

Kinetics of Plasticizer Migration between Nitrocellulose and Ethylcellulose in Double-Base Propellant Systems

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INTRODUCTION

Internal-burning double-base-propellant rocket charges are necessarily covered with a combustion-resistant substance on all surfaces except the interior perforation. The restricted burning, coupled with a specific geometric form for the internal propellant surface, permits some control over thrust level and burning time. This inhibitor, which also acts as a liner to reduce heat transfer to the motor wall, is generally made of some plastic material that has a coefficient of thermal expansion not greatly different from that of the propellant. It is a requirement of the restricted-burning rocket that the inhibitor remain adhered to the propellant grain, although their temperature may change, drastically, several times before the unit is fired.

Whenever dissimilar plastic materials are placed in physical contact, the probability exists that the plasticizers present will interdiffuse slowly between the two materials. This process occurs with an inhibiting plastic applied to double-base propellant, whereby substantial quantities of nitroglycerine may be lost from propellant to inhibitor.

This transfer may lead to a significant change in the performance of the round, depending largely on the caliber. The effect is more serious in small calibers because (1) the ratio of inhibitor mass to propellant mass is larger, leading to a greater ultimate depletion of the nitroglycerine content of the propellant, and (2) the diffusion path is shorter, causing the effect to appear more quickly. If stored for a sufficient time, the total impulse drops from its normal value, and the burning time is increased. Toward the end of burning, the burning rate slows down, with a resultant drop in pressure, which further prolongs the burning time. These ballistic modifications occur during storage mainly because of the plasticizer exchange between propellant and inhibitor. Nitroglycerine migrates from the propellant to the inhibitor and, in many cases, plasticizer migrates from the inhibitor to the propellant, contributing a negative quantity to the heat of explosion.

Initially, cellulose acetate was the preferred inhibitor material. However, when the cause of the ballistic property change was discovered, ethylcellulose was used instead of the cellulose acetate. Ethylcellulose is superior to cellulose acetate in its resistance to nitroglycerine. Even with the use of ethylcellulose, the migration effect is not eliminated.

Surveillance studies are necessarily conclusive in all questions of storage stability of a propellant round; however, any method of obtaining valid information more quickly would be advantageous. Ordinarily, surveillance studies are accelerated by storing at elevated temperatures in order that physical and chemical processes may occur more rapidly. However, the extrapolation of accelerated test data to lower temperatures involves serious questions. By what factor were the physical and chemical processes accelerated by a rise in temperature? Would the observed processes occur at all at a lower temperature? Elevated-temperature surveillance can produce useful results, but in many cases there is a distinct possibility that totally erroneous conclusions will be drawn.

In principle, it is possible to accelerate evaluation of processes such as plasticizer migration by some means other than elevated-temperature studies. Plasticizer migration is a case of diffusion between solid phases, and such processes can be treated quantitatively if given the boundary conditions and diffusion constants. The purpose of this study was to explore the possibilities and limitations of such an approach.

CALCULATIONS

Diffusion processes are treated normally by solving Fick's law of diffusion, with appropriate boundary conditions. This law has the form:

$$\partial C/\partial t = (\partial/\partial x)[D(\partial C/\partial x)] \quad (1)$$

where C is the concentration of diffusing material at an arbitrary point x along the one-dimensional diffusion path at the elapsed time t , and D is the diffusion coefficient associated with the process.

Solutions of this equation are given by Barrer¹ for a number of different geometrical cases. For the boundary conditions of instantaneous equilibrium of the polymer surface with the surrounding vapor, the solution is:

$$\frac{Q}{Q_0} = 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp \left\{ - \frac{(2n+1)^2 \pi^2 D t}{l^2} \right\} \quad (2)$$

where Q is the weight change at time t , Q_0 is the equilibrium weight change, D is the diffusion coefficient, and l is the polymer film thickness. The series converges rapidly, and only the first term of the series will be important for times larger than zero. One can therefore evaluate D when it is independent of concentration, for there is the implicit assumption in this form of the equation (eq. (1)) that D is independent of concentration and time. That this is not ordinarily the case for diffusion of solvents

in polymers is pointed out theoretically and experimentally by Crank and Henry,² Mandelkern and Long,³ King,⁴ Rouse,⁵ Long and Thompson,⁶ and others. Therefore, one would not expect to apply eq. (1) to the propellant inhibitor system without modification.

The concentration dependence of the diffusion coefficient can be treated mathematically^{1,7} by introducing the assumption that C is a function of $x/t^{1/2}$. The complete solution, giving concentration distribution as a function of time, then becomes unwieldy. One discusses, instead, the net quantity Q of diffusing material that has passed through unit area at time t . The unit area is placed conveniently at the phase interface, perpendicular to the diffusion direction. This leads to the simple semi-empirical equation:

$$Q/Q_e = K(t^{1/2}/l) \quad (3)$$

or, expressed in terms of average concentration,

$$C_t = K'(t^{1/2}/l) \quad (4)$$

The constant K is independent of concentration and of sample thickness, but is a function of temperature. There is the limitation that this simple linear relationship cannot hold much beyond the half-time to equilibrium for a finite system.

The extensive recent literature in this field indicates that these equations may be applied to the propellant-inhibitor system. For nonpolar solvent-polymer systems, which are well above their second-order transition, temperature diffusion appears to be Fickian with a diffusion coefficient that increases with concentration. Such a system is isobutane and polyisobutylene, studied by Prager and Long.⁷ For polar solvent-polymer systems, close to or below the second-order transition temperature of the polymer, the agreement is not so good. A sigmoid relationship commonly occurs³ between the variables, in place of the linear one predicted by eq. (3), and K becomes somewhat dependent on sample thickness.

These aberrations (often called anomalous, or non-Fickian, diffusion) may be explained qualitatively. The sigmoid shape of the sorption isotherm probably is caused by the stress release that occurs as the diffusing solvent begins to soften the last of a film of polymer and the film expands in area.^{8,9} The slight dependence of K on sample thickness is explained⁹ by postulating a time delay in the establishment of an equilibrium value for the diffusion coefficient (and/or the surface concentration). It is an observed fact that thicker samples may reach sorption equilibrium somewhat sooner than predicted by eq. (3).

The polar polymer-polar solvent system of cellulose nitrate, ethylcellulose, and nitroglycerine has been found to depart from theory in both the respects just discussed. In rockets using a double-base-propellant ethylcellulose, specifications permit a variation of $\pm 20\%$ in the inhibitor thickness. Therefore, a high degree of accuracy is not required in the treatment of plasticizer migration; moreover, the direction of the error

is known, and something of its magnitude, so it is possible to make reasonably accurate allowances. Thus, there seemed to be sufficient promise in this approach to justify an experimental test for later correlation with results of actual surveillance studies of rockets made with the double-base ethylcellulose system.

MATERIALS AND PROCEDURES

The propellant-inhibitor combination selected for study was specified for a 2 in. diameter rocket. The propellant contained nitrocellulose (12.6% N), nitroglycerine, diethyl phthalate, and smaller quantities of stabilizers and manufacturing aids. The inhibitor was made from 50 cpoise standard ethylcellulose, 1,1,4,4-tetramethylbutylphenyl ether, and diethyl phthalate.

Samples were prepared from propellant sheets by facing one side in a lathe until the thickness was reduced to the desired value, usually 58 mils. From this area, a circular disk was punched with a 0.968 in. ID sharpened brass ring. The ring was left in place during the experiment, to restrict evaporation to the two flat surfaces. Because both flat surfaces are exposed, the effective thickness will be one half of the measured value, or 29 mils; the same treatment will apply to the inhibitor.

The inhibitor plastic was received in the form of a tube that had been extruded around a wooden model of the rocket grain and subsequently stripped. A section of the tube was opened and flattened between metal plates in an oven at 95°C., resulting in a sheet about 50 mils thick. This sheet was machined to the desired thickness, about 10 mils, and a circular disk of the same diameter as the propellant was punched out. It was unnecessary to consider edge effects with such a thin disk.

Samples of propellant and inhibitor were taken purposely from material with orientation approximating that in the completed charge, because orientation is known to have a considerable effect on the rate of diffusion in polymers.⁹

Dimensions were chosen to provide equal areas of exposed surface and to duplicate the propellant-to-inhibitor weight ratio of approximately 7.9 in the rocket charge (Table I).

TABLE I
Some Dimensions of the 2 in. Rocket Charge

| | |
|---------------------------------------|-------------|
| Inhibitor thickness, in. | 0.040-0.060 |
| Propellant web thickness, in. | 0.302-0.514 |
| Inhibitor wt., g. (mean) | 175 |
| Propellant wt., g. (mean) | 1,380 |
| Propellant inhibitor wt. ratio (mean) | 7.9 |

Apparatus

Plasticizer migration can be conveniently followed in the laboratory by utilizing the principle of vacuum contact. The plastics to be studied

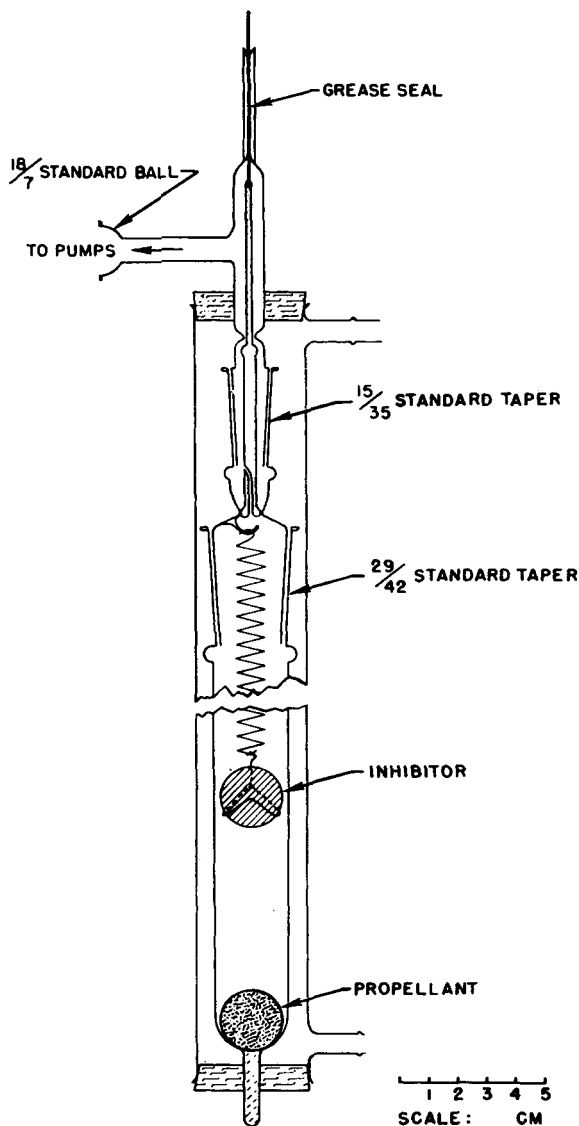


Fig. 1. Diagram of the apparatus used to study the kinetics of plasticizer migration.

are placed in reasonable proximity in an evacuated system. When all fixed gases (air, etc.) have been removed, the transfer of plasticizers through the vapor phase is so rapid that the rate-determining process is solid-state diffusion, precisely as though the materials were in physical contact over all exposed surfaces.

Considerable attention was given to the problem of designing a practical apparatus (Fig. 1). The chief problem was to ensure the virtual absence of fixed gases and at the same time avoid pumping away the plasticizer

vapors. As a solution to the problem, the system was evacuated continuously during the experiment through a capillary leak designed to permit the escape of fixed gases while holding the rate of vapor loss at a tolerable level (0.2 mg./hr. at the highest experimental temperature). The section of capillary tubing that comprised the leak was made movable, with a ball-and-cone ground joint on each end. When the movable section was lifted up, the system was connected through the capillary leak; in any intermediate position the valve was wide open, permitting rapid evacuation. This arrangement eliminated light molecules of air or water about three times as fast (on a molecular basis) as the heavy plasticizer molecules.

The weight gain of the inhibitor sample was followed as a function of time by observing the length of the calibrated quartz helix.

Samples of propellant and inhibitor were placed in the sorption tube in the positions indicated in Figure 1, and the system was evacuated to a pressure of about 10 mm. This pressure is sufficient to prevent plasticizer interchange, yet low enough to allow the escape of most dissolved gases. After an overnight period at room temperature, the circulation of thermostatted water was begun, the initial spring length was observed, and the remainder of the air was removed by evacuation for 1 min. with the valve wide open. For the rest of the experiment, the valve was placed in the capillary-leak position. The position of the lower end of the spring was observed with a cathetometer at intervals proportional to the square root of the time elapsed after the final evacuation.

Equilibrium Migration

For the study of migration equilibrium, it is not necessary to use equipment as elaborate as that in Figure 1, provided the kinetics of the system under study is sufficiently well known so that the time necessary for attainment of equilibrium can be estimated. For such cases, it is sufficient to put the two plastics together into a test tube, which is then evacuated, sealed, and stored at the desired temperature for the required length of time.

The film of ethylcellulose to be tested (not more than 0.010 in. thick) is weighed, then placed in a vacuum desiccator, and pumped continuously with a good mechanical pump for 1 hr. The loss in weight is a measure of the volatile materials in the film. The same piece is then placed in a 20 × 150 mm. test tube containing a weight of propellant turnings equal to twice the weight of the film. The test tube is then "necked" down and a piece of 7 mm. tubing provided with two constrictions is sealed on. This unit is evacuated for 1 hr. (either alone or in manifold with others) by a pump capable of reducing the pressure to 1 μ or less, then sealed off at the upper constriction. After being stored several days completely immersed in an oil or water thermostat at the desired temperature, the tube is removed and the vacuum released *at once* by breaking the 7 mm tube at the second constriction. The tube is then opened, the propellant and

the inhibitor are weighed, and the inhibitor film is analyzed polarographically for nitroglycerine content. Each film sample is run in duplicate. An additional set of tubes is permitted to remain in the bath for a longer period of time as a check on whether equilibrium had been attained. The gain in weight and the gain in nitroglycerine are reported in terms of per cent of original sample weight.

The choice of a 2:1 ratio of propellant to inhibitor represents a compromise of several factors. For the storage times considered, not all of the propellant web enters into plasticizer interchange with the inhibitor. The ratio is arbitrary; however, heat-of-explosion experiments indicate that this ratio is in near agreement with the propellant thickness actually taking part in the process. Moreover, this ratio approaches equilibrium more quickly than a higher ratio because the amount of material transferred is less; yet, ample material is transferred to permit accurate measurement of weight gain and nitroglycerine content.

Chemical Analysis

The measured weight gain gives the net plasticizer transfer, but does not provide information as to what proportion of this transfer is nitroglycerine. Such information can be obtained only by chemical analysis of the inhibitor at the conclusion of the experiment. A polarographic procedure was used for this analysis.¹⁰⁻¹²

The inhibitor sample was dissolved in such a volume of 95% alcohol that 10 ml. of solution contained about 1 mg. of nitroglycerine. A 10 ml. aliquot of this solution was added to 5 ml. of aqueous 0.1*M* tetramethylammonium chloride containing maximum-suppressing dye, purged with nitrogen, and polarographed. A measured volume of a standard alcoholic solution containing about 1 mg. of nitroglycerine was added and the sample was polarographed again. The weight of nitroglycerine in the aliquot was calculated from the wave heights on the two polarograms.

RESULTS

The curves in Figure 2 represent the results of kinetic experiments at three temperatures, chosen to correspond to those commonly used in full-

TABLE II
Experimental Parameters

| Temp., °F | Propell. thickness, mils ^a | Propell. wt., mg. | Inhib. thickness, mils ^a | Inhib. wt., mg. | Propell. inhib. wt. ratio |
|--------------|---|----------------------|---|--------------------|---------------------------------|
| 70 | 58.0 | 1,078 | 10.5 | 134.3 | 8.02 |
| 95 | 58.0 | 1,093 | 10.0 | 132.0 | 8.28 |
| 130 | 60.0 | 1,118 | 10.3 | 135.2 | 8.22 |
| | 77.2 | 1,445 | 14.0 | 184.5 | 7.83 |

^a The effective thicknesses of propellant and inhibitor are one half the values tabulated, because diffusion proceeds symmetrically from both surfaces of the disks.

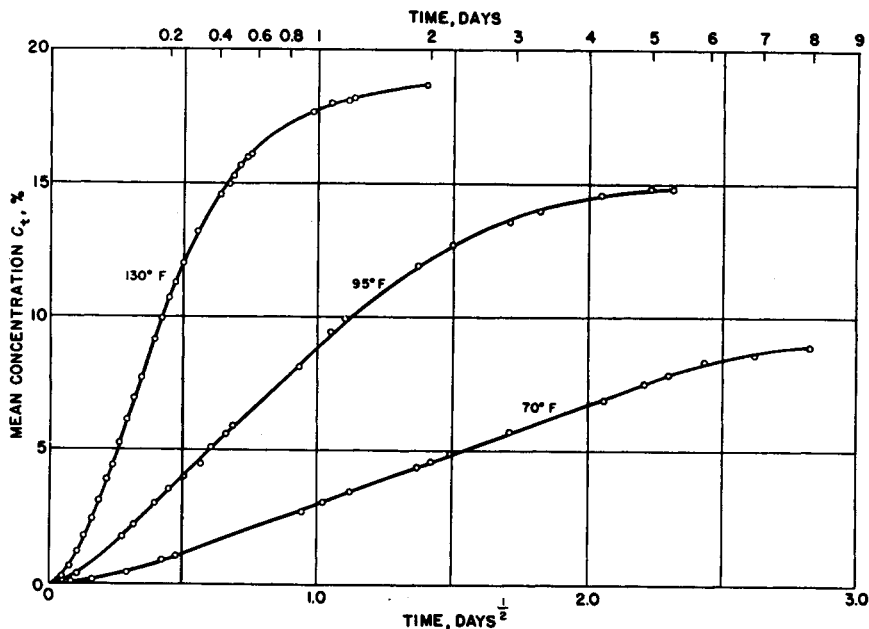


Fig. 2. Plasticizer-migration curves for the small-scale one-dimensional model of the 2 in. rocket propellant charge.

scale surveillance. Experimental parameters are given in Table II; experimental and calculated results are given in Table III.

Figure 3 shows two approximately coincident curves. The 130°F. data are replotted with data from another experiment at the same temperature; the propellant and inhibitor thicknesses are in the ratio of

TABLE III
Experimental Results

| Temp., °F. | Equilibrium conditions | | K', in./day ^{1/2} |
|---------------|------------------------|--------------------------------------|-------------------------------|
| | Inhib. wt. gain, % | Wt. gain due to nitroglycerine, % | |
| 70 | 13.7 | 10.7 | 0.020 |
| 95 | 17.9 | 15.0 | 0.040 |
| 130 | 22.4 | 18.3 | 0.15 |
| | 22.4 | 18.3 | 0.14 |

1:1.3. The root-time data from the latter experiment were all divided by the factor 1.3 before plotting. This demonstrates the predictability of the relationship between linear dimension and time, as will be discussed in more detail below.

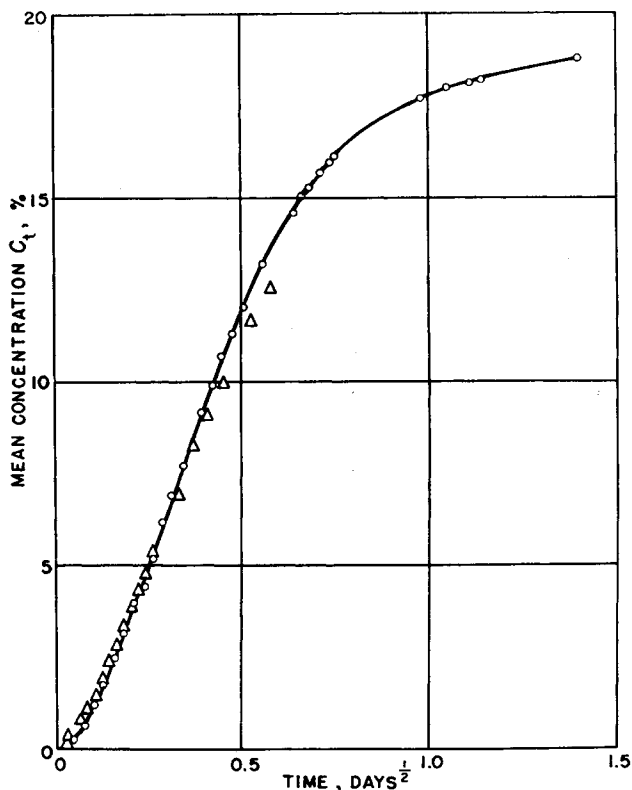


Fig. 3. Plasticizer-migration isotherm (130°F.): (O) standard sample; (Δ) thick sample.

DISCUSSION

The experimental curves (Fig. 2) come very close to obeying the simple relationship of eq. (3); the initial curvature characteristic of polar-solvent-polymer systems is slight. The slope, K' , of the near-linear portion of each curve has been calculated and entered in Table III.

To use these values in extrapolating small-scale measurements to the full-scale rocket grain, it must be demonstrated that K' is essentially independent* of sample thickness. The near-coincidence of the curves in Figure 3, which results in substantially equal K' values in Table III, is encouraging. Admittedly, this test covers only a relatively small thickness ratio (1:1.3), but samples of a larger ratio are difficult to handle with laboratory apparatus.

A plot of $\ln K'$ versus $1/RT$ gives a straight line with a slope equal to 11.6 kcal. This value is of the correct order of magnitude for the energy of activation in diffusion processes. An interpretation of this value would

* A rocket system having a larger propellant web, which utilized an inhibitor that contained approximately half the percentage of plasticizer used in the present work, showed a measurable increase of K' with sample thickness when this test was applied.

be highly speculative; however, it can be useful in predicting K' to a first approximation at any temperature.

Given the experimental values of K' for the unit under consideration, one can use eq. (3) to calculate C_i , the average concentration of migrated plasticizer into inhibitor of nominal thickness $l = 0.050$ in. stored for any period of time at a selected temperature. Actually, it is not necessary to make such a calculation, because the result can be read directly from Figure 2 by applying a scale factor of 100 to the time coordinate; that is, 1 day of the sorption experiment is equivalent to 100 days of storage of the full-scale charge. This convenient scale factor was provided by the deliberate choice of a factor of 10 in scaling down the linear dimension of the 2 in. rocket system; the 0.050 ± 0.010 in. inhibitor thickness of the 2 in. rocket became 0.005 ± 0.001 in. for the sorption experiment.

An approximation is involved in treating the 2 in. rocket as a one-dimensional system, that is, as a pair of infinite flat slabs of propellant and inhibitor. It would be a closer approximation to consider the 2 in. rocket as two-dimensional, a pair of infinite concentric cylindrical tubes. The additional mathematical complication is not justified for this exploratory experiment, particularly since it can be shown that the errors introduced by the one-dimensional simplification are in opposite directions and in some degree compensating. If one rolls a portion of the infinite flat slab of the one-dimensional case into an infinite tube, keeping the mass constant, the propellant phase must become thicker. However, the radial convergence of the diffusion paths tends to compensate for this increase in path length.

The data of Figure 2 probably cannot be used to predict plasticizer migration in the rocket charge much beyond the linear portions of the curves. The final stages of diffusion in a finite system are not treated by the theory, and one cannot predict whether the equivalence between linear dimension and square root of time is valid under these conditions. Moreover, an unavoidable experimental error caused by the small but continuous loss of vapor from the system begins to have its effect, causing a premature approach to a limiting value of plasticizer migration, which is too low. This is demonstrated by separate experimental determinations of the equilibrium migration in a completely closed system (Table III).

It is important to observe that the equilibrium value for plasticizer migration is different at the three experimental temperatures. This illustrates the risk involved in assuming that storage at high temperatures affects only the rate of physical and chemical processes. In some cases, the effect on equilibrium may be more important than the effect on rate. This limited work indicates that a month of storage at 130°F . may not be equivalent to any storage period at 70°F ., because at the latter temperature the plasticizer migration may never reach the level attained in a month at 130°F .

Another factor affecting the kinetics of plasticizer migration in the practical case is the possible presence of low molecular weight solvent.

The 2 in. rocket charge was chosen for this study because it has an extrusion-coated grain. However, the charge may contain 1% of ethyl lactate-butyl acetate solvent, because precoating is necessary to ensure satisfactory bonding of the inhibitor. The bulk of this solvent will be concentrated initially near the propellant-inhibitor interface, where its concentration may be quite high for a short time. Only qualitative statements can be made about the influence of this solvent; it will distribute itself much more rapidly (probably by a factor of 10) than the high molecular weight plasticizers; it will tend somewhat to accelerate the migration of the plasticizers, because of its softening effect on the propellant.

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Synopsis

Recent experimental and theoretical work on the concentration-dependent diffusion of solvents in polymers has led to a reasonably detailed understanding of the processes involved. The possibility exists of supplementing standard but costly surveillance testing with rapid laboratory-scale measurements of plasticizer migration. Many solvent-polymer systems conform rather closely to the assumption that the amount of material transferred by diffusion is a function of the composite variable $t^{1/2}/l$. This means that any scale factor applied to the diffusion path length, in a one-dimensional system, may be compensated for by applying the square of this factor to the time. It has been shown that this assumption can apply with reasonable accuracy to plasticizer diffusion in the propellant-inhibitor systems. By approximating a particular 2 in. rocket charge with a one-dimensional model, it is possible to predict the time required for the unit's inhibitor to reach specified levels of plasticizer migration at any storage temperature of interest. A large temperature coefficient appears to be associated with the equilibrium plasticizer migration in the system chosen for experiment, as well as with the rate. Unless suitable allowance is made, this can lead to serious misinterpretation of the results of the surveillance that has been accelerated by raising the storage temperature.

Résumé

Des expériences récentes et des travaux théoriques sur la diffusion des solvants en fonction de la concentration dans les polymères, ont permis d'en comprendre le mécanisme d'une façon suffisamment détaillée. On a trouvé la possibilité de compléter le travail d'analyse standard mais coûteux par une mesure rapide pratiquée au lab-

oratoire sur la migration des plastifiants. Beaucoup de systèmes solvant-polymère se conforment d'une façon plutôt regoureuse à la conception suivant laquelle la quantité de matériel transféré par diffusion est une fonction de la variable complexe $t^{1/2}/l$. Cela signifie que n'importe quel facteur d'échelle appliqué à la longueur du parcours de diffusion, dans un système uni-dimensionnel, peut être compensé par application du carré de ce facteur de temps. On a montré que cette conception peut être appliquée avec une précision raisonnable à la diffusion de plastifiant dans les systèmes accélérateur/inhibiteur. En assimilant une charge spéciale de 2 pouces à un modèle uni-dimensionnel, il est possible de prédire le temps requis par l'inhibiteur pour atteindre un niveau spécifié de migration de plastifiant à n'importe quelle température à laquelle on veut stocker. Un grand coefficient de température est associé à la migration plastifiant à l'équilibre dans le système choisi pour l'expérience, il existe de même pour la vitesse. A moins d'en tenir compte, ceci peut conduire à des interprétations complètement fausses des résultats de la surveillance par suite d'une augmentation de la température de stockage.

Zusammenfassung

Neuere experimentelle und theoretische Untersuchungen über die konzentrationsabhängige Diffusion von Lösungsmitteln in Polymeren haben zu einem ausreichenden Verständnis der dabei ablaufenden Vorgänge geführt. Es besteht die Möglichkeit, die kostspieligen Standardüberwachungstests durch rasche Messung der Weichmacherwanderung im Labormassstab zu ergänzen. Viele Lösungsmittel-Polymeresysteme entsprechen ziemlich genau der Annahme, dass die durch Diffusion transportierte Substanzmenge eine Funktion der zusammengesetzten Variablen $t^{1/2}/l$ ist. Das bedeutet, dass jeder Massstabfaktor, der für die Länge des Diffusionsweges in einem eindimensionalen System verwendet wird, durch Anwendung des Quadrates dieses Faktors auf die Versuchsdauer kompensiert werden kann. Es wurde gezeigt, dass diese Annahme mit brauchbarer Genauigkeit auf die Weichmacherdiffusion in Propellant-Inhibitorsystemen angewendet werden kann. Durch Annäherung einer speziellen 2-inch Raketenladung durch ein eindimensionales Modell ist es möglich, die für die Erreichung einer spezifischen Weichmacherwanderung durch den Inhibitor der Einheit notwendige Zeit bei einer beliebigen in Frage kommenden Lagerungstemperatur zu berechnen. Ein grosser Temperaturkoeffizient scheint in dem gewählten Versuchssystem sowohl für das Weichmacherwanderungsgleichgewicht als auch für die Geschwindigkeit zu bestehen. Ohne geeignete Berücksichtigung kann das zu ernstlichen Fehldeutungen der Ergebnisse der durch Erhöhung der Lagerungstemperatur beschleunigten Überwachungstests führen.

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