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Hollow Cylinder Sample Test Using A Dynamic Mechanical Analyser

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A hollow cylindrical specimen was designed for testing rubber in shear deformation using a Dynamic Mechanical Analyser series 7. Test results showed that a lower coefficient of variation for storage modulus is obtained with this system than using a compressive pellet sample in a cup-and-plate deformation. This shear test has been developed to characterise four grades of natural rubber: Standard Thai Rubber (STR) 5 L; STR 20; RSS and skim block. Over a frequency range of 0.1 Hz to 50.0 Hz, and 0.5% strain, the effect of rubber grade, temperature, degree of mastication and the amount of carbon black (N330) was investigated. Dynamic complex and real viscosities were determined and showed good agreement with the shear flow viscosity obtained from experiments with a Rosand capillary rheometer. However, limitations of this shear technique were found, such as a failure of test specimen at temperatures approaching the molten state of rubber, and insufficient force control for very stiff specimens with high carbon black loading.

There are a number of literature sources[1,2] on the dynamic properties of elastomers (natural and synthetic rubbers). These studies are important with respect to engineering applications involving dynamic forces on vulcanised materials. Most of the published work characterised rubber material as vulcanised solids over the range of glassy, transition and rubbery states. Only a few[2] studied the dynamic properties of rubber in the unvulcanised state or in the 'rubbery' region. Raw natural rubber is fed directly to a processing instrument, *e.g.* extrusion or injection moulding. During the process, characteristics of rubber in melt form is directly related to product properties. Therefore, a number of experiments[2] have been carried out on raw rubber in the molten state, such as testing with capillary rheometers or viscometers. Dynamic properties of raw rubber can also be referred to flow properties and they are widely tested in vibration between a cone-and-plate or a plate tool kit in torsion.

A Dynamic Mechanical Analyser series 7 (DMA 7) is the only dynamic tester available in Prince of Songkla University. It has become an important instrument to characterise rubber in unvulcanised form together with the other available instruments. The problem encountered while testing non-vulcanised rubber, loaded in dynamic force at high temperatures approaching the molten state led this study to develop a new configuration of test.

Figure 1a shows a sample held in a small cup and compressed by an upper plate, con-

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nected to a load cell. Edge effects are expected to give higher values of modulus than the expected compression value. This will be clearly seen from the experimental results as an extremely high value of measured modulus compared with shear test result. As we have only a dynamic tester, a hollow cylinder sample self-adhering to the walls of an annular cylindrical gap was designed (*Figure 1b*) in order to test a specimen in shear.

To confirm the results obtained from the DMA 7, dynamic viscosities were determined and compared with the shear flow viscosity at low shear rate region. The dynamic real and complex viscosities were calculated[8] from *Equations 1* and *2*.

$$\eta'(\omega) = \frac{E''}{\omega} \quad \ldots 1$$

$$\eta^*(\omega) = \sqrt{\left(\frac{E'}{\omega}\right)^2 + \left(\frac{E''}{\omega}\right)^2} \quad \ldots 2$$

where E' is the storage modulus at angular frequency ω
E'' is the loss modulus at angular frequency ω
η' is the real viscosity at angular frequency ω
η^* is the complex viscosity at angular frequency ω.

The flow viscosity, tested in the extrusion method using the Rosand capillary rheometer, was determined as the viscosity varying with shear rate, denoted as $\eta(\dot{\gamma})$. Based on the equivalent of the shear rate and the angular frequency ($\dot{\gamma} = \omega$), following Cox and Merz's assumption[5,6], the comparison was taken.

EXPERIMENTAL AND METHOD OF TESTING

The experiment was divided into two main parts as shown below.

Compression and Shear Tests

The aim of this experiment was to clarify an appropriate test method using the DMA 7. Testing raw natural rubber at temperatures just below the temperature of a molten state is

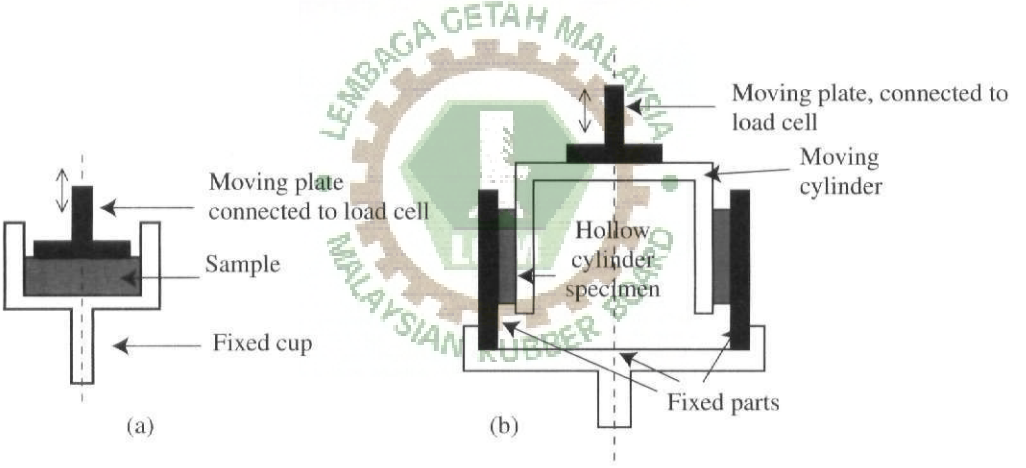


Figure 1. Sample geometry diagram tested in (a) compression between cup-and-plate and (b) shear as a hollow cylinder specimen.

recommended by using a cup-and-plate kit, an accessory kit of the machine, for a honey-like material testing. However, the preliminary study[3,4] showed sensitive data with a high coefficient of variation, as expected, because of the cup-and-plate used. Therefore, a hollow cylinder sample configuration was designed in order to obtain more consistent test data.

Comparison of Dynamic and Shear Flow Viscosity

In order to confirm the results from DMA 7 tested in shear mode, a comparison with another test method was made. The shear flow tested on the extrusion method using the Rosand capillary rheometer was used and compared with the calculated dynamic viscosities following the ideas of Cox and Merz[5,6] and Bueche[7].

Four grades of raw natural rubber were tested using hollow cylinder specimens. The aims of the tests were to investigate the effect of the degree of mastication, rubber grade, temperature, and fillers. The confidence in these results will be realised when it is in conformance with other test equipment.

EQUIPMENT

- Two-Roll mill – A commonly used mastication equipment having 25.4 cm of diameter, 50.8 cm length, and friction ratio of 1:1.25
- Mooney Viscometer – AC/684/FD model, SPRI Limited UK. The unit of measurement is indicated as Mooney Viscosity (MV)
- Dynamic Mechanical Analyser series 7 (DMA 7) – a Perkin Elmer machine
- Capillary Rheometer – a Rosand RH-710 single barrel model machine

RESULTS

Compression and Shear Tests

The sample tested in compression and shear modes were investigated. Small cut pieces of raw natural rubber were heated in a sample preparation equipment in order to mould a compression and shear sample. The sample was loaded in a cup, provided as an accessory kit, recommended for a honey-like material, *e.g.* raw natural rubber at temperatures approaching a molten state. The sample was compressed between the cup and a parallel plate connected to a force detector. The cup diameter used was about 6 mm and the approximate sample height was 2 mm *(Figure 1a)*. The hollow cylinder sample was self-attached to two thin metal cylinders *(Figure 1b)* (see *appendix A*). The approximate outer and inner diameters of this specimen were 16 mm and 12 mm respectively, and the sample height and thickness were around 6 mm and 2 mm, respectively. Both compression and shear tests were performed using Standard Thai Rubber (STR) 5 L in a frequency scan mode over the frequency range of 0.1 Hz to 50 Hz, 0.5% dynamic strain and at 100°C.

Figure 2 shows the storage modulus varying with angular frequency, ranging approximately from 0.6 rad/s to 300 rad/s. The number of samples tested in compression were much higher than those used in shear; *i.e.* 20 and 3, respectively. However, the variation of the compression data was not lower than that of shear mode. The average coefficient of variation of the storage modulus over the frequency range 0.1 Hz to 50 Hz for the compression and shear were approximately

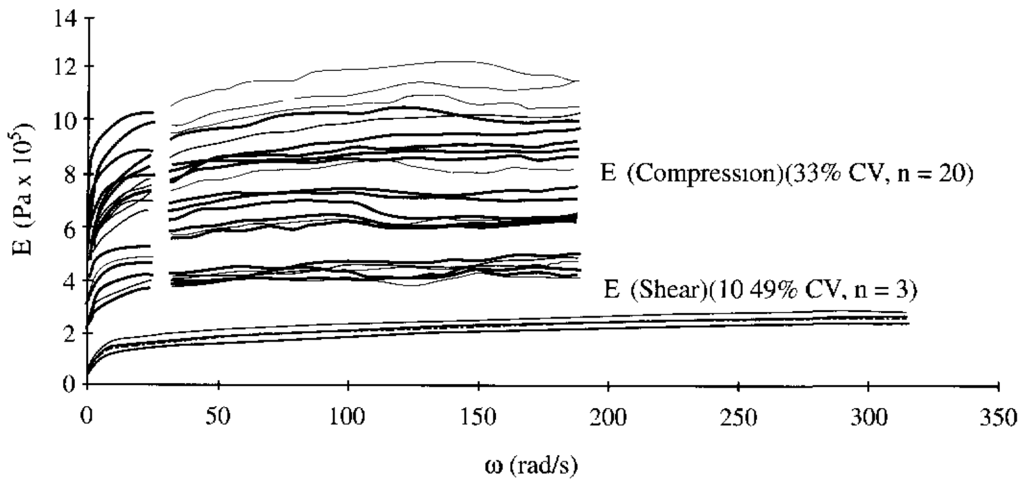


Figure 2 STR 5 L storage modulus tested in compression and shear (20 samples were tested in compression with 33 0% coefficient of variation, 3 samples were tested in shear with 10 49% coefficient of variation)

33% and 10%, respectively Moreover, the compression mode test showed a discontinuity at about 5 Hz, which could be a resonance frequency of the DMA 7 and the sample Therefore, the designed hollow cylinder specimen was suited to test raw natural rubber using the DMA 7 at a specific range of temperature.

Comparison of Dynamic Viscosities and Shear Flow Viscosity

Four grades of natural rubbers, STR 5 L, STR 20, RSS and skim block having a degree of mastication of 74, 85, and 80 MV, respectively, were tested at 100°C and 0 5% strain over the frequency scan range of 0 1 Hz to 50 Hz *Figure 3* show the influence of rubber grades in the variation of storage and loss moduli with angular frequency At frequencies higher than 5 Hz, the storage modulus of RSS, STR 20 and STR 5 L decreased in order. The skim block rubber shows the most rapid change in the storage modulus than the others, while all grades have the loss modulus decreasing in the order as follows. skim block, STR 20, RSS and STR 5 L

Dynamic storage and loss moduli of these four grades of raw natural rubber were calculated for complex and real viscosities using *Equation 1* and *2* and compared with the shear flow viscosity tested at 100°C from capillary rheometer The comparisons were plotted on log-log scale viscosity and angular frequency or shear rate and shown in *Figures 4a* to *4d* for STR 5 L (74 MV), STR 20 (85 MV), RSS (92 MV) and Skim block

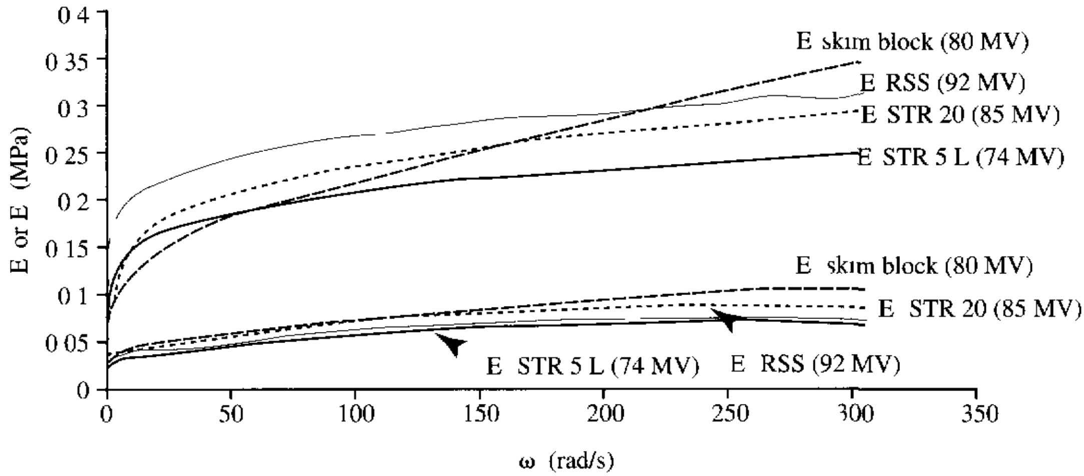


Figure 3 Storage and loss moduli of four grades of raw natural rubber tested in shear hollow cylinder at 100°C

(80 MV), respectively The results show that viscosity as a function of shear rate is nearly identical to the absolute value of the complex viscosity as a function of angular frequency, in accordance with a study by Cox and Merz's[5,6]

DISCUSSION AND CONCLUSION

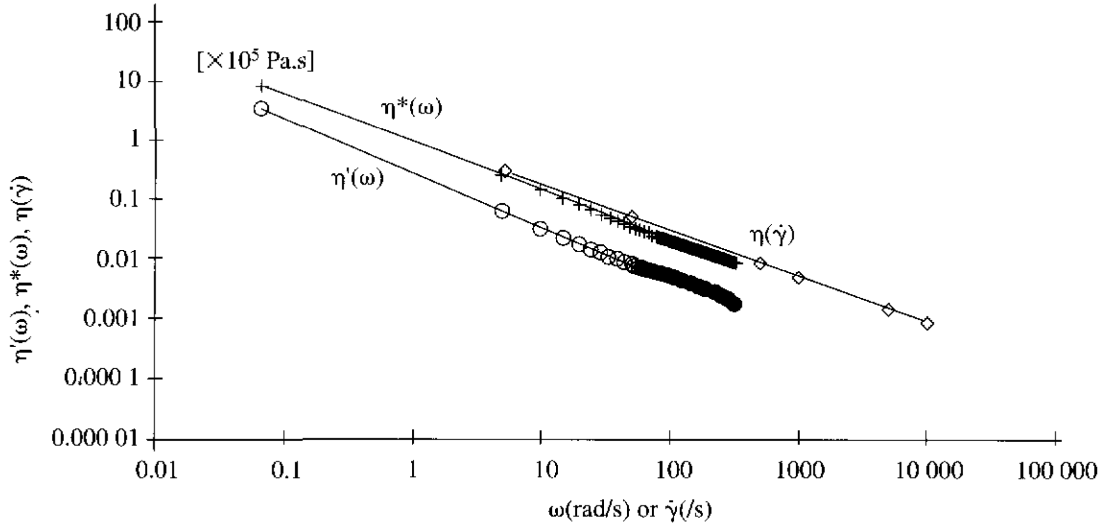
The study of dynamic properties of raw natural rubber using a hollow cylindrical shear sample on the DMA 7 found an appropriate test with limited range of sample stiffness and temperatures of testing Samples of overt high stiffness were out of the capability of the machine to perform an accurate test For materials of overt softness, due to a high content of non-reinforcing agent and testing material at temperatures approaching a molten temperature, a specimen no longer remained as a cylindrical shape

The dynamic properties of various grades of natural rubber tested within a certain temperature range were confirmed by comparison with the other test methods However, the influence of temperature and filler should be further studied.

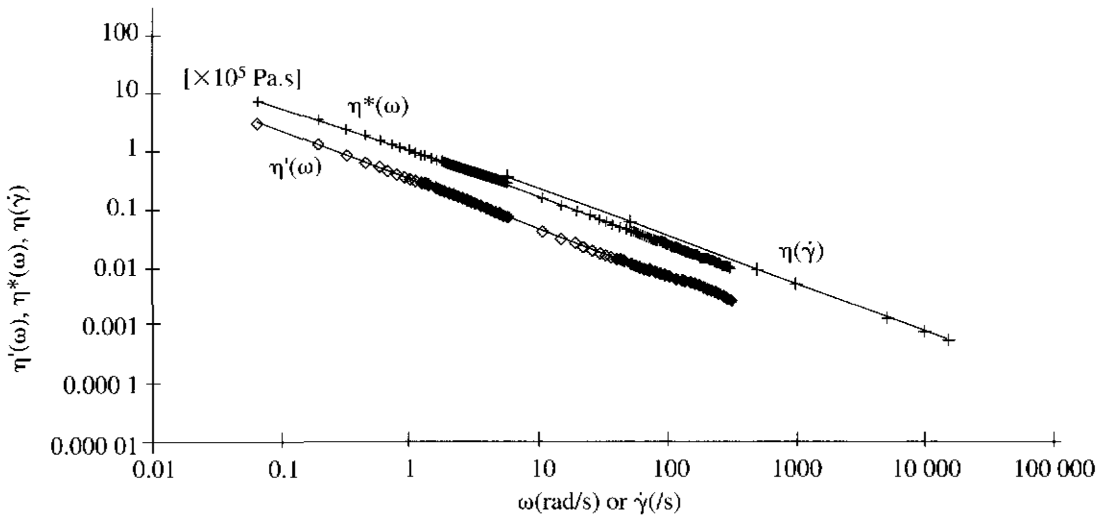
The study of dynamic properties of raw natural rubber over a frequency range from 0 1 Hz to 50 Hz can be concluded as follows

- Testing raw rubber using a hollow cylinder shear sample is better than using a pellet compression sample between cup-and-plate kit
- Based on the assumption of an equivalent of angular frequency and shear rate, the complex viscosity as a function of angular frequency calculated from the

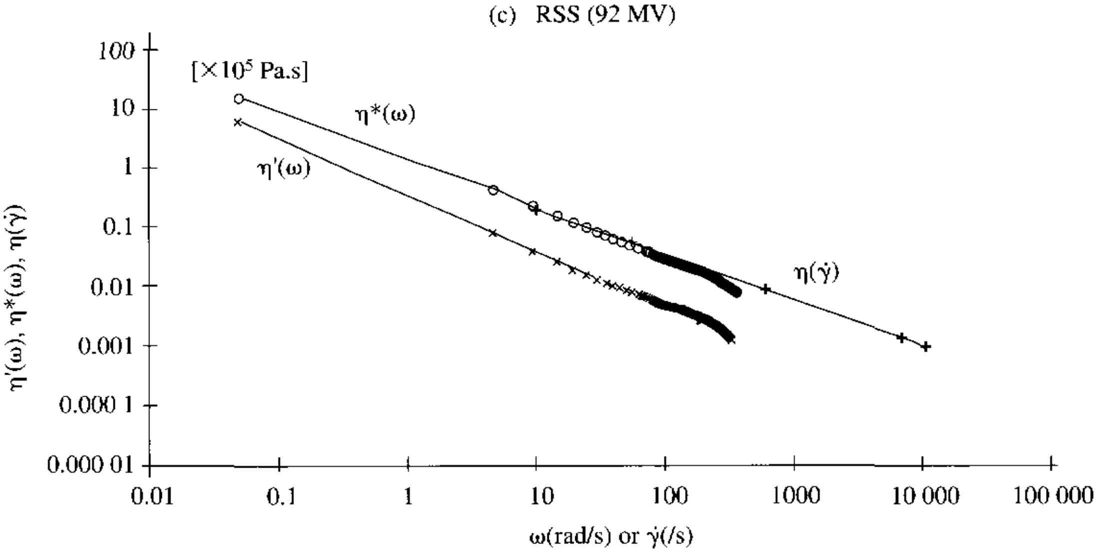
(a) STR 5 L (74 MV)

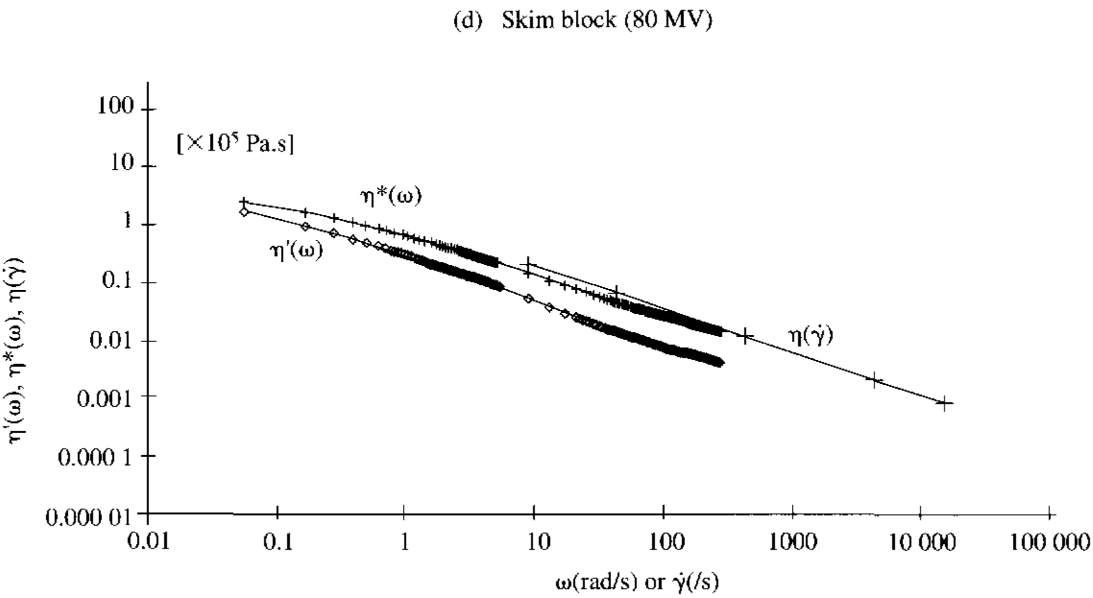


(b) STR 20 (85 MV)



Figures 4a and b. Comparison of shear viscosity, [$\eta(\dot{\gamma})$], real viscosity [$\eta'(\omega)$] and complex viscosity [$\eta^*(\omega)$] at 100°C.





Figures 4c and d. Comparison of shear viscosity, [$\eta(\dot{\gamma})$], real viscosity [$\eta'(\omega)$] and complex viscosity [$\eta^*(\omega)$] at 100°C.

DMA results is nearly equal to the viscosity as a function of shear rate, for all unfilled raw natural rubber tested here.

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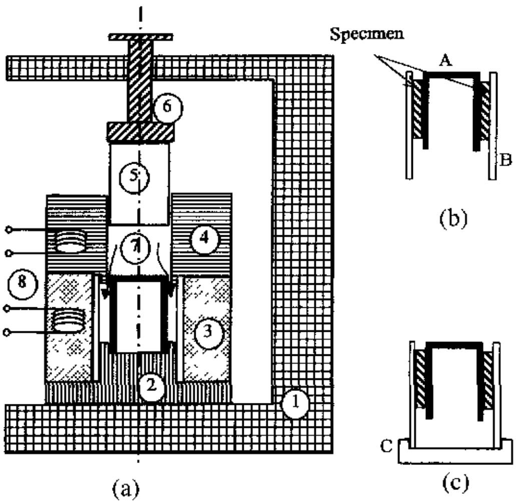
APPENDIX A



A = Inner metal holder cylinder
B = Outer metal holder cylinder
C = Supported metal plate
1 = Metal clamp, holding the whole set
2 = Lower part of the mould, holding a molten rubber
3 = Surrounded hollow cylinder, where the heat is supplied
4 = Heated barrel used to collect rubber melts
5 = Piston
6 = Screw to drive the piston
7 = Rubber melt
8 = Electric heater wires

Figure A-1. Diagram of sample preparation for moulding: (a) Arrangement of the whole set of devices: (b) A ready moulded specimen and (c) Sample configuration held in the DMA 7.

The following are the steps involved in the sample preparation

1 Cut small pieces of masticated raw rubber and put into the barrel (4), where the electric heater coil (8) is wound in order to heat the rubber

2 Carefully move the piston (5) down by turning a screw (6) Rubber melt is then transferred into the gap between cylinder A and B

3 Turn the electric heater (8) off and remove the upper and lower parts of the mould The final set is showed in (b)

4 Place the sample obtained from *Step 3* on to a supported metal plate (C) and load this sample into the DMA 7 holder, where the vibration probe of DMA 7 will be forced on the metal (A)