and prolonged plastic deformation with the resultant increase in the area under the curve. This behavior could possibly be due to a strong interaction between the neighboring polymer particles at and above the coalescence

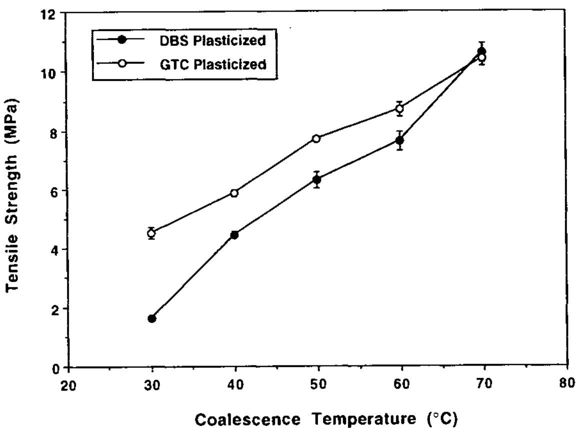

Fig. 2. Plot of tensile strength of free films as a function of coalescence temperature.

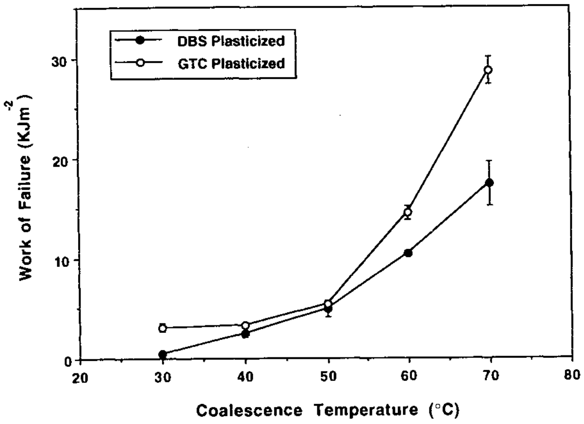

Fig. 3. Plot of work of failure of free films as a function of coalescence temperature.

temperature of 50°C, thus creating a smooth and continuous structure of the films.

Figure 4 illustrates the representative load-time profiles obtained for the films cast from DBS plasticized Surelease coalesced at temperatures below and above 50°C. Similar profiles were obtained for the films cast from GTC plasticized Surelease. As evident from Fig. 4, the fracture strain or percentage elongation at break of the films is directly proportional to the coalescence temperature within the range studied. This indicates that the films coalesced at a higher temperature are more ductile. The fracture strain values of the films obtained in the present study were in the range of 1–5%. Arwidsson *et al.* (7) have also reported fracture strain values in the range of 1–5% for the sprayed films of Aquacoat plasticized with different levels of triethyl citrate, a water-soluble plasticizer, and of DBS plasticized Surelease. These authors have reported that the sprayed films of Surelease with an additional quantity of DBS exhibited higher fracture strain values (ranging between 10 and 15%) compared to those of Surelease without additional DBS. However, the authors have shown that the fracture strain of sprayed films of Surelease without additional DBS did not show a distinct trend throughout the temperature range studied (50 to 90°C). This observation is in contrast to the observation of the present study. The difference in the temperature dependency of the strain values between those reported by Arwidsson *et al.* (7) and the present study could be due to several reasons, e.g., the difference in the technique of preparing the film (sprayed vs cast) and the difference in the process conditions. Arwidsson (6) has also reported similar fracture strain values (2–8%) for the films sprayed from an organic solution of ethylcellulose without a plasticizer as a function of selected coating process variables. The strain rate (cross-head speed) used in the referenced studies was 0.4 mm/min, compared to the 5 mm/min used in the present study. This is an important parameter to be considered, especially when comparing the results of two studies, since the strain or percent elongation of a polymeric film is normally inversely proportional to the rate of strain (9). Li and Peck (8) studied mechanical properties of free films of an analogous aqueous polymeric dispersion (silicone elastomer latex) plasticized with polyethylene glycols of three different molecular weights, each at 10, 20, and 30% concentration, and reported strain values in the range of 1–8%. In this study, the strain rate used was 1 mm/min.

The influence of coalescence temperature on the elastic

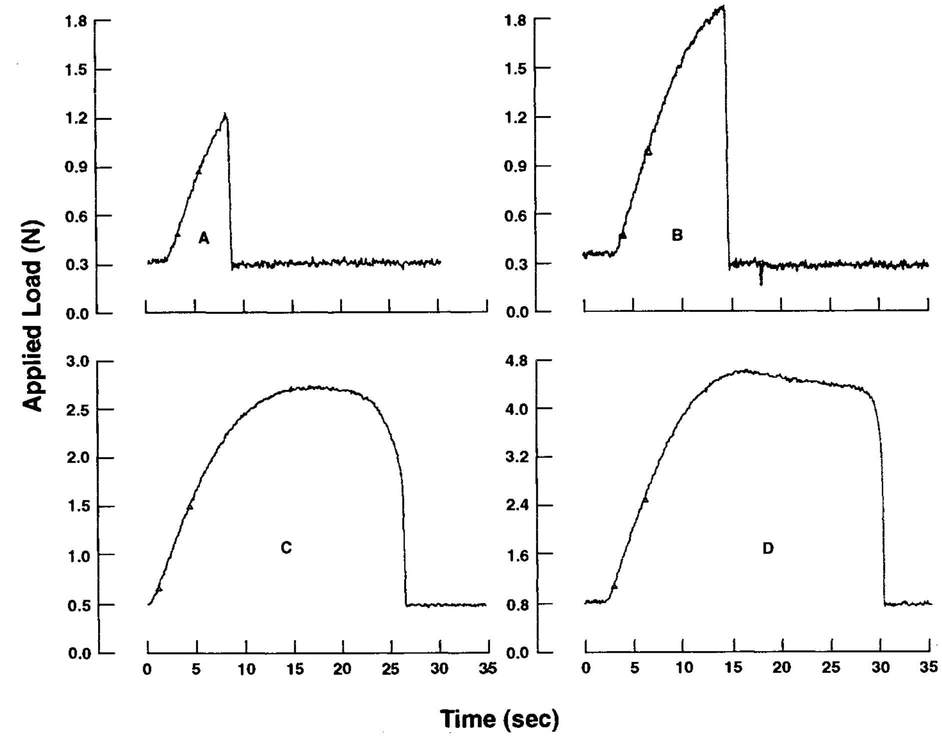

Fig. 4. Representative load-time profiles of free films of DBS plasticized Surelease coalesced at (A) 30°C, (B) 40°C, (C) 60°C, and (D) 70°C.

modulus values of the films cast from both Surelease formulations is presented in Fig. 5. It can be observed that, in general, as the coalescence temperature increased, the elastic modulus values of the films increased almost linearly for both the formulations. As observed for the values of tensile strength, the films cast from GTC plasticized Surelease showed consistently higher values of elastic modulus than those cast from DBS plasticized Surelease up to coalescence temperatures of 60°C, beyond which the reverse trend was observed. Within the normal range of drying temperature (40–60°C), the lower values of elastic modulus observed for the films of DBS plasticized Surelease indicate a higher elasticity compared to those of GTC plasticized Surelease.

The three parameters, i.e., tensile strength, work of failure, and elastic modulus, are good indicators of the mechanical properties of the polymeric films. However, independent interpretation of these parameters may not always lead to meaningful conclusions with respect to the *in situ* performance of coating on pharmaceutical substrates (i.e., tablets or pellets) produced under dynamic coating conditions. The ratio of tensile strength to elastic modulus of free films has been related to their *in situ* performance, and it has been shown that lower values of tensile strength-to-elastic modulus ratio correlated with increased coating defects (13). Since coating with minimal defects is critical to provide and maintain consistent and reproducible drug release rates from the coated controlled release dosage forms, a higher value of this ratio is desirable. Figure 6 shows the plot of the ratio of tensile strength to elastic modulus as a function of coalescence temperature for the films cast from both Surelease formulations. It is apparent that an increase in the coalescence temperature for films of both polymeric formulations led to an increase in this ratio up to a coalescence temperature of 50°C, beyond which the ratio decreased slightly. Since the highest tensile strength-to-elastic modulus ratio for the films cast from both Surelease formulations was observed at a drying temperature of 50°C, it may be presumed that the films coalesced at or around this temperature are less susceptible to physical defects. However, further studies would be needed to establish this relationship.

In summary, the mechanical properties of free films cast from two formulations of Surelease containing DBS and GTC as plasticizers, and coalesced at temperatures ranging between 30 and 70°C were evaluated using tensile stress analysis. The mechanical properties of the films were observed to depend on the type of plasticizer and the coalescence temperature. The films cast from DBS plasticized Surelease yielded lower values of tensile strength and elastic modulus compared to the films from GTC plasticized Surelease, indicating that the films cast from DBS plasticized Surelease were relatively softer than those cast from GTC plasticized Surelease. This suggests that DBS might be a more effective plasticizer than GTC. With an increase in the coalescence temperature, the films of Surelease formulations exhibited a characteristic transition in their mechanical properties, i.e., from a soft and weak (as evident from lower values of tensile strength, strain at break and elastic modulus) to a hard and tough system (as evident from higher values of tensile strength, strain at break, and elastic modulus). The optimal values of the tensile strength-to-elastic modulus ratio obtained for films of both formulations dried at 50°C indicate that the films coalesced at or around this temperature may be less susceptible to physical defects. The approach for the evaluation of tensile properties presented here can be used to evaluate the performance of plasticizers and their levels in the polymeric film coating formulations.

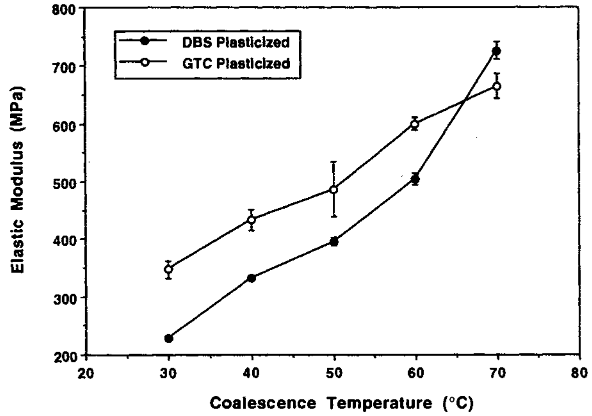

Fig. 5. Plot of elastic modulus of free films as a function of coalescence temperature.

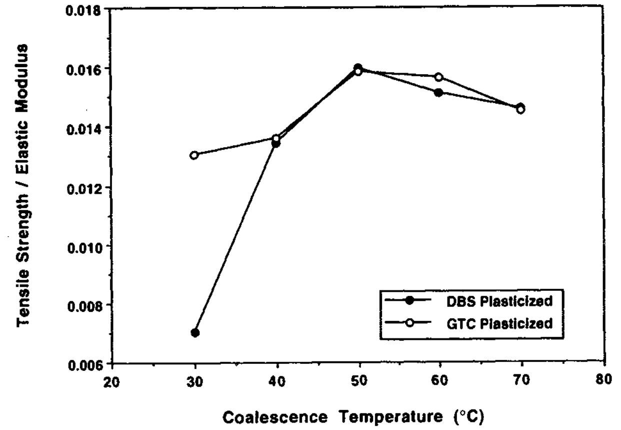

Fig. 6. Plot of the ratio of tensile strength to elastic modulus of free films as a function of coalescence temperature.

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