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TECHNICAL REPORT

By

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EFFECT OF SEVERAL GAS ATMOSPHERES ON HYDRAULIC FLUID SHEAR STABILITY

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ABSTRACT

The sonic-shear stability of polyisobutylene and polymethacrylate solutions in oil was examined under several gases and at pressures up to five atmospheres. Tests were conducted in a pressurized sonic treatment cup under nitrogen, hydrogen and helium atmospheres. Comparison tests under air were made at one atmosphere.

Results indicated that sonic-shear stability varied inversely with gas solubility for all the gases, except air, at one atmosphere. Both polymer solutions displayed highest viscosity losses under the helium atmosphere. The use of gases other than air and sonic test cell pressurization showed promise as a modification of the sonic-shear method, to make it more closely reflect mechanical-shear tests.

FOREWORD

The work reported here was performed under DA Project No. 1C024401A108, AMS Code 5025.11.802 on Power Transmission Fluids under Work Unit Title, "Effect of Contaminants on the Shear Stability of Hydraulic Fluids." It was carried out for the purpose of examining the effects of sonic test cell atmospheres other than air on the shear stability of polymer-thickened oils.

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PROBLEM

To examine the shear stability of polymer-thickened fluids under several gas atmospheres and at pressures greater than atmospheric.

BACKGROUND

Investigations into use of the sonic oscillator for determination of shear stability of hydraulic fluids showed that sonic and mechanical shear test methods did not agree.(1) This disagreement had been attributed to differences in response of different chemical types of polymers to different types of shear tests. To find the cause of such discrepancies several factors have been investigated.

The first factor examined was the difference in temperature between the sonic and R.I.A. pump tests.(1) Thereafter, other factors examined were particulate and water contamination in the sheared oils.(2) None of these were found to contribute to the discrepancies between the test methods.

Another test method difference was found in the air pressure variations between the sonic and R.I.A. pump tests. The latter test imposes a 25 psig. dry air pressure upon the surface of the fluid-to-be-sheared prior to its passage through the pump and capillary. In contrast the sonic test cell is operated at 0 psig. (atmospheric pressure).

This difference in gas pressure above the fluid was seen to have two possible effects: (1) More dissolved gas would be available in the oil, in the R.I.A. pump test, to participate in turbulence-cavitation phenomena at the areas of shearing, and (2) The atmospheric pressure employed in the sonic tests would, of course, not cause additional gas to be dissolved, but in addition, the sonic-induced cavitation in the oil would expell most of the dissolved gas from the test liquid.

Dissolved gas can participate in cavitation phenomena. In an ultrasonic fluid, Boyle, et.al.(3) have shown that bubbles of gas are formed by the union of microscopically small bubbles originally present in the liquid. The smaller bubbles are driven towards the nodes of a sound wave. Then too, the expansions of liquid in the wave give rise to negative pressures within the liquid. Boyle and Taylor(4) relate this phenomena to cavitation wherein formation of "hollows" in the liquid is forced by the applied ultrasonic field. Dissolved gases will collect in these hollow areas. Sorensen(5) further found that the power required is reduced as applied ultrasonic frequency is lowered. Since, in the R.I.A. sonic oscillator, the sound frequency (10 Kc) used is much lower, it can be expected that the sonic-shear test will efficiently, and rapidly coalesce dissolved gas into bubbles. Cavitation is also affected by the external gas pressure applied to the liquid. Bondy and Sollner(6) indicated that this applied pressure must not exceed a "critical" value if there is to be degassing. However, cavitation can still occur even when no gas is present, as in a degassed liquid. In this case, much smaller bubbles will be formed but will be, according to Fitch, et.al.(7) filled with vapor of the liquid in which cavitation is occurring. In the present case, most polymer-thickened hydraulic fluids major constituent is a light base oil. This oil should be able, even in absence of any gas, to provide sufficient vapor to form bubbles under cavitational stress.

In a more practical sense, many hydraulic systems reservoirs are pressurized in order to maintain a ready supply of oil at the pump inlet. This reservoir pressurization will, of course, increase the amount of gas dissolved in the oil, which in turn, could affect cavitation phenomena occurring at other parts of the system, for example, pump and orifice components.

To examine some of the factors related to dissolved gas effects; and to determine whether such effects were related to sonic-pump shear test differences; tests were conducted in a sonic unit modified for control of the test cell atmosphere.

APPROACH

Polymer-in-oil solutions were sheared in a sonic oscillator at several pressure levels under several pure gases. Viscosity loss was used to measure the shearing activity.

Materials Used

Fifteen percent by weight solutions of linear polyisobutylene and polymethacrylate polymers were dissolved in a light base oil (free of additives) such as is used for formulating fluids defined by Military Specification MIL-H-5606B.

The molecular weight of the polyisobutylene was 44,000; that of the polymethacrylate was 92,000; while the base oil viscosities at 100°F and 210°F, respectively, were 3.62 and 1.33 centistokes. Prior to testing, the polymer solutions were filtered through 0.45 and 0.20 micron pore-sized membrane filter papers in series, using a pressure filtration funnel.

The test cell containing the test oil was filled with nitrogen, hydrogen or helium gas, which was dried by passing it over prepared silica gel.

Equipment Used

A Raytheon Model DF 101, 250-watt magnetostrictive oscillator, operating at 10 Kc, was the basic unit. It was operated according to a proposed ASTM Method.(8) It is illustrated in Figures 1 and 2 along with modifications that were made.

Illustrated in Figure 1 is the oscillator unit (O) which feeds electromagnetic vibrations into the treatment cup (TC) shown in Figure 2. Also shown in Figure 1 are the modifications: (CC) polarizing current control switch and (CM) polarizing current meter used to monitor this current. The treatment cup in Figure 2 consists of the basic test cell (TC), whose lid was modified to permit passage of pressurized gas into or out of the closed cup. The gas pressure gage (M) and valves (V_1 and V_2) used for this purpose are also shown. Not shown is a cylinder containing dried silica gel, through which the test gas was passed to remove any moisture. The test gas was then introduced into the treatment cup which contained the oil to be sheared. The entire bottom of the treatment cup was connected to a rod which, when caused to vibrate by high frequency current in its surrounding coil, in turn, radiated sonic vibrations into the treatment cup.

Procedure Used

Prior to each days operation the unit was run for 40 minutes at full power (1.5 amps.) to warm up the unit. The sonic treatment cup was maintained at 100°F ± 10°F by an adjacent source of temperature controlled water. The treatment cup was twice cleaned with petrolene and benzene, dried and 15 ml ± 0.5 ml of test oil added to the cup. The lid was then affixed, the gas connections made and gas was

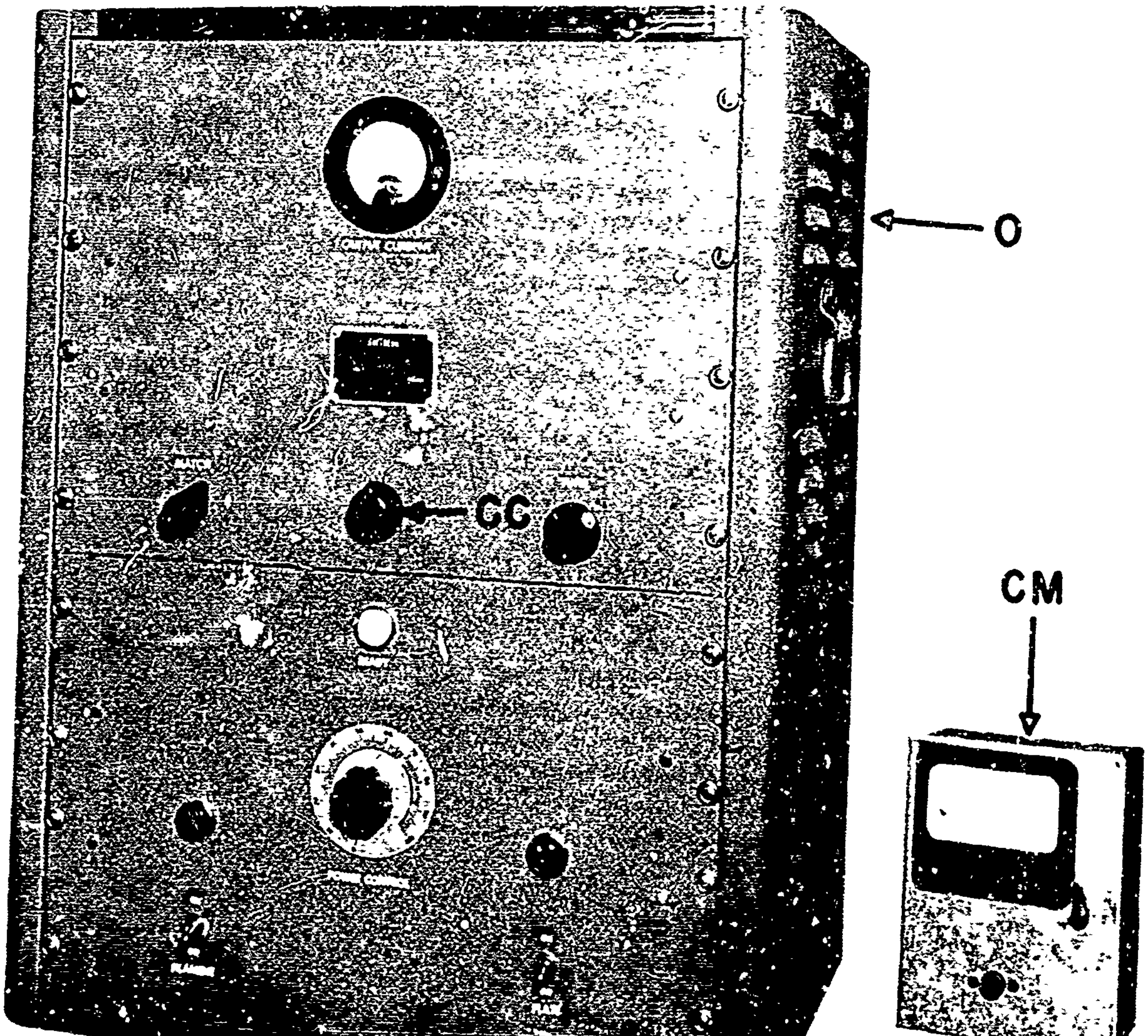

FIGURE 1 SONIC OSCILLATOR

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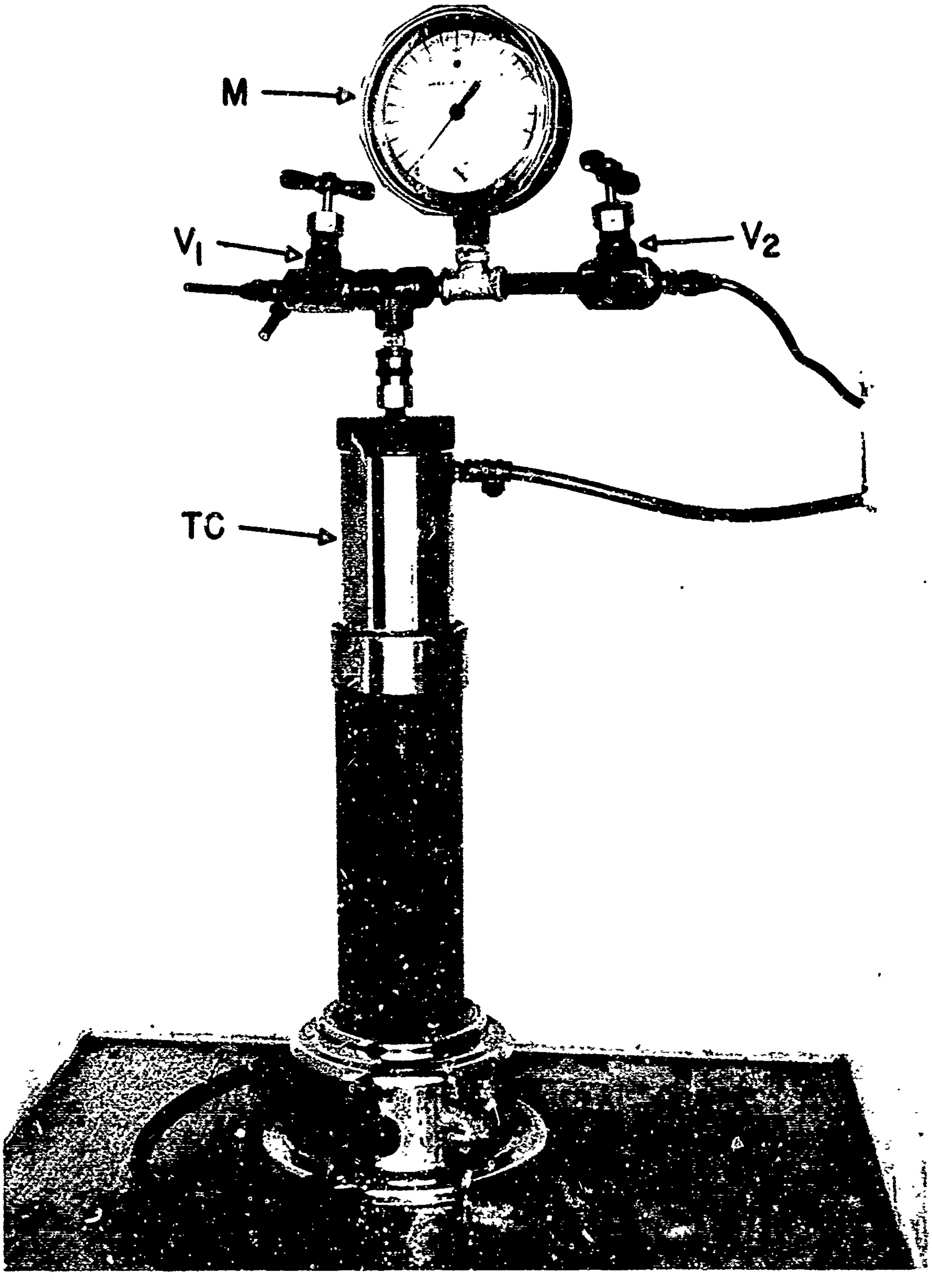

FIGURE 2   PRESSURIZED SONIC TREATMENT CUP

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passed into the cup slowly until a pressure of 50 psig. was reached. Then the system was purged slowly. This addition and purging of gas was repeated six times to remove the residual air in the cup. After the sixth addition of gas, the system was allowed to stand 5-10 minutes to check for leaks (while at 50 psig.). After the last purge, the system was again allowed to stand 5 minutes at a pressure selected from one of the following test pressures:

1 atmosphere - 0 psig.

2 atmospheres - 14.7 psig.

3 atmospheres - 29.4 psig.

4 atmospheres - 44.1 psig.

5 atmospheres - 58.8 psig.

The sonic oscillator was then started and operated under the following conditions:

Time = 40 ± 1 minutes.

Power = 1.0 ± 0.05 amps.

Polarizing Current = 7.4 ± 1 amp.

At the end of the sonic treatment, the system was slowly purged of gas, the lid removed, and the kinematic viscosity of the oil determined at $100^{\circ}F$.

The percent viscosity loss was determined according to the following formula:

$$\% \text{ loss} = \frac{(\text{Orig. - final) viscosity}}{\text{Orig. viscosity}} \times 100.$$

RESULTS

Percent viscosity loss data are shown in Table I. The data indicated both polymers under helium at atmospheric pressure showed a greater viscosity loss than under air. This is also true for the polyisobutylene solution under hydrogen. In all cases, the amount of viscosity loss decreased as gas pressure is increased. This decrease was

TABLE I

GAS AND PRESSURE EFFECTS ON SHEAR-INDUCED VISCOSITY LOSSES OF POLYISOBUTYLENE AND POLYMETHACRYLATE SOLUTIONS

Pressure (Atmospheres)	Polymer	Mean Viscosity Loss(1)					
		Polyisobutylene(2)			Polymethacrylate(3)		
	Gas	N_2	H_2	He	N_2	H_2	He
1		13.3	21.3	29.3	8.2	10.5	14.2
2		9.3	14.3	22.4	6.3	7.2	9.2
3		7.8	11.4	15.6	4.2	4.8	6.6
4		5.1	9.4	11.6	1.3	1.8	2.0
5		1.4	6.4	7.8	0.6	0.7	1.0

(1) Mean of Duplicates (Population Standard Deviation = 1.75; 95% Confidence Limits = ± 2.5%).

(2) Under air at 1 atm. = 20.2% viscosity loss.

(3) Under air at 1 atm. = 11.7% viscosity loss.

about equally rapid for both polymer solutions. The fact that the isobutylene solutions losses were higher at 5 atmospheres pressure, reflects their higher initial breakdown at 1 atmosphere.

There was visual evidence of excess gas dissolved in all the tests at pressures above 1 atmosphere. The oil samples foamed upon removal from the sonic treatment cup and during the first one or two runs in the viscosity tube. Viscosity values were not recorded until after the foaming activity had ceased.

DISCUSSION

The data at one atmosphere for the two polymers under the various gases show considerably different reactions when compared with the results under an air atmosphere in the sonic test cell. Referring to the viscosity losses for the solutions tested under nitrogen, both the polyisobutylene and the polymethacrylate solutions showed decreased shear attack. Under hydrogen, both types of solutions showed viscosity losses nearly equal to that under air, while under helium, however, both showed a higher level of breakdown when compared to tests under air.

These results appear to be anomolous in some respects. Thus, first, assuming gas density to be important, air and nitrogen should give similar behavior and hydrogen and helium would also have a similar effect. Second, assuming gas reactivity with free radicals formed during shear to be basic; different shear effects would be expected for air (the oxygen content of air could react with such free radicals) while similar behavior should be shown by the nitrogen, hydrogen and helium, since it is unlikely that any of these would engage in polymer-free radical reactions. Yet, tests under these gases showed different levels of shear breakdown. A third possible basis for these effects was the solubility of the gases in the oil. Typical solubility values taken from several sources are:

Air(8) = 0.11 ml. per 1 ml. of oil.

Nitrogen(9) = 0.09 ml. per 1 ml. of oil.

Hydrogen(9) = 0.05 ml. per 1 ml. of oil.

Helium(10) = 0.02 ml. per 1 ml. of oil.

Comparison of this data with viscosity losses in tests (see Table I) under nitrogen, hydrogen and helium indicated that the solutions percent viscosity loss varied inversely, approximately, with the square root of the gas solubility. However, this relationship did not hold for the air-atmosphere tests since a viscosity loss, lower than would be predicted, was found. Possibly, as observed by Henglein and Schulz,(11) oxygen (present in the air) reacted with polymer free radicals formed during shear and prevented their recombination with other polymer fragments. Since this could not occur under nitrogen, the result would be, comparatively, a larger viscosity loss under air that would also be greater than that predicted by gas solubility considerations. Other workers such as Jellinek(12) have theorized that degradation rate will decrease with gas solubility. Brett and Jellinek(13) found polystyrene-in-benzene degradation rates to be higher in tests under gases more soluble in the solution.

Referring to the high pressure tests, degradation was found to vary inversely with applied gas pressure. This was to be expected since more gas would be dissolved in the polymer media. Reduction of degradation (observed viscosity losses) proceeded rather uniformly as the pressure was increased. Under nitrogen, both polymers degradation was reduced to 0.6 to 1.5% at five atmospheres. Under hydrogen and helium, polymethacrylate solution viscosity loss was less than 2 percent at 4-5 atmospheres pressure. However, the polyisobutylene solution showed 6.4 to 7.8 percent losses even at 5 atmospheres; though this likely reflected the relatively higher viscosity losses noted at 1 atmosphere. In fact, the pressure increase produced a larger reduction in degradation than was found for the polymethacrylate solution. The data trend suggests that test pressures above 5 atmospheres would have caused the polyisobutylene degradation to approach zero in all cases. Higher pressures were not employed because it was suspected they would be hazardous in the sonic treatment cup used.

The observed effect of pressure on sonic degradation, though expected on the basis of dissolved gas considerations, can also be fitted to the observation of Hamann(14) who stated that sound absorption decreases with pressure. Thus, a pressure (or sound) wave after application to a liquid is followed by a fast contraction and a second slower contraction. The second, delayed contraction (derived from a potential barrier to molecular rearrangement) is said to be responsible for absorption of high-frequency sound. Superimposed pressures can reduce the amount of the delayed contraction, and alter sound absorption with, possibly, an attendant reduction on effects on shear-labile constituents in the fluid.

Referring to the problem of discrepancies between sonic and mechanical tests which had resulted from the fact that the sonic method favored polyisobutylenes over polymethacrylates shear-stability-wise, the test data indicated the comparative stability of the two polymers was somewhat affected. Table II shows the ratios of polyisobutylene to polymethacrylate percent viscosity losses. The ratios were somewhat larger under hydrogen and helium than under air at one atmosphere and indicated some correction of the sonic methods' deficiency. The large increases noted at 4 to 5 atmospheres mainly reflect that the polymethacrylate solutions' viscosity losses had approached zero. Generally, at the lower pressures, the polyisobutylene solutions' relative sensitivity to sonic-shear was increased most under helium and hydrogen gases.

TABLE II

COMPARISON OF VISCOSITY LOSSES FOR THE TWO POLYMER TYPES

Ratio of $\frac{\text{Polyisobutylene Visc. Loss}}{\text{Polymethacrylate Visc. Loss}}$

	Gas Pressure (atm.)				
Gas	1	2	3	4	5
Nitrogen	1.6	1.5	1.9	3.9	2.3
Hydrogen	2.0	2.0	2.4	5.2	9.1
Helium	2.0	2.4	2.4	5.8	7.8
Air	1.7	-	-	-	-

From the more practical point of view of sealed hydraulic systems for which helium is coming into increasing use as a pressurizing gas, this work suggested that the use of helium may be accompanied by more rapid shearing of polymer-thickened fluids in such systems.

CONCLUSIONS

The sonic-shear stability of polyisobutylene and polymethacrylate solutions in oil is influenced by the nature of the dissolved gases. For three of the gas atmospheres tested, the polymer degradation as reflected in solution viscosity loss, varied inversely approximately as the square root of the gas solubility.

Polymer shear was larger under helium than for any of the other gases tested.

The use of special gases and test cell pressurization showed some promise for causing sonic-shear tests to more closely reflect mechanical tests. Thus, polyisobutylene solutions were more severely degraded relative to polymethacrylates in several instances.

RECOMMENDATIONS

Sonic test modifications comprised of controlled test cell atmospheres under pressure are recommended as promising for causing sonic tests to reflect mechanical shear tests.

Helium should not be substituted for other gases for use as a pressurizing medium in hydraulic systems containing polymer-thickened fluids until its effect on the fluids' shear stability is established.

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