

# Powder Flow Studies III: Tensile Strength, Consolidation Ratio, Flow Rate, and Capsule-Filling-Weight Variation Relationships

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**Abstract** □ The tensile strength of consolidated powder beds was studied by applying a series of loads to the surface of the powder beds in a tensile tester. The results were plotted as tensile strength *versus* consolidation pressure. The linearity of these plots suggests a direct relationship between tensile strength and consolidation pressure. The following plots gave linear relationships: (a) tensile strength *versus* consolidation ratio, (b) tensile strength *versus* coefficient of variation of the filled weight of the capsules, and (c) logarithm of the tensile strength *versus* logarithm of the flow rate. These results suggest a direct relationship between tensile strength and consolidation ratio and their usefulness in studying powder flow. The physical significance of the empirical equation used in consolidation studies was explored. A comparison of the empirical equation with a theoretically derived equation, under certain assumptions, suggests that the consolidation ratio is a function of the ratio of the initial volume to the net volume and a function of the coefficient of Rankine. The coefficient of Rankine is a function of the angle of internal friction in the static powder bed.

**Keyphrases** □ Tensile strength—powder flow behavior, relationship to consolidation ratio □ Powder flow—tensile strength, consolidation ratio relationships □ Consolidation ratio—powder flow behavior, relationship to tensile strength

The knowledge of powder behavior is important in understanding the powder properties during storage, transportation, mixing, granulating, capsule filling, and tableting. Tensile strength, defined as the force per unit area of a broken face required to split a powder compact, provides useful information on powder behavior. The effects of powder density, particle-size distribution, and interparticle forces were considered in developing a theory of the tensile strength of powders (1).

Theoretical developments in studying powder flow must make far-reaching simplifying assumptions because of the numerous parameters involved, such as size, shape, and surface texture. These developments, although interesting, are of limited practical utility.

## BACKGROUND

Recent studies (2, 3) reported the consolidation of loosely packed powders and powder mixtures in cylindrical containers by applying a series of loads. The results of simple measurements in which the decrease of powder volume was measured as a function of the compressive load were plotted as the logarithm of the change in volume *versus* the logarithm of the applied pressure. The slopes of these plots were similar, and the intercept (consolidation ratio) gave a linear relationship with the logarithm of the flow rate and the coefficient of variation of the filled weight of capsules produced on an automatic capsule-filling machine.

Simple consolidation measurements and the use of an empirical equation suggested a good relationship to flow properties of the powders and powder mixtures. A literature survey indicated a paucity of any relationship between powder flow behavior and tensile strength.

This paper reports linear relationships between (a) tensile strength and consolidation ratio, (b) tensile strength and coefficient of variation of the filled weight of capsules, and (c) logarithm of the tensile strength and logarithm of the flow rate. These results suggest a direct relationship between tensile strength and consolidation ratio and their usefulness in studying powder flow. A comparison of the empirical equation with a

theoretically derived equation, under certain assumptions, suggests that the consolidation ratio is a function of the ratio of the initial volume to the net volume and a function of the coefficient of Rankine. The coefficient of Rankine is a function of the angle of internal friction in the static powder bed.

## EXPERIMENTAL

**Materials**—The drug<sup>1</sup>, tromethamine salt of (±)-2-benzoyl-1-azabicyclo[3.3.0]octa-2,4-diene-6-carboxylic acid, was at least 99% pure. The excipients were spray-dried lactose USP<sup>2</sup>, starch USP<sup>3</sup>, and magnesium stearate USP<sup>4</sup>.

**Mixing**—The powders of each formulation were mixed by the geometric dilution method on a piece of glassine paper. The powder mixture was screened through a 40-mesh screen to ensure proper mixing and to avoid powder agglomeration due to compaction. Three types of formulations were used. Drug and spray-dried lactose were mixed in the indicated proportions in the first formulation. The second formulation contained 0.5% magnesium stearate in addition to the drug and spray-dried lactose. The final formulation contained 10% starch, 0.5% magnesium stearate, spray-dried lactose, and drug in the indicated proportions. Powder mixing and all tensile strength measurements were carried out at 24 ± 1° and 40 ± 5% relative humidity.

**Tensile Strength**—The tensile strength of powder mixtures was determined by conducting a tensile test with an instrument<sup>5</sup> consisting of a main unit, an operational amplifier, and an x-y recorder. The main unit included four major components: (a) a constant-speed motor for exerting a pulling force through a steel string to break the powder bed, (b) a cylindrical powder cell with one half fixed to the main unit and the other half movable by suspension onto a metal frame, (c) a strain gauge of 200-g force for the quantitative measurement of tensile stress required to break the powder bed, and (d) a linear, variable differential transformer for the measurement of powder bed displacement.

The operational amplifier contained a strain amplifier, a displacement meter, and an operational unit. The latter was designed to indicate only the actual load by subtracting the necessary blank load that varied with displacement.

Blank tests were run to ensure that there was no extra load between the two half-cells before the powder was loaded. An accurately weighed amount of the powder was charged evenly through the cell extension tube, and the compaction weight was inserted gently into this tube. After 20 sec, additional weights, from 100 g to 2 kg, were loaded, with 20 sec between each loading. The weights and the cell extension tube were unloaded after 5 min. A steady-state condition in the powder bed thus was reached. The pulling string connecting the powder cell through a strain gauge was tightened under suitable tension before the clamp of the two half-cells was released. Then the constant-speed motor switch was turned on to pull the movable half-cell.

During the breaking process, the x-y recorder automatically recorded the tensile force profile over the total displacement of the two half powder beds. The peak value on the vertical axis of the tensile force-displacement curve gave the maximum tensile force of the powder. The area of breakage was normally 5 cm<sup>2</sup>, provided the compaction weight descended to the proper position. Otherwise, the area was corrected for the powder bed thickness difference. The tensile strength of the powder bed was obtained by dividing the maximum tensile force with the area of powder bed breakage. Three determinations were made, and the mean and the standard deviation were calculated. Only results obtained from the tensile

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<sup>5</sup> Cohetester, Hosokawa Micromeritics Laboratory, Osaka, Japan.

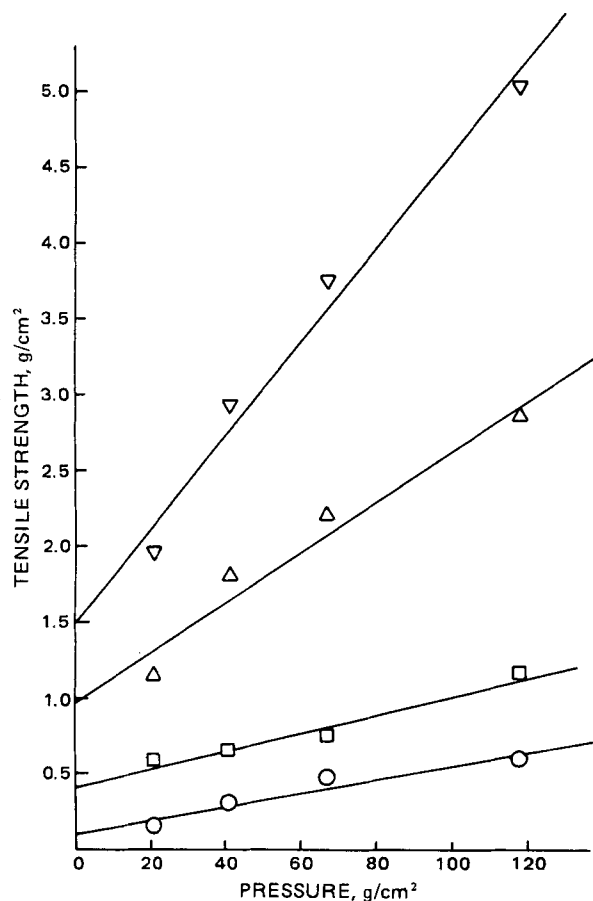
**Table I—Tensile Strength of Powder Mixtures Containing Drug and Excipients at Various Consolidation Pressures**

Percent Drug	Pressure, g/cm <sup>2</sup>	Tensile Strength, g/cm <sup>2</sup>	Slope	Intercept	Correlation Coefficient, <i>r</i>
89.5	20.99	1.95 ± 0.026	0.03078	1.507	0.9897
	41.36	2.92 ± 0.11			
	66.84	3.74 ± 0.07			
	117.78	5.02 ± 0.32			
40	20.99	1.14 ± 0.098	0.0167	0.9686	0.9752
	41.36	1.81 ± 0.17			
	66.84	2.20 ± 0.08			
	117.78	2.85 ± 0.06			
20	20.99	0.58 ± 0.01	0.006122	0.4020	0.9776
	41.36	0.64 ± 0.06			
	66.84	0.74 ± 0.06			
	117.78	1.16 ± 0.07			
10	20.99	0.15 ± 0.04	0.00443	0.1039	0.9609
	41.36	0.30 ± 0.08			
	66.84	0.47 ± 0.05			
	117.78	0.59 ± 0.08			

fracture where the powder bed broke into two halves along the joint of the two half-cells were recorded.

### RESULTS AND DISCUSSION

The tensile strength results of powder mixtures containing 10, 20, 40, and 89.5% drug at various consolidation pressures are given in Table I and Fig. 1. The excipients in these powder mixtures were 10% starch, 0.5% magnesium stearate, and spray-dried lactose to make 100%. The results indicate that the tensile strength is related linearly to the consolidation pressure in the pressure range studied. The dimensionless slope of these lines may be regarded as material constant, which is a measure of the increase in cohesive forces between adjacent particles due to consolidation. The intercept indicates the cohesive forces in an unconsolidated powder bulk. For free-flowing powders, the intercept is zero.



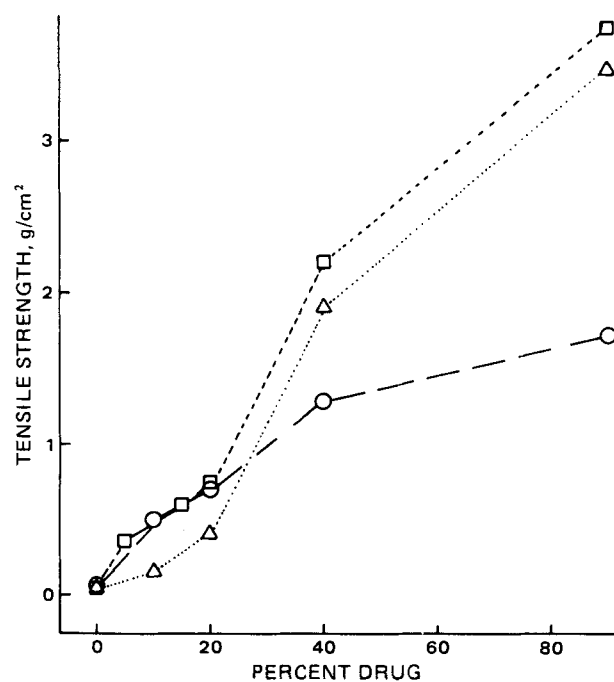
**Figure 1—Tensile strength of powder mixtures versus consolidation pressure plots showing linear relationships. Key: ○, 10% drug; □, 20% drug; △, 40% drug; and ▽, 89.5% drug.**

The profiles of the tensile strength *versus* percent drug at a constant consolidation pressure (66.84 g/cm<sup>2</sup>) are given in Fig. 2. The tensile strength of the mixtures containing 10 and 20% drug, lactose, and magnesium stearate was lower than that of mixtures containing drug, lactose, starch, and magnesium stearate and of mixtures containing drug and lactose. At higher drug concentrations, the tensile strength of the mixtures containing starch and magnesium stearate was higher than that of the mixtures containing only drug and lactose.

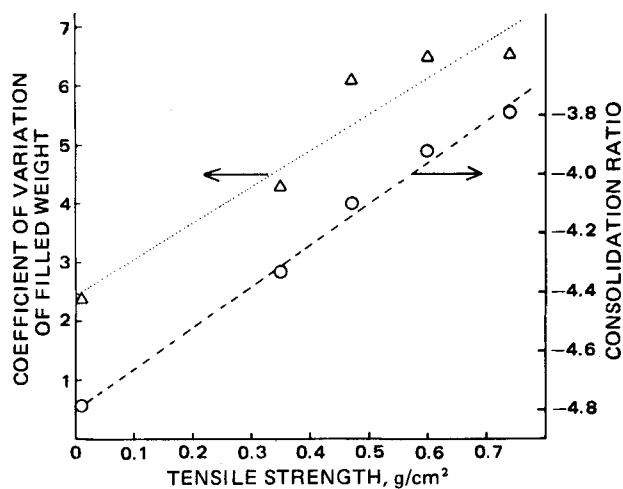
The consolidation ratio and the coefficient of variation of the filled weight of capsules reported previously (2) were plotted *versus* tensile strength (Fig. 3). The results indicate linear relationships between tensile strength and consolidation ratio and between tensile strength and coefficient of variation of the filled weight of the capsules. The logarithm of the flow rate reported earlier (3) and the logarithm of the tensile strength are plotted in Fig. 4. These results suggest that the tensile strength and consolidation ratio provide useful information of powder flow behavior.

The powder consolidation ratio was obtained (2) from the intercepts of the relative volume *versus* consolidation pressure plots according to:

$$\ln \frac{V_0 - V}{V} = m \ln \frac{P}{P_0} + I \quad (\text{Eq. 1})$$



**Figure 2—Profiles of the tensile strength as a function of the formulation composition. Key: ○, drug and lactose mixture; △, drug, lactose, and 0.5% magnesium stearate mixture; and □, drug, lactose, 10% starch, and 0.5% magnesium stearate mixture.**



**Figure 3**—Plots of the tensile strength versus consolidation ratio and coefficient of variation of the filled weight of the capsules. Key: O,  $r = 0.9964$ ; and  $\Delta$ ,  $r = 0.9583$ .

where  $V_0$  is the initial powder volume;  $V$  is the powder volume at a given surface pressure,  $P$ ;  $P_0$  is the surface pressure  $P$  (1 atmospheric pressure approximately equals  $1 \text{ kg/cm}^2$ ); and  $m$  and  $I$  are constants.

By assuming  $I = \ln C$ , dividing both sides by  $V_0$ , and rearranging, Eq. 1 can be written as:

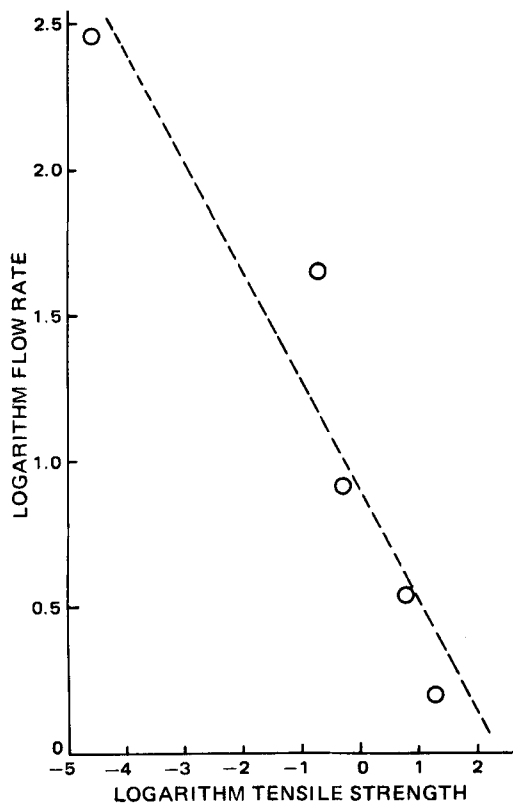
$$\frac{1}{V} - \frac{1}{V_0} = \frac{C}{V_0} \left(\frac{P}{P_0}\right)^m \quad (\text{Eq. 2})$$

Multiplying both sides by  $V_\infty$ , adding 1 to both sides, and rearranging give:

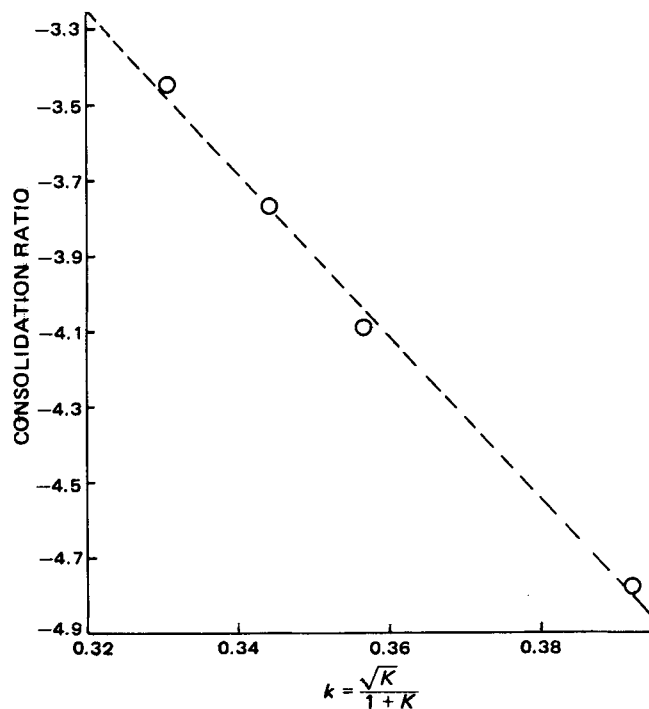
$$\frac{V - V_\infty}{V} = \frac{V_0 - V_\infty}{V_0} - \frac{CV_\infty}{V_0} \left(\frac{P}{P_0}\right)^m \quad (\text{Eq. 3})$$

where  $V_\infty$  is the net volume of the powder.

Since porosity  $n$  equals  $(V - V_\infty)/V$  and the powder bed is assumed to be homogeneous, Eq. 3 can be written:



**Figure 4**—Logarithm tensile strength versus logarithm flow rate showing a linear relationship ( $r = 0.9523$ ).

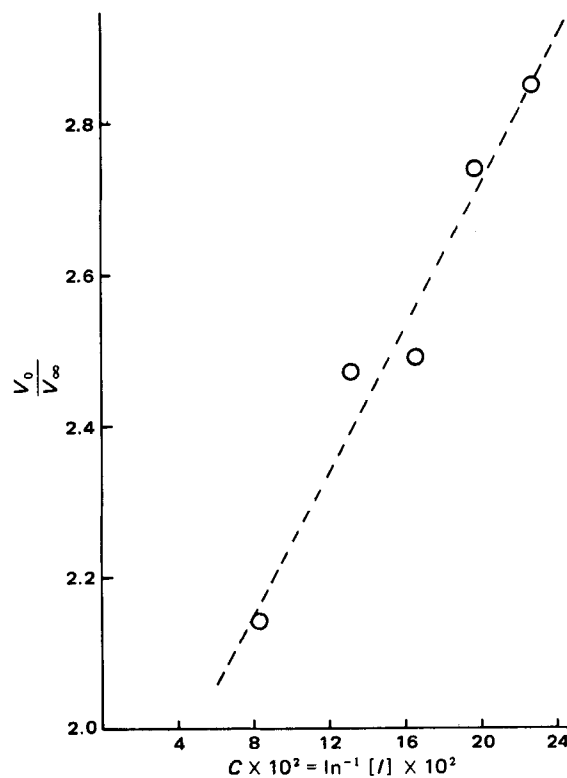


**Figure 5**—Plots of consolidation on ratio versus  $k$  showing a linear relationship ( $r = -0.9980$ ).

$$n - n_0 = -\frac{CV_\infty}{V_0} \left(\frac{P}{P_0}\right)^m \quad (\text{Eq. 4})$$

Theories dealing with the pressure and volume change relationship have been reported in the field of powder technology. Kawakita and Ludde (4) obtained a theoretical compression equation using Mohr's circle of stress:

$$\Delta\phi = \frac{\sqrt{K} d\sigma_1}{1 + K \sigma_1} \quad (\text{Eq. 5})$$



**Figure 6**—Plots of  $V_0/V_\infty$  versus  $C$  showing a linear relationship ( $r = 0.9764$ ).

**Table II—Values of  $\phi$ ,  $K$ ,  $k$ , and  $I$**

Drug in Mixture, %	$\phi$	$K$	$k$	$I$
40	48.6	0.1428	0.3307	-3.45
20	46.5	0.1592	0.3442	-3.77
10	44.5	0.1758	0.3566	-4.09
0	38.4	0.2337	0.3918	-4.78

where  $\sigma_1$  is the maximum principal stress,  $\phi$  is the angle of internal friction, and  $K$  is the coefficient of Rankine (5) as given by:

$$K = \frac{1 - \sin \phi}{1 + \sin \phi} \quad (\text{Eq. 6})$$

Thus,  $\Delta\phi$  represents the probability of a packing change of the particles and is considered to be proportional to some quantities related to the state of packing in one of the several described manners. It was shown that various compression equations could be derived by making different assumptions about the properties that determine  $\Delta\phi$ .

If  $\Delta\phi$  is proportional to the decrease in porosity and inversely proportional to the  $m$ th power of consolidation pressure,  $-dn/P^m$ , then:

$$-\frac{dn}{P^m} = k \frac{dP}{P} \quad (\text{Eq. 7})$$

where  $P$  is the consolidation pressure,  $m$  is a constant, and  $k$  is a function of the coefficient of Rankine.

Equation 7, on rearrangement, gives:

$$-dn = kP^{m-1} dP \quad (\text{Eq. 8})$$

Integration of Eq. 8 from initial porosity  $n_0$  to porosity  $n$ , corresponding to the zero initial consolidation pressure to the consolidation pressure  $P$ , gives:

$$n - n_0 = -\frac{k}{m} P^m \quad (\text{Eq. 9})$$

Equation 9 is similar to Eq. 4, which is the modified form of Eq. 1. Equations 4 and 9 suggest that  $C$  is a function of  $(V/V_\infty)(k/m)$ .

A previous study (2) showed that, for different powder mixtures of the same drug,  $m$  was a constant. The value  $k$  is a function of the coefficient of Rankine  $K$  given by Eq. 6, and  $\phi$  may be approximated from the angle of repose.

For the powder mixtures containing drug, spray-dried lactose, starch, and magnesium stearate, the values of  $\phi$ ,  $K$ ,  $k$ , and  $\ln C$  are given in Table II. A plot of  $C$  versus  $k$  gave a linear relationship with  $r = -0.9666$ . Figure 5 gives a plot of  $\ln C$  versus  $k$ . The excellent correlation ( $r = -0.998$ ) indicates that  $\ln C$  is inversely proportional to  $k$ .

Figure 6 gives a plot of  $V_0/V_\infty$  versus  $C$  for the same powder mixture. A linear relationship ( $r = 0.9809$ ) between  $V_0/V_\infty$  versus  $C$  suggests that these parameters are directly related.

The results of this study suggest that the consolidation ratio is a function of tensile strength and that both parameters are useful in studying flow behavior of powders and powder mixtures. The physical significance of the consolidation ratio was explored. The consolidation ratio is a function of the ratio of the initial volume to the net volume of the powder and of the coefficient of Rankine, which, in turn, is a function of the angle of internal friction in the static powder bed.

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# Quantitation of Solvent Polarity Based on Molecular Structure

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**Abstract** □ A numerical index is proposed that ranks solvents according to their polarity. It is based entirely on structure, encoding the relative content of exterojacent electrons in the molecule. The index is the first-order valence molecular connectivity index,  $^1\chi^v$ . The index is modified for the number of isolated functional groups in the molecule. A comparison with solvent polarity indexes based on several experimental methods reveals a good relationship. The polarity index proposed can be quickly calculated, it does not depend on the availability of the actual molecule, and it permits prediction of solvent polarity or the polarity of mixtures.

**Keyphrases** □ Solvent polarity—quantitative method based on molecular structure □ Polarity index—solvents, quantitative method based on molecular structure □ Molecular structure—quantitative method for ranking solvent polarity

The term "solvent polarity" is widely used by chemists to characterize the observed manifestations of intermolecular interactions in a solution process. The processes may be chromatography, dissolution, or chemical reactions. The manifestations of the interactions are recorded as the separation or retention on stationary phases, the solubility, and the partitioning or reaction course and rate.

Solvent polarity is a comparative term which the chemist uses intuitively to rank commonly employed compounds. This intuition, based on experience, permits the generalization that hydrocarbons are less polar than esters while alcohols are more polar than esters of the same molecular weight.

## BACKGROUND

The quantitation of solvent polarity is difficult in practice due to the variety of intermolecular forces that may operate between molecules in a given system. Much has been written about solution theory, but the forces influencing solvent-solute interactions can be summarized as those due to dispersion and dipolar and electron donor-acceptor properties (1).

Dispersion forces account for virtually all of the attractive energy between nonbonded atoms among hydrocarbons. These compounds are regarded as nonpolar when their solvent properties are characterized. Dipolar and electron donor-acceptor interactions influence the attractive energy among molecules possessing unsaturation or atoms other than carbon or hydrogen. Solvents possessing structures capable of these interactions are characterized as being polar. Thus, molecular structure governs solvent polarity, but this relationship has yet to be quantified in any simple way.