POLYMOLECULARITY AND MECHANICAL PROPERTIES 
OF CELLULOSE ACETATE

By Arnold M. Sookne and Milton Harris 1

ABSTRACT

The tensile strengths, ultimate elongations, and folding endurances of films prepared from a series of cellulose acetate fractions and blends were studied. When the mechanical properties are plotted against the intrinsic viscosities (or relative weight-average degrees of polymerization), the results for the fractions and different blends fall on separate curves. In contrast, when the mechanical properties are plotted against the number-average degrees of polymerization, the results for the fractions and all of the blends fall approximately on a single curve for each property. The results are shown to be qualitatively consistent with the hypothesis that the mechanical properties of blends are the weight-averages of the properties of their components. The results emphasize the importance of the determinations of the number-average degree of polymerization in studying commercial polymeric materials.

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I. INTRODUCTION

High polymers are in general polymeric in that there is, heterogeneous with respect to molecular size. The problem of the influence of the degree of polymericity on the mechanical properties of such materials is an important one, and it has received considerable study in recent years. The detailed results of these investigations are reviewed elsewhere [1, 2, 3]2. The procedure generally followed in these studies was to fractionate a polymer with respect to molecular size, and compare the mechanical properties of the fractions with those of blends. There has been substantial agreement among the more recent investigators that fractionated cellulose materials are superior in mechanical properties to blends of the same intrinsic viscosity; i.e., the same weight-average molecular weight [1, 4, 5]. In particular, it has been found that the presence of material of low

1 Research Associates at the National Bureau of Standards representing the Textile Foundation.

2 Figures in brackets indicate literature references at the end of this paper.
molecular weights exerts a harmful influence on the mechanical properties of blends. These results appear to be unambiguous only for unoriented or slightly oriented specimens; it is quite possible that other conclusions may be reached when oriented or crystallized materials are considered.

In an approach to this problem some time ago, a large quantity of commercial cellulose acetate was fractionated in this laboratory [6], and some of the mechanical properties of the fractions and their blends were studied [4]. It was found that these mechanical properties were more closely related to the number-average than to the weight-average \( DP_n \)'s of the samples. However, the \( DP_w \)'s were at that time calculated from viscosity measurements at only one concentration (by means of Staudinger's rule), and the \( DP_n \)'s of the blends were calculated under the assumption that the fractions were perfectly homogeneous. The \( DP_n \)'s of the fractions have now been determined experimentally by osmotic pressure measurements in acetone solution, and their intrinsic viscosities have been determined in the same solvent [7]. It is the purpose of this discussion to re-examine the data for the mechanical properties of the fractions and blends in the light of these new measurements of \( DP_n \) and intrinsic viscosity. In addition, examination of the data shows that to a first approximation the mechanical properties of the blends are the weight-averages of the properties of their components; i.e.,

\[
\text{Property}_{\text{blend}} = \frac{\sum w_i P_i}{\sum w_i},
\]

(1)

where \( w_i \) is the weight of the molecular species with a mechanical property \( P_i \). It will be shown that the experimentally obtained relationships between the mechanical properties and the \( DP_n \)'s and intrinsic viscosities are all consistent with this relationship.

II. RESULTS AND DISCUSSION

The separation of the fractions [6], measurement of the osmotic pressure values and intrinsic viscosities [7], and the preparation and mechanical testing of films of the fractions and blends [4] have been described in detail elsewhere.

1. MECHANICAL PROPERTIES AS A FUNCTION OF INTRINSIC VISCOSITY AND NUMBER-AVERAGE DEGREE OF POLYMERIZATION

Figure 1 shows the tensile strengths of the fractions as a function of their intrinsic viscosities (upper graph) and \( DP_n \)'s (lower). In considering these and subsequent data (figs. 1 to 6), the intrinsic viscosity may be taken as a direct measure of \( DP_n \) on the basis of Kraemer's results [8] and because of the linear relationship between \( DP_n \) and \([\eta] \) (intrinsic viscosity) found for the fractions [7]. Because of the uncertainty in the value of Kraemer's constant, the data have been plotted as a function of \([\eta] \) rather than of \( DP_n \).4 The curves have

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4 The weight-average and number-average degrees of polymerization will henceforth be written \( DP_w \) and \( DP_n \), respectively.

4 The scale of the axis of abscissae in each upper graph of figures 1 to 6 has, however, been so adjusted that it compares directly with the \( DP \) axis of the lower graph, assuming \( DP_w = 230 [\eta] \) (Kraemer's relationship).
Figure 1.—Tensile strengths of cellulose acetate fractions as a function of intrinsic viscosity (upper graph) and DP, (lower).

A large slope in the region of small chain length, followed by a gradually decreasing slope which may represent an approach to an asymptotic value for very high DP's. Similarly shaped curves have been observed for numerous polymers [9]. The fraction of lowest chain length (DP,=43) was so brittle that it would not form a coherent film of sufficient size to be tested by ordinary technics, and it has therefore been assigned a strength of zero. Small fragments of this sample were tested by special technics, however, and a strength as high as 250 kg/cm² was obtained. This result suggests that the failure to obtain measurable strengths for this and other samples of very low DP results from the inadequacy of the film casting and testing technics, rather than from the weakness of the sample. Although it is true that such materials have zero strength as far as their practical utilization is concerned, the possibility exists that the curve relating strength to DP would pass through the origin if suitable testing technics were used.

Figure 2 shows the ultimate elongations of the fractions, plotted in the same manner as the tensile strength results. The shapes of the latter curves are generally similar to those for the tensile strength data, except that the approach to a limiting value is completed at a lower DP for the ultimate elongation results. As shown in figure 3, entirely analogous results were obtained for the measurements of the folding endurances of the fractions (made on a Massachusetts Institute of Technology Fold Tester).

In order to study the effect of distribution of chain length on mechanical properties, blends were prepared from pairs of the fractions. The major portion of the work on blends was done with three series, prepared by mixing in various proportions pairs of fractions of
the following $\overline{DP}_n$'s: 502+43, 502+127 and 502+204. (These same pairs of fractions are identified by their intrinsic viscosities of 2.75+.23, 2.75+.70, and 2.75+1.37, respectively.) A number of miscellaneous blends and the starting material were also studied. The intrinsic viscosities and $\overline{DP}_n$'s of the blends were calculated from
their known weight compositions and the intrinsic viscosities and $\overline{DP}_n$'s of the fractions.$^5$

![Figure 4](image_url)

**Figure 4.—Tensile strengths of blends of cellulose acetate fractions as a function of intrinsic viscosity (upper graph) and $\overline{DP}_n$ (lower).**

The curve for the fractions (traced from fig. 1) is included for comparison.

Figure 4 shows the tensile strengths of the blends as a function of both their intrinsic viscosities (upper graph) and $\overline{DP}_n$'s (lower). The respective curves for the fractions, traced from the smooth curves of figure 1, are included for comparison. As shown by the upper graph of figure 4, the strengths of the blends are in general lower than those of the fractions when the results are plotted against intrinsic viscosity (or relative $\overline{DP}_n$). Further, the discrepancy between the curve for the fractions and that for a series of blends appears to increase with increasing difference in chain length between the components in a given series. Thus, the open circles, which represent blends of the longest and shortest chain lengths, fall farthest

$^5$ The intrinsic viscosities of the blends were calculated from the formula

$$[\eta]_{\text{blend}} = f_1[\eta]_1 + f_2[\eta]_2,$$

where $f_1$ and $f_2$ are the weight fractions in the blend of the components of intrinsic viscosities $[\eta]_1$ and $[\eta]_2$, respectively. The values of $\overline{DP}_n$ for the blends were obtained from the relationship

$$\overline{DP}_n^{\text{blend}} = \frac{1}{\frac{f_1}{\overline{DP}_n^1} + \frac{f_2}{\overline{DP}_n^2}},$$

where $f_1$ and $f_2$ are the weight fractions in the blend of the components of $\overline{DP}_n^1$ and $\overline{DP}_n^2$, respectively.

Calculated rather than experimental values of $[\eta]$ and $\overline{DP}_n$ for the blends were used because of the unquestioned validity of the calculation procedure, and because samples of the original blends sufficient for the experimental measurements were not available.
below the curve for the fractions. In contrast, when the tensile strengths are plotted against \( \overline{DP}_n \) (lower graph, fig. 4), the results for the blends, including the starting material and a number of miscellaneous blends not shown in the upper graph, in general fall closer to the smooth curve for the fractions. However, the open circles (which represent blends of the longest and shortest chain lengths) fall consistently somewhat above the smooth curve. The possible cause for this will be discussed below.

The results of the ultimate elongation measurements on blends (fig. 5) are qualitatively similar to the tensile strength data, but the differences between the upper and lower graphs are more clearly defined. In the upper graph, the curves for the blends of fractions of intrinsic viscosities 2.75+ .23 fall markedly below the smooth curve for the fractions (traced from fig. 2), the results for the series 2.75+.70 are intermediate, and those for the series 2.75+1.37 are quite close to the curve for the fractions. In the lower graph, in which the results are plotted against \( DP_n \), all of the results for the blends fall fairly close to the curve for the fractions; the open circles, however, again consistently fall somewhat above the smooth curve.

The results for the folding endurance measurements (fig. 6) are analogous to those for the ultimate elongations. The differences between the blends and fractions are again large when the results are plotted against intrinsic viscosity. The results for the series 2.75+1.37 appear to fall somewhat above the smooth curve for the fractions, but it is possible that this difference results from the variability inherent in folding endurance measurements rather than any actual superiority of the blends. As shown by the lower graph of figure 6, the results for all the blends once more fall fairly close to the curve for the fractions when \( DP_n \) is taken as the independent variable. Again, however, the results for the series \( DP_n \)'s 502+43 (open circles) fall slightly above the curve for the fractions.
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2. ADDITIVITY OF MECHANICAL PROPERTIES

The results described above demonstrate that the mechanical properties of blends are more closely related to $\overline{DP}_n$ than to $\overline{DP}_w$. It can be shown that this closer relationship between mechanical properties and $\overline{DP}_n$ is consistent with two factors: the shape of the curve relating mechanical properties to $DP$ for the fractions, and the fact that the mechanical properties of a blend are the weight-averages of the properties of their components (eq 1). The second point will be considered first.

Examination of the tensile strength data for the blends reveals that if a strength of approximately 260 kg/cm$^2$ is assigned to the fraction of lowest $DP$, for which no precise experimental value could be obtained, the strength of each blend is the weight-average of the strengths of its components. This is shown in figure 7, in which the observed strength of each blend has been plotted against the value calculated (according to eq 1) from the weight percentages and strengths of the fractions of which it is composed. A line of 45-degree slope passing through the origin is included in the figure for comparison. The one blend which departs seriously from the line (observed strength, 0; calculated strength, 310) was a 95:5 mixture by weight of the fractions of shortest and longest chain lengths, respectively. This blend, like the fraction of lowest $DP$, was too brittle to give a film suitable for testing.

A similar relationship between the observed and calculated ultimate elongations of the blends can also be demonstrated, but because of the lower precision of these results, the agreement between the observed and calculated values is not so good as for the tensile strength measurements. The folding endurances of all the blends except those contain-
ing the fraction of smallest chain length ($DP_n = 43$) are also approximately additive. For the latter blends, the folding endurance values are additive only if the fraction with a $DP_n$ of 43 is assumed to have a negative folding endurance. This somewhat unreasonable result is, however, consistent with the shape of the curve relating folding endurance to $DP_n$ for the fractions (fig. 3), as extrapolation of this curve to a $DP_n$ of 43 would lead to a negative folding endurance.

It is now of interest to examine the consequences of applying the concept of additivity of mechanical properties to curves relating such properties to chain length for fractionated materials. For this purpose, the curve shown in figure 8 was chosen. It should be emphasized, however, that the results described below would be qualitatively similar if any of the experimental curves of figures 1 to 3 were used in this discussion. Figure 8 is a theoretical curve calculated from the equations of Sullivan [10], relating strength and fiber length of yarns which are perfectly homogeneous with respect to fiber

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**Figure 7.**—Observed tensile strengths of blends of cellulose acetate fractions as a function of strengths calculated according to assumption that strengths of fractions are additive (eq 1).

The $45^\circ$ line is included for reference. For complete description, see text.

**Figure 8.**—Theoretical curve, relating strength to $DP$ for perfectly homogeneous polymers.
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length. It differs qualitatively from typical experimental curves only in that it passes through the origin. As suggested above, however, the failure of the experimental strength-$DP$ curves to pass through the origin may well result from technical difficulties in film preparation and testing. In any case, this difference does not seriously affect the conclusions drawn below.

Figure 9 shows the strengths of blends, calculated from the curve of figure 8, assuming additivity of strength. As in the experimental curves, the strengths have been plotted on both weight- and number-average bases. Results for four series of blends have been plotted, representing varying proportions of the following pairs: $4+10$, $2.5+10$, $1+10$, and $.01+10$. The numbers have been so chosen that the first three of these approximately represent the three series of blends studied experimentally, and the last is roughly equivalent to blends of the fraction of highest $DP$ and a monomer. As shown by the upper graph of the figure, when the results are plotted on a weight-average basis, the calculated strengths of the blends fall on straight lines below the curve for the fractions. The extent of the departure from the curve for the fractions increases with the disparity between the chain lengths of the components in a pair. In contrast, when the results are plotted on the number-average basis, the curves for the blends of pairs $4+10$ and $2.5+10$ fall directly on the curve for the fractions. The curve for the pair $1+10$, which corresponds to the

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*According to Sullivan's analysis [10], to the left of the broken line in figure 8, yarn breakage occurs solely by slippage of the fibers. As the strength is contributed solely by frictional forces in this range, it is directly proportional to fiber length. To the right of the broken line, the frictional forces are sufficiently large to cause some fiber breakage, and the strength approaches asymptotically to a maximum value at which all fibers break. The analogy between frictional forces between fibers in a yarn and interchain forces in a film or filament is fairly close, but it is not suggested that the breaking in tension of a film can be explained in terms of this analysis. Thus, the breaking of a film includes a complex process of flow and crystallization, for which there is no counterpart in a yarn. The two processes do, however, contain a number of interesting points of similarity.
experimental blends of the longest and shortest chain lengths, falls slightly above the curve for the fractions, as did the open circles in the lower graphs of figures 4, 5, and 6. The curve for the pair \(0.01+0.1\), however, falls far above the curve for the fractions, which result is also in accord with experience [3]. Thus, these curves, which merely represent a statement of the weight-additivity of the strength, reproduce all of the features of the experimental data. It is perhaps worth noting that the series 1+10 (and the corresponding experimental series, as well) contains blends considerably more heterogeneous than the common unplasticized commercial polymers. Thus, some of the blends in this series have a ratio of \(\frac{D_P}{D_P} n\) higher than 3, as compared with a value of about 2 for most ordinary polymers. Even for this series, however, the mechanical properties are much more closely related to \(D_P n\) than to \(D_P w\).

The discussion above provides only a limited and qualitative approach to the problem of the mechanical properties of blends as related to the properties of their components. Further, the conclusions should be taken as applying principally to strength properties, rather than to the more complicated properties such as ultimate elongation and folding endurance. It is felt, however, that the concept of additivity of mechanical properties should prove helpful in some types of practical work, and may provide a basis for a more detailed analysis of the problem.

III. CONCLUSION

The results described above indicate the importance of \(D_P n\) in considering the mechanical properties of linear polymolecular polymers. It has been shown that the interdependence of mechanical properties and \(D_P n\) over a considerable range of chain lengths can be explained in terms of the weight-additivity of mechanical properties, and the shape of the curves relating these properties and the \(DP\) for fractionated materials. The results suggest that further attention be given to the use of osmotic pressure methods and other methods for determining \(D_P n\) in industrial work.

IV. REFERENCES


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