JOURNAL OF APPLIED POLYMER SCIENCE VOL. 8, PP. 25-35 (1964)

Dilatation of Granular Filled Elastomers under High Rates of Strain

R. J. FARRIS, *Aerojet-General Corporation, Sacramento, California*

Synopsis

An apparatus has been developed for measuring the dilatation of specimens tested in uniaxial tension in a constant volume chamber by determining the change in the volume of air around the specimen through precise measurement of air pressure. The dilatometer can be used on standard testing machines and has shown reproducible values at testing speeds from 0.66 to 666%/min., and temperatures from −75 to 210°F. Typical stress strain curves are shown with the simultaneously recorded dilatation versus strain.

A. INTRODUCTION

1. Significance of Measurement

Hank Thompson electrified the baseball world with his homer in 1951. The impact of Smith's dewetting studies[1] has been no less dramatic upon scientists engaged in mechanical properties studies of elastomeric materials containing filler particles. In the scant four years since Smith's paper appeared in the *Transactions of the Society of Rheology*, dewetting has become an accepted byword in solid propellant research.

Interest in the nature and significance of dewetting, however, has been severely restricted by the limited and often inadequate means available for assessing or measuring this phenomenon. This paper describes a newly developed instrument, a gas dilatometer, which has been designed specifically for assessing the dewetting process from dilatational properties. This instrument is relatively inexpensive, and versatile. It is operable over wide ranges of temperatures and strain rates, may be hydrostatically pressurized, and provides precise measurements. It may be used independently or can be automated as an accessory to existing testing equipment. It is readily adaptable for either routine or research testing.

2. Nature of Dewetting

The stress-strain properties of granular filled elastomers beyond low strains are complicated by a multiplicity of factors, of which "dewetting" (i.e., failure of the filler-elastomer bond) is perhaps the most important. Time and temperature dependencies of the dewetted and undewetted composite material and the dewetting phenomenon itself complicate the overall behavior and interpretation of physical properties measurements.

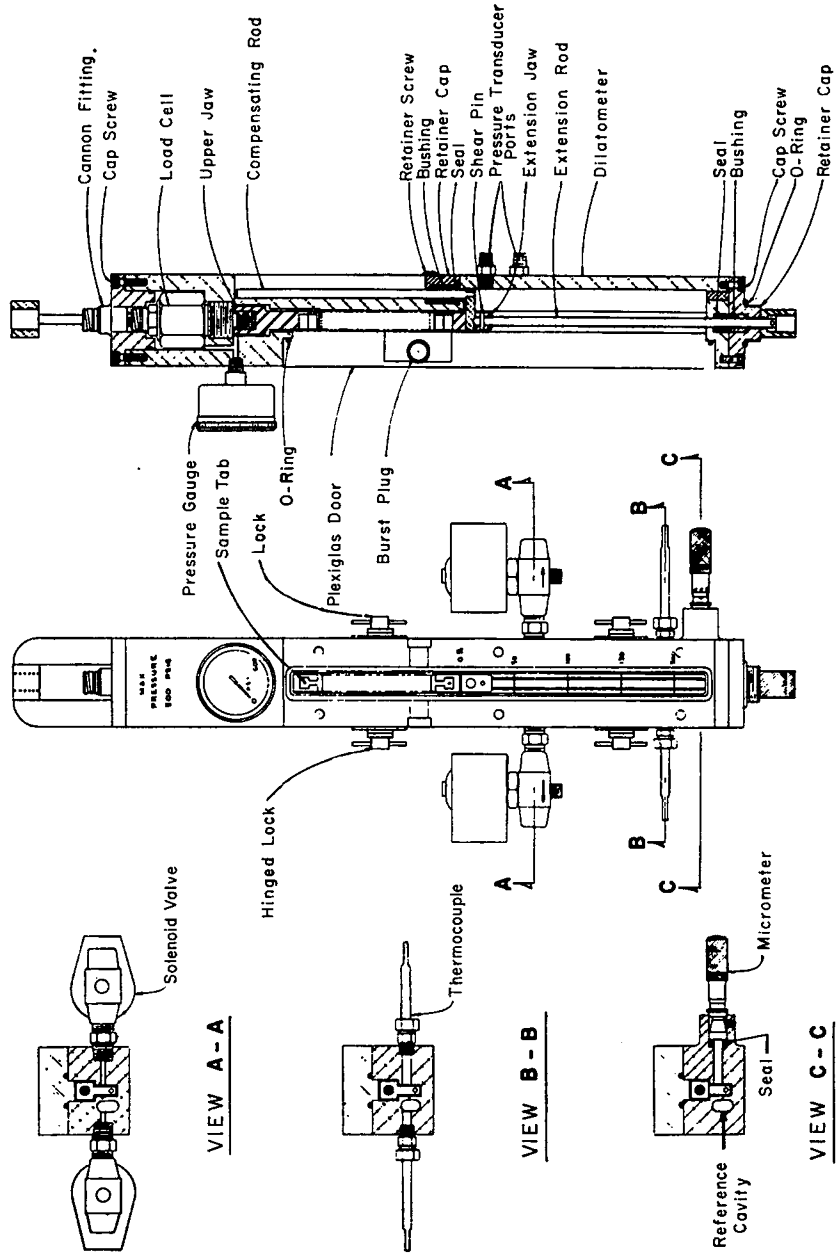

Fig. 1. Detail of gas dilatometer.

The dewetting phenomenon produces, in these materials, small elliptical vacuoles about the dewetted filler particles. These vacuoles enlarge and propagate with additional strain application with their major axis being parallel to the axis of straining.

Dilatation in granular filled elastomers is essentially produced by vacuole formation. The granular fillers in these materials undergo no real straining because they are of a high modulus and are contained in a weak rubber matrix. If these particles undergo no straining, they cannot dilate.

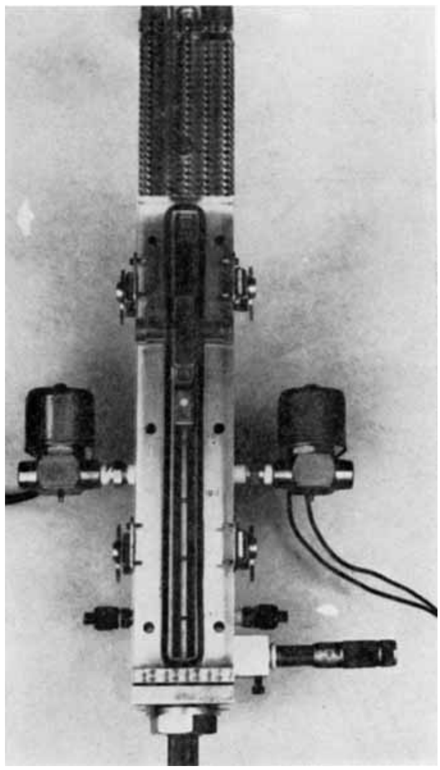

Fig. 2. Front view of gas dilatometer.

The rubber that comprises the filler containing matrix is, for all practical purposes, incompressible and therefore does not dilate. Therefore, dilatation results from the filler-elastomer failure and in some cases from elastomer-elastomer cavitation (cohesive failure).

Dewetting in these materials greatly influences their stress-strain behavior as well as other physical properties. Tools for these measurements have been, however, both cumbersome and the data has been minimal. As there is no direct measure of dewetting, one must therefore measure a byproduct of dewetting. Of the byproducts, dilatation is the most open avenue of approach for it is solely induced from vacuole formation.

3. Survey of Existing Test Equipment

Early experimental methods for determining the volumetric change[1] in granular filled elastomers under static strain conditions were conducted by the straining of samples in a fluid medium and determining volumetric change by capillary tube fluid displacement. More recently an experimental method using a gravimetric buoyancy technique was developed[2]

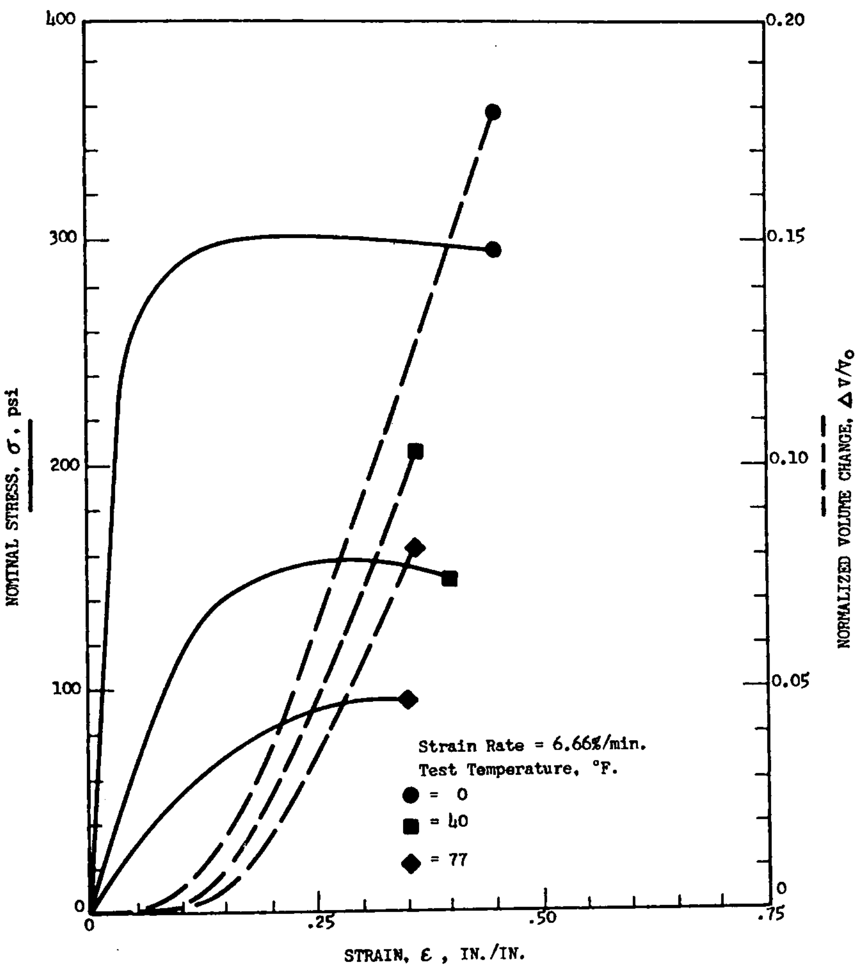

Fig. 3. Effect of temperature on the stress-strain-dilatation behavior of a granular filled elastomer.

which may be best described as a series of incremental strains applied to a sample with the density change calculated for each increment. Although accuracy is quite precise by this method, the excessive time involved and data unavailable between increments of strain dictated the design of an alternate experimental apparatus that would not only lend itself to high rate production testing, but continuously record all measured parameters under dynamic stress loading conditions.

B. APPARATUS

As an accessory to the existing Instron Tensile Testers, but not limited to this specific tester, a gas dilatometer, designed for the simultaneous and continuous measurement of the volume change (vacuole content) of propellant and its stress-strain characteristics, was recently made operational at Aerojet-General Corporation.

Basic to insight into the logic of the system is an understanding of the process involving interpretation of differential pressures as volume changes. To assist in the discussion, a schematic representation is shown in Figure 1 and a photograph of the operational instrument is shown in Figure 2.

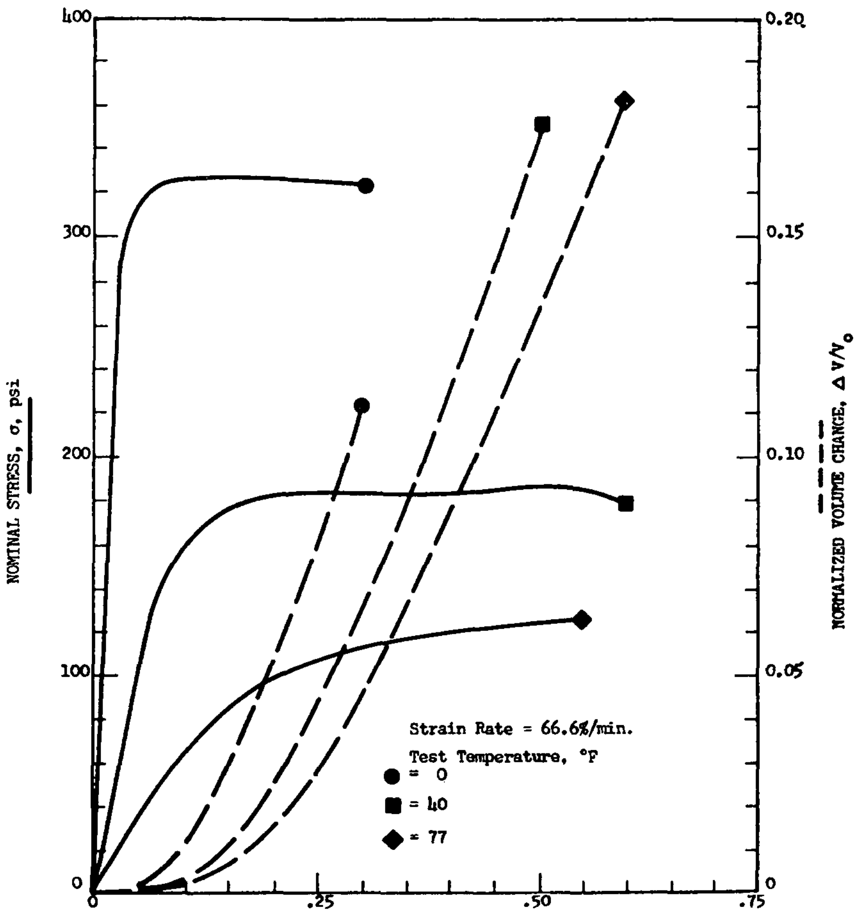

Fig. 4. Effect of temperature on the stress-strain-dilatation behavior of a granular filled elastomer.

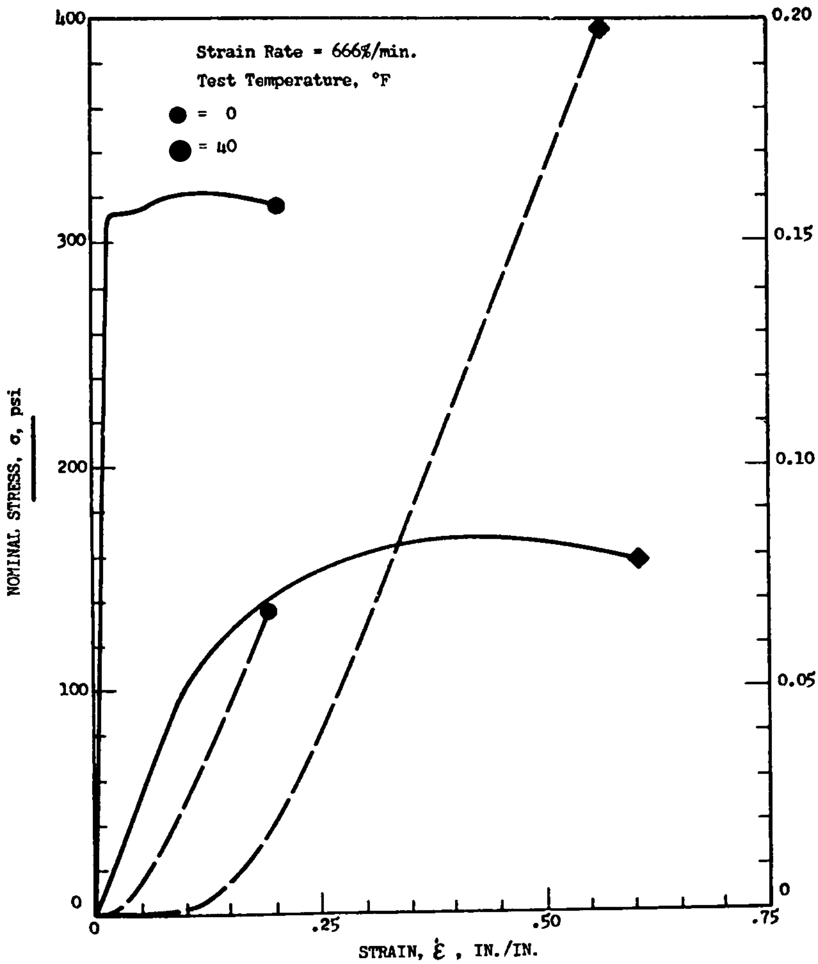

Fig. 5. Effect of temperature on the stress-strain-dilatation behavior of a granular filled elastomer.

The instrument has an aluminum body with two cavities, a test cavity and a pressure reference cavity. A hinged Plexiglass door with an O-ring seal and swivel locks provides access to the test cavity where an end bonded specimen, (0.5 × 0.5 × 3.0 in. in dimension) is fitted into upper and lower sample holder jaws. The upper jaw is mounted to a force transducers that is completely contained within the test cavity thus providing accurate force measurements. The lower jaw is mounted on a precision machined rod which passes out of the body through a bushing and seal. As the rod moves out of the cavity to extend the specimen, an identical rod, yoke mounted to the extension rod, enters the test cavity through another bushing and seal to compensate for the volume change caused by the extension rod leaving the test cavity, thus providing an extension mechanism without

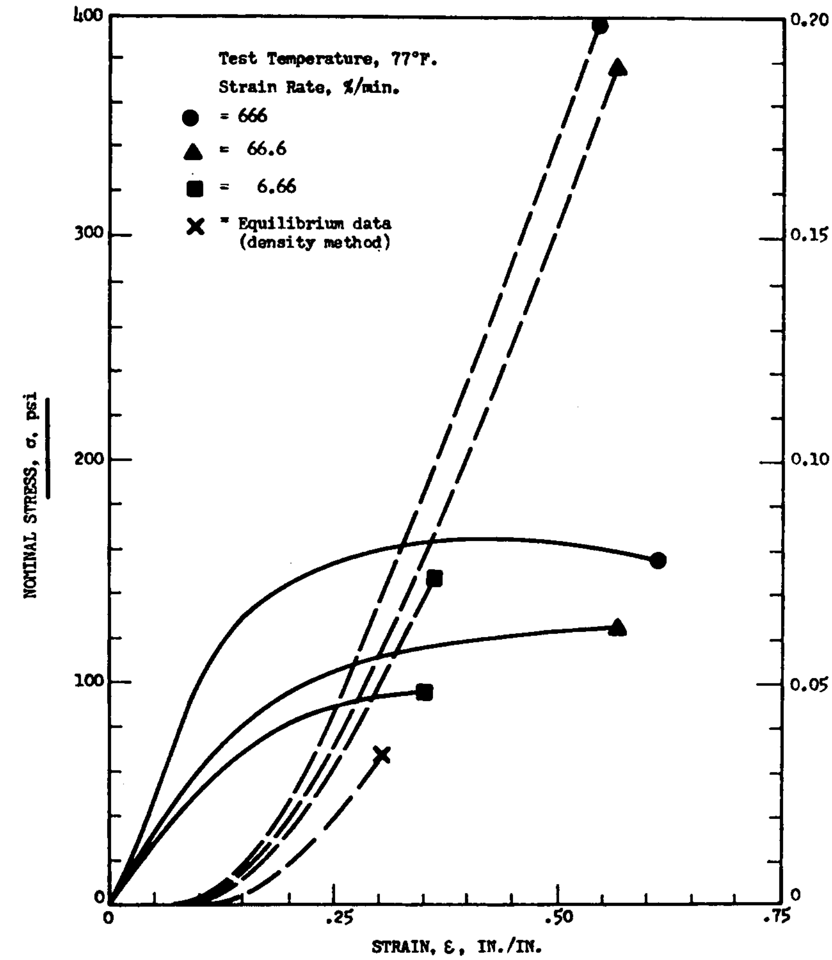

Fig. 6. Effect of strain rate on the stress-strain-dilatation behavior of a granular filled elastomer.

changing the volume of the test cavity. The pressure reference cavity, being of constant volume, is separated from the test cavity by a fine differential pressure transducer. When the specimen is extended, any change in specimen volume will be reflected in an opposite but equal change in air volume in the test cavity, resulting in a pressure change between the test and reference cavities.

In the testing of propellant under hydrostatic pressure, the Plexiglass door is replaced by an aluminum door and the low range differential pressure transducer (±.5 psia) is replaced by one capable of measuring small pressure differentials (±.15 psia) when used with high line pressures. Solenoid valves provide simultaneous pressurization and pressure equalization of the test and pressure reference cavities after the specimen is placed in the test cavity and the door bolted in place. The measurement technique is identical as described previously except for the change in base pressure. The instrument was designed for operation to 500 psi pressure and may easily be extended by building a more rigid case. A force transducer, of a vented type, is totally contained in the test cavity, making it free from pressure effects.

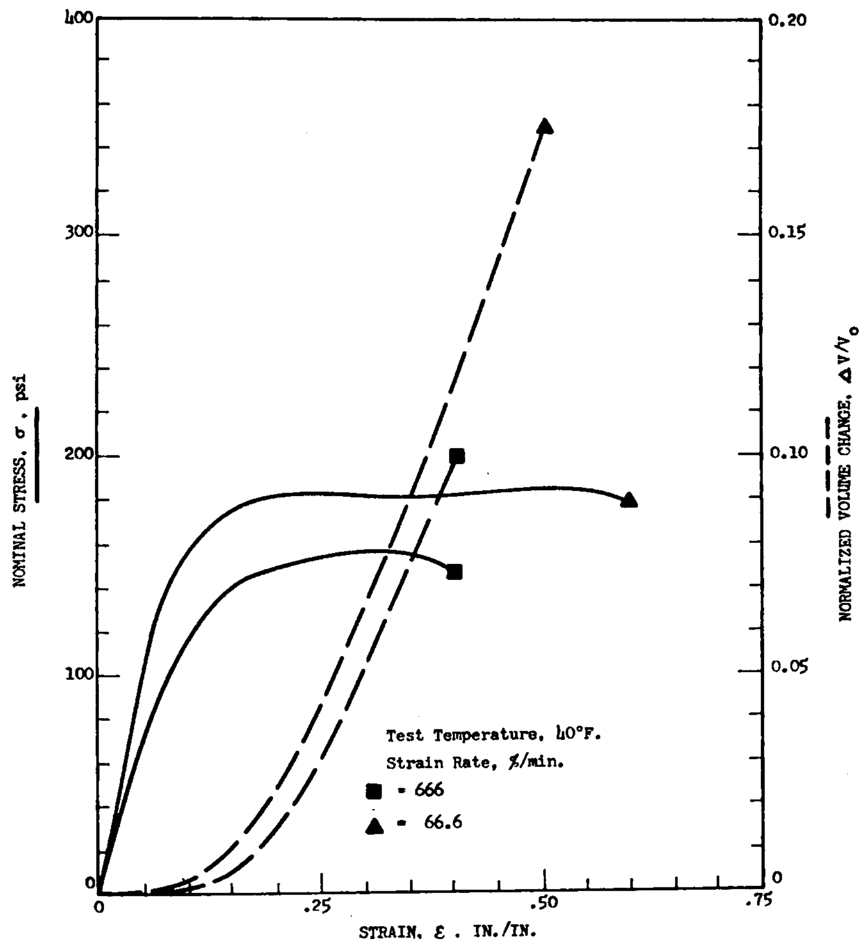

Fig. 7. Effect of strain rate on the stress-strain-dilatation behavior of a granular filled elastomer.

The gas behavior within the test cavity is isothermal at extension rates up to 200 in./min. because the aluminum is an excellent conductor and the cavity surface to volume ratio is high. This was verified experimentally by forcing volume changes equal to specimen strain which would be the most extreme case possible. Under these circumstances Poisson's ratio would be equal to zero. There are also no significant thermal contributions from the specimen during straining since these materials show no recorded transient temperatures.

In order to maintain conditions even at relatively high strain rates, without requiring very elaborate temperature conditioning facilities, the instrument body was voluminously designed so as to effect an excellent heat

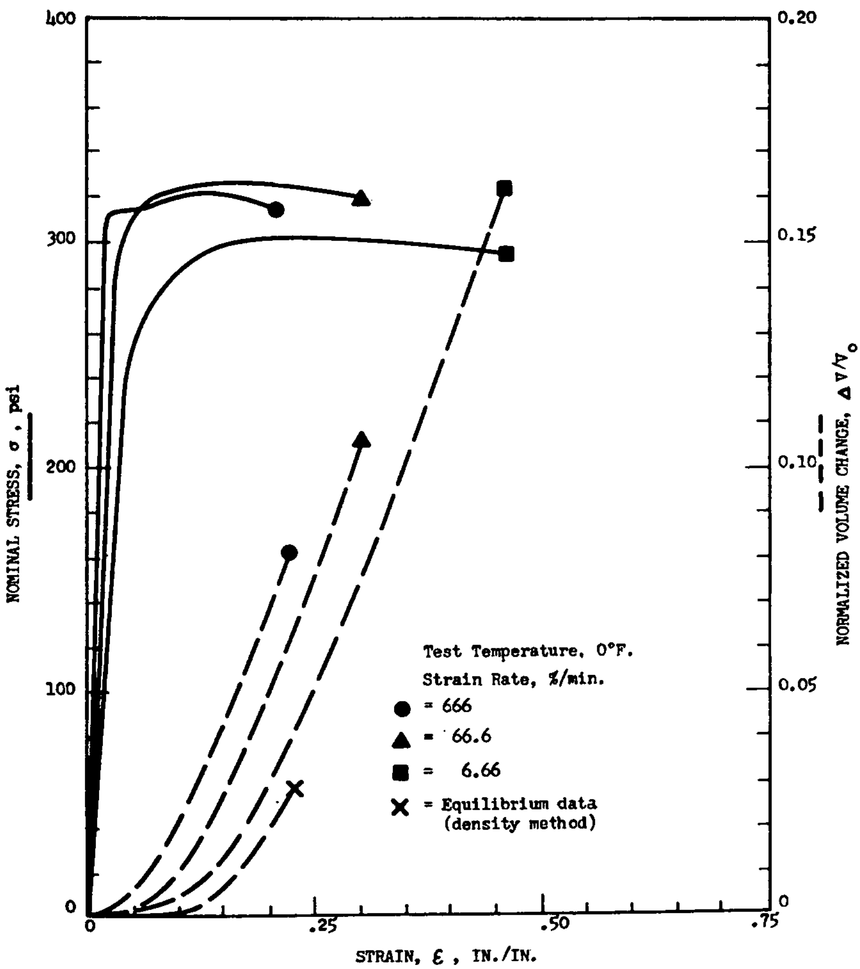

Fig. 8. Effect of strain rate on the stress-strain-dilatation behavior of a granular filled elastomer.

sink. The elimination of the effects of all pressure changes resulting from a temperature differential between the test and reference cavities, and compensation for adiabatic gas behavior at high strain rates were made by connecting two high speed thermocouples in a differential analog circuit with the differential pressure transducer continuously compensating the pressure signal for changes in gas temperature.

Present operating limits of the apparatus are strain rates between .66 to 66,600%/min. (estimated) with temperature range of −75–+210°F. and operating pressures between .1 and 500 psia.

C. MEASUREMENTS

Typical stress-strain-dilatation measurements of a granular filled elastomer (73% by volume filler and 27% elastomer) are shown in Figures 3 through 8. Measurements were made at temperatures from 0 to 77°F. and at strain rates from 6.66 to 666%/min.

Figures 3, 4, and 5 illustrate the effect of temperature on the stress-strain-dilatation behavior at constant-strain rate. Increasing modulus and stress with decreasing temperature is characteristic of these materials. The increase in dilatation with decreasing temperature is also characteristic as the particles are being subjected to higher stresses at all strains.

Figures 6, 7, and 8 illustrate the effect of strain rate at constant temperature on the stress-strain-dilatation properties. It is evident that the effect of strain rate is similar to that of temperature which is probably due more to the viscous behavior of the composite material than that of the elastomer itself.

Equilibrium dilatation data taken by a buoyancy density technique was available for this material. These data are presented as equilibrium data on the figures. These equilibrium dilatation measurements are in agreement with those data taken at higher strain rates.

D. PHENOMENOLOGICAL OBSERVATIONS

It is evident that most irregularities in the stress-strain behavior of granular filled elastomers is due to dilatation which results from the rupturing of filler-binder bonds and in some cases, cavitation within the binder. The latter results when the cohesive properties of the binder are less than the adhesive properties of the filler-binder interface bond. The true stress-strain behavior is essentially linear until dilatation commences, at which time the material begins to lose the structural support of the filler and the stress-strain curve deviates downward from linearity. This transition in stress-strain behavior is induced by dewetting.

The observed transition areas of the stress-strain-dilatation behavior is not common only to filled elastomers, but to all materials for which these data were available. Steel undergoes a transition from elastic to plastic deformation at yield. In the elastic region, the dilatation and stress are linear with strain and in the plastic region it is well documented that the plastic deformation is deformation with no additional dilatation.[3] Linear elastic and linear viscoelastic materials do not exhibit transition areas in either stress or dilatation with respect to strain as expected, for this is the definition of a linear material. Rubbers that tend to crystalize with straining show an increasing modulus as crystalization occurs with a negative dilatation[4] (density increase). The behavior of soils[5] is remarkably similar to granular filled elastomers and could be considered as an elastomeric system with an extremely weak matrix binding the crystaline materials.

One of the difficulties in analyzing test data on new materials, especially composite materials, is in determining its behavior type. Simple guides to rank materials in several behavior categories may be developed from the stress-strain-dilatation properties of the material.

E. CONCLUSION

It is felt that this instrument should be particularly valuable to researchers engaged in the fundamental and applied studies in the behavior of elastomeric materials. Its versatility, wide range of test conditions, relative ease in operation, and its simplicity coupled with its precision characterize it as an excellent laboratory tool.

References

1. Smith, T. L., *Trans. Soc. Rheol.*, **3**, 113 (1959).
2. Svob, G. J., P. C. Colodny, L. A. Waddle, and T. B. Lefferdink, *Volume Changes of Polyurethane Propellants Subjected to Small Strains*, Joint Army-Navy-Air Force-ARPA-NASA Panel on Physical Properties of Solid Propellants, **I**, October 1961, pp. 295–306.
3. MacGregor, G. W., *J. Appl. Mech.*, **6**, 1939.
4. Mandelkern, L., *Chem. Rev.*, **56**, 903 (1956).
5. Rowe, P. W., The Stress-Dilatancy Relationship for Static Equilibrium of an Assembly of Particles in Contact.

Résumé

On a construit un appareil pour mesurer la dilatation d'échantillons soumis à une tension uniaxiale dans une chambre à volume constant. Cette mesure se fait en déterminant le changement dans le volume de l'air autour de l'échantillon au moyen de mesures précises de la pression de l'air. Le dilatomètre peut-être employé sur des machines d'essais standards et donne des valeurs reproductibles à des vitesses d'essais variant de 0.66 à 666%/min.; et à des températures de −75 à 210°F. On présente les courbes typiques tension-élongation ainsi que l'enrégistrement simulténé de la dilatation en fonction de l'élongation.

Zusammenfassung

Es wurde ein Apparat zur Messung der Dilatation von Proben entwickelt, die mit uniaxialer Spannung in einer Kammer konstanten Volumens durch Bestimmung der Änderung des Volumens der die Probe umgebenden Luft durch genaue Luftdruckmessung getestet wurden. Das Dilatometer kann an Standardtestmaschinen benützt werden und zeigt reproduzierbare Werte bei Testgeschwindigkeiten von 0,66 bis 666%/min und Temperaturen von −75° bis 210°F. Typische Spannungs-Dehnungskurven und gleichzeitig aufgenommene Dilatation-Belastungskurven werden mitgeteilt.