

## ***Effects of Drying Conditions on the Technological Properties of Sheet Natural Rubber***

M.M. JAYASURIYA<sup>\*#</sup>, K.M.U. MITHRANANDA<sup>\*</sup>, S. YAPA<sup>\*</sup>, U. RATNAYAKE<sup>\*</sup>,  
S. SIRIWARDENA<sup>\*</sup>, G. KUMARA<sup>\*</sup>, R. LIYANAGE<sup>\*</sup>, C. KURUPU<sup>\*</sup>  
AND M. WEJESEKARA<sup>\*</sup>

*Attempts were made to prepare natural rubber sheets by varying the drying time and drying conditions. Rheological and technological properties of rubber compounds based on ACS-1 and tread formula were evaluated. It was found that sun drying of rubber sheets for up to 2 days, followed by smoking for up to 2 days could impart equivalent physical properties with that of purely smoked rubber. In addition, it was found that drying of rubber sheets under sunlight for 2–4 days did not impart inferior physical properties.*

Recently various research works have been carried out on the use of sunlight for drying of raw rubber. Nevertheless, in the past, sun drying of raw rubber sheets was not recommended since the UV content of sunlight can generate free radicals in the rubber which initiate photodegradation<sup>1</sup>. This process leads to deteriorated physical properties. Moreover, it has been reported that photodegradation can be autocatalytic after a certain stage of exposure is reached<sup>2</sup>.

Previous research conducted on the use of sunlight for drying of rubber sheets indicated that drying could be done under sunlight for short periods, followed by smoking for up to 2–3 days. Furthermore, it is reported that deterioration of physical properties was not prominent in rubber sheets which were dried for short periods<sup>3</sup>. Hence, in this study further attempts were made to evaluate the effects of drying conditions on rheological characteristics

of rubber compounds based on ACS-1 formula. In addition, certain technological properties of a tyre tread formula based on variably dried rubber sheets were determined.

### EXPERIMENTAL

Field latex was diluted with water to 15% dry rubber content (DRC) and the diluted latex was coagulated by using 1% formic acid. The coagulum, thus obtained, was passed through mills with both smooth and diamond rolls and then subjected to various drying conditions. In this study, rubber sheets were dried using sunlight and/or smoke. In the case of two days' smoked dried rubber, it was subjected to further drying at room temperature for up to four days' since two days' of drying by the smoking process was not sufficient. Therefore, the two day sun-dried sheets were also subjected to the same treatment.

<sup>\*</sup> Rubber Research Institute of Sri Lanka, Telawala Road, Ratmalana, Sri Lanka

<sup>#</sup> Corresponding author

The first part of this study was carried out by using the ACS-1 formula to evaluate the effect of smoking on cure characteristics of gum compounds. The latter part of this study was conducted using tyre tread compounds and the formulations used are given in *Table 1*. Raw rubber properties of sheet rubber were determined according to *ISO* methods.

Rheological properties of rubber compounds were determined using a Monsanto rheometer. Physical properties such as hardness, resilience, compression set, abrasion resistance and tensile properties were determined according to *ISO* standards. Furthermore, retention of tensile strength was determined by heating the tensile pieces at 100°C for 3 days and it was calculated as follows:

$$\text{Retention tensile strength, (\%)} = \frac{\text{Tensile strength, obtained after ageing}}{\text{Tensile strength, obtained before ageing}} \times 100$$

## RESULTS AND DISCUSSIONS

The physical properties of the raw rubber are given in *Table 2*. The results clearly indicated that the plasticity retention index (PRI) of rubber sheets that were subjected to sun drying was greater than the recommended minimum PRI value (60) for good grades of ribbed smoked sheets (RSS). As expected, the PRI values of smoked sheet rubber were comparatively higher than those of sun-dried samples. Furthermore, it was observed that the PRI values of rubber samples dried by using sunlight and smoking were comparatively higher than those of either purely sun-dried rubber or smoked rubber. The same trend was observed in previous research work<sup>3</sup>. When compared to sun-dried samples, this behaviour was probably due to the deposition of phenolic cresotic substances during the smoking process and it may exert an antioxidant effect on rubber. However, the exact reason for having high PRI values in the above-mentioned samples compared to the purely smoked samples is not clear. Most of the PRI values seem to be independent from  $P_0$  values.

TABLE 1. FORMULATION FOR ACS-1 AND TYRE TREAD

| Item/Material                                    | ACS-1 (g) | Tyre tread (g) |
|--------------------------------------------------|-----------|----------------|
| Natural rubber                                   | 100       | 100            |
| Carbon black                                     | —         | 50             |
| Processing oil                                   | —         | 5              |
| Zinc Oxide                                       | 6         | 4              |
| Stearic acid                                     | 0.5       | 2              |
| IPPD (N-Isopropyl-N'-phenyl-ppd)                 | —         | 1.5            |
| Sulphur                                          | 3.5       | 1.5            |
| MBTS (Dibenzothiazyl disulphide)                 | 0.5       | —              |
| CBS (N-Cyclohexyl-1-2-benzothiazolesulphenamide) | —         | 1.8            |

TABLE 2. PHYSICAL PROPERTIES OF RAW RUBBER

| Sample                                                                        | Dirt (%) | Ash (%) | Volatile matter (%) | N <sub>2</sub> (%) | Initial plasticity ( <i>P<sub>0</sub></i> ) | Plasticity retention Index (%) |
|-------------------------------------------------------------------------------|----------|---------|---------------------|--------------------|---------------------------------------------|--------------------------------|
| 2 days' sun dried, then 2 days' smoked (Sd <sub>2</sub> and Sm <sub>2</sub> ) | 0.93     | 0.27    | 1.23                | 0.11               | 43                                          | 88                             |
| 3 days' sun dried, then 1 day smoked (Sd <sub>3</sub> and Sm <sub>1</sub> )   | 0.079    | 0.27    | 0.50                | 0.10               | 47                                          | 89                             |
| 2 days' sun dried (Sd <sub>2</sub> )                                          | 0.064    | 0.29    | 0.75                | 0.10               | 47                                          | 66                             |
| 4 days' sun dried (Sd <sub>4</sub> )                                          | 0.054    | 0.21    | 0.61                | 0.11               | 53                                          | 68                             |
| 6 days' sun dried (Sd <sub>6</sub> )                                          | 0.052    | 0.28    | 0.54                | 0.11               | 46                                          | 61                             |
| 2 days' smoked (sm <sub>2</sub> )                                             | 0.042    | 0.23    | 0.41                | 0.15               | 51                                          | 78                             |
| 4 days' smoked (Sm <sub>4</sub> )                                             | 0.056    | 0.26    | 0.26                | 0.10               | 40                                          | 85                             |
| 6 days' smoked (Sm <sub>6</sub> )                                             | 0.059    | 0.28    | 0.16                | 0.10               | 39                                          | 85                             |

However, as the drying time increases,  $P_0$  tends to decrease in sun-dried rubber sheets or smoked sheets, except in the Sd<sub>2</sub> sample. Similar observations were made in a previous study<sup>4</sup>.

### Cure Characteristics of ACS-1 Formulation

Cure characteristics of gum compou vulcanisation temperatures. The results are given in Table 3. On account of the ease of mixing the rubber with reproducible results, and due to the sensitivity of vulcanisation of the non-rubber components, ACS-1 gum formulations were used in this study to determine the effect of the intensity of smoking on cure characteristics. Cure rate is one of

the important factors which determines the technological properties of rubber. It mainly depends on the polyisoprenoid character<sup>5</sup>. It can also be influenced by the non-rubber materials present<sup>5</sup>. In the past, research work had been carried out to determine the state of cure/rate of pure gum NR vulcanised compounds by using modulus - time curves of a simple mathematical form equivalent to that of a first order chemical reaction<sup>6</sup>. For those cases, the rate of cure can be measured by the value of the first-order velocity constant, which is a measure of the reciprocal of the vulcanisation period required to produce a given percentage cure<sup>6</sup>. In this study, cure rates of NR vulcanisates were calculated from

TABLE 3. EFFECT OF SMOKING INTENSITY ON CURE CHARACTERISTICS OF GUM COMPOUNDS

| Cure Characteristics | Curing temperature |                 |                                     |                 |                 |                 |                                     |                 |                 |                 |                                     |                 |
|----------------------|--------------------|-----------------|-------------------------------------|-----------------|-----------------|-----------------|-------------------------------------|-----------------|-----------------|-----------------|-------------------------------------|-----------------|
|                      | 140°C              |                 |                                     |                 | 150°C           |                 |                                     |                 | 180°C           |                 |                                     |                 |
|                      | Sm <sub>4</sub>    | Sm <sub>6</sub> | Sd <sub>2</sub> and Sm <sub>2</sub> | Sd <sub>4</sub> | Sm <sub>4</sub> | Sm <sub>6</sub> | Sd <sub>2</sub> and Sm <sub>2</sub> | Sd <sub>4</sub> | Sm <sub>4</sub> | Sm <sub>6</sub> | Sd <sub>2</sub> and Sm <sub>2</sub> | Sd <sub>4</sub> |
| Scorch time (min)    | 3.51               | 2.83            | 2.92                                | 4.09            | 2.30            | 2.73            | 1.85                                | 3.4             | 1.04            | 1.32            | 1.19                                | 1.12            |
| 90% Cure time (min)  | 19.1               | 15.6            | 17.5                                | 23.3            | 13.8            | 13.8            | 11.8                                | 15.7            | 3.40            | 3.2             | 3.40                                | 3.46            |
| 95% Cure time (min)  | 24.8               | 19.1            | 21.1                                | 29.0            | 16.8            | 16.8            | 14.6                                | 19.2            | 3.80            | 3.83            | 3.80                                | 3.9             |
| Cure rate index      | 6.41               | 7.83            | 6.86                                | 5.21            | 8.69            | 9.0             | 10.05                               | 8.13            | 42.37           | 53.19           | 45.25                               | 42.74           |

rheographs by using the percentage of reciprocal value of the difference between 90% cure time and scorch time (s.t). [Cure rate index =  $100/(t_{90\%} - s.t)$ ]

Results indicated marked differences in cure time among the compounds of smoked RSS sample itself and also between the vulcanisates of smoked and sun-dried sheet rubber cured at 140°C and 150°C. It was found that compared to sun-dried samples, the cure rate increased significantly with smoking time at low vulcanising temperatures such as 140°C – 150°C. Nevertheless, when vulcanisation was carried out at 180°C, it is difficult to observe a marked difference in cure rates between sun-dried and 4 days' smoked RSS samples. It is known that fatty acids<sup>7</sup>, which are actually not very soluble in rubber, solubilise the accelerator during vulcanisation process. Hence, it may be reasonable to assume that the presence of volatile aldehydes which convert to carboxylic acid may act as a carrier for zinc and it helps to solubilise the accelerator. It may cause an activating effect on the accelerator. Therefore, such effect could impart higher cure rate in smoked rubber sheets.

From these results, it is very clear that as the curing temperature is increased the cure rate also increases significantly. Hence, the effect of cresotic substances on cure rate between 4 days' smoked RSS and sun-dried samples were not marked at high temperatures.

### Effect of Drying Conditions on Vulcanisation Characteristics

Vulcanisation characteristics of rubber compounds that were based on tyre tread formula are given in Table 4. Comparisons of torque values indicate that smoked dried rubber have higher  $M_H$  values (maximum torque) except for sample  $Sm_6$ . Concerning the rate of vulcanisation, no marked differences in cure rate were observed between smoked and sun-dried samples. Therefore, the effect of smoking intensity on cure rate is quite insignificant in smoked rubber sheets, which were compounded according to the tyre tread formula.

Physical properties of rubber compounds, prepared by using rubber samples that were

TABLE 4. CURE CHARACTERISTICS OF TYRE TREAD FORMULATIONS

| Sample            | $M_L$ | $M_H$ | $t_{10}$<br>(mins) | $t_{50}$<br>(mins) | $t_{90}$<br>(mins) | Cure rate<br>index (%) |
|-------------------|-------|-------|--------------------|--------------------|--------------------|------------------------|
| $Sd_2$            | 1.5   | 10.6  | 3.57               | 6.31               | 11.53              | 13.33                  |
| $Sm_2$            | 1.5   | 14.3  | 4.18               | 6.26               | 11.27              | 13.90                  |
| $Sd_2$ and $Sm_2$ | 1.9   | 15.1  | 3.31               | 5.68               | 11.01              | 12.70                  |
| $Sd_3$ and $Sm_1$ | 1.9   | 14.7  | 3.37               | 5.83               | 11.01              | 12.82                  |
| $Sd_4$            | 1.6   | 9.3   | 3.53               | 6.56               | 12.56              | 11.90                  |
| $Sm_4$            | 1.4   | 11.8  | 4.26               | 7.07               | 13.03              | 11.38                  |
| $Sd_6$            | 2.0   | 14.4  | 3.51               | 6.56               | 12.39              | 11.14                  |
| $Sm_6$            | 1.7   | 12.7  | 4.06               | 6.46               | 13.14              | 10.95                  |

dried under different conditions, are given in Table 5. Noticeable changes in hardness values between sun-dried and smoked samples were not seen. Similar observations were made for the resilience values of rubber samples.

Laboratory abrasion resistance results are shown in Table 5. Results clearly indicated that there was not any significant variation in abrasion values between sun-dried and smoked samples. The same trend could be observed for the comparison data values.

Tensile properties (Figure 1) indicated that as the drying time increases, tensile strength tends to decrease in sun-dried samples. The same trend could be observed for retention tensile strength (Figure 1). It indicates that there is a tendency to decrease ageing resistance as the time of exposure to sunlight is increased

It could be observed that as the smoking time increases the retention of tensile strength tends to increase up to an optimum level, and thereafter, shows a downward trend. Low levels of ageing resistance shown by the two-

day smoked sample and the six-day smoked sample, may be due to insufficient drying conditions and may be due to the degradation caused by over-drying, respectively. The results indicated that with respect to sun-dried samples, the tensile strengths of smoked samples are comparatively high. Further, it was noted that, the tensile strengths of rubber samples, Sd<sub>2</sub> and Sm<sub>2</sub> and Sd<sub>3</sub> and Sm<sub>1</sub> were 25 MPa. However, marked differences in retention tensile strength could be observed between Sd<sub>2</sub> and Sm<sub>2</sub> sample and Sd<sub>3</sub> and Sm<sub>1</sub> sample as 95.9% and 85.3%, respectively. Moreover, it was evident that smoked samples have higher ageing resistance than that of purely sun-dried samples and it may be due to the phenolic cresotic substances which are deposited during smoking.

## CONCLUSIONS

Smoking of RSS up to 4–6 days imparts higher cure rate in gum compounds of natural rubber compared to compounds based on sun-dried rubber, especially at low curing temperatures.

TABLE 5. PHYSICAL PROPERTIES OF RUBBER COMPOUNDS BASED ON SUN DRIED AND / SMOKED RUBBER

| Sample                              | Hardness (IRHD) | Resilience (%) | Compression set (%) | Din abrasion (g/cm <sup>3</sup> ) |
|-------------------------------------|-----------------|----------------|---------------------|-----------------------------------|
| Sd <sub>2</sub> and Sm <sub>2</sub> | 66              | 52             | 14.0                | 0.22                              |
| Sd <sub>3</sub> and Sm <sub>1</sub> | 63              | 54             | 14.6                | 0.23                              |
| Sd <sub>2</sub>                     | 57              | 54             | 16.3                | 0.33                              |
| Sd <sub>4</sub>                     | 59              | 54             | 12.7                | 0.22                              |
| Sd <sub>6</sub>                     | 61              | 58             | 15.2                | 0.31                              |
| Sm <sub>2</sub>                     | 62              | 52             | 19.0                | 0.27                              |
| Sd <sub>4</sub>                     | 59              | 53             | 16.5                | 0.21                              |
| Sm <sub>6</sub>                     | 60              | 54             | 13.2                | 0.31                              |

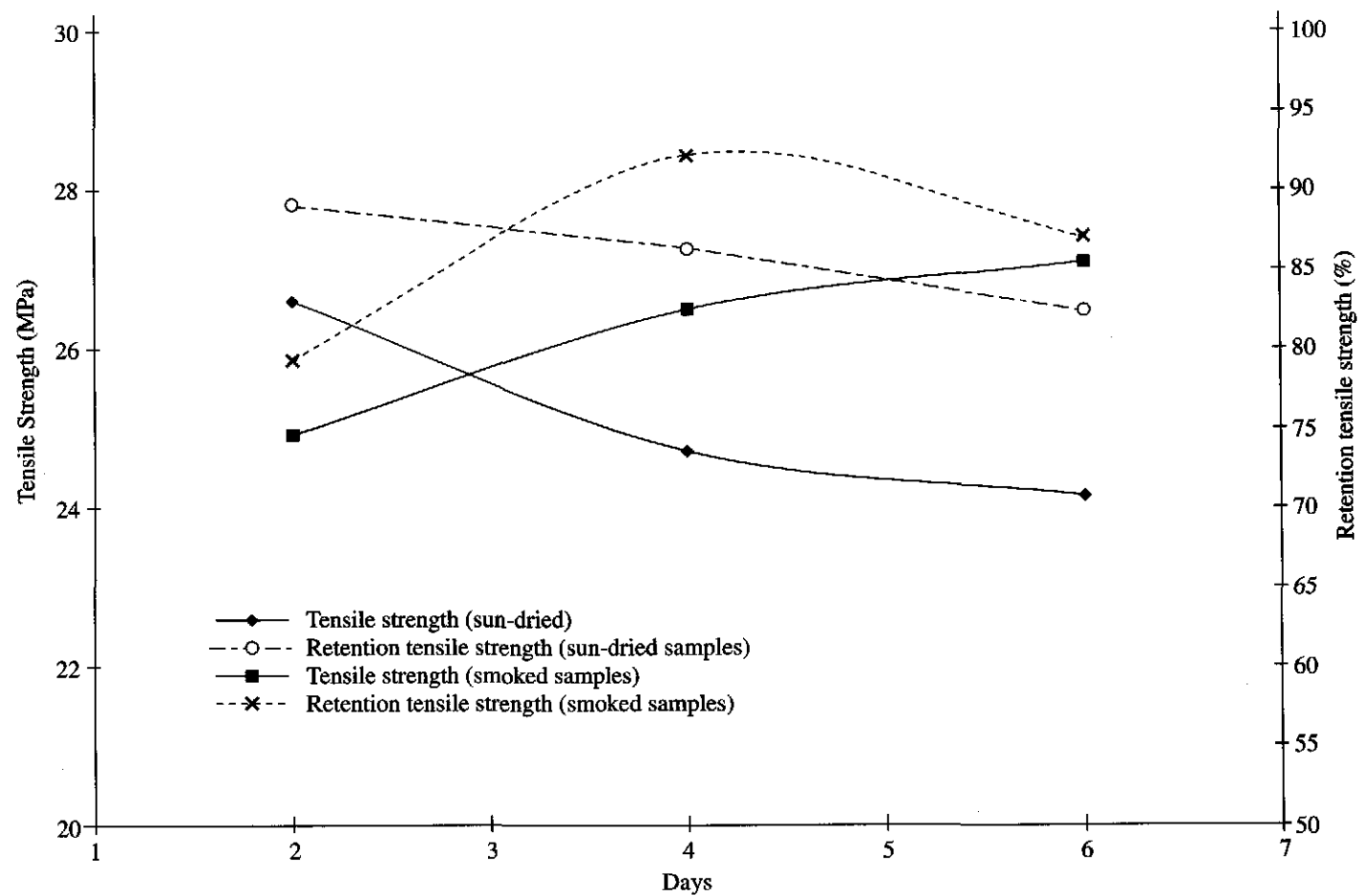


Figure 1. Effects of drying methods on tensile and retention tensile properties.

Further, it was found that, compared to smoked samples, even though the PRI of sun-dried rubber were comparatively lower, they were higher than the recommended PRI value (60)

When physical properties such as hardness, resilience and abrasion resistance were considered, sun drying under moderate sunlight did not cause adverse effect on physical properties. However, physical properties of purely sun-dried rubber samples could be further improved by the smoking process of up to 2 days. In addition, rubber sheets that were dried under sunlight up to two days followed by drying in smoke house up to two days exhibited much better physical properties and ageing resistance that are approximately equivalent to smoked samples which were dried up to 4–6 days. However, it was found that when compared to purely sun-dried samples, ageing resistance of smoked samples were comparatively higher.

#### ACKNOWLEDGEMENTS

The authors thank Dr L M K Tillekeratne, Director, RRISL, for his valuable guidance given throughout this project and wish to extend sincere thanks to Dr Karnika De Silva and Dr Kalyani Liyanage, Mr K E Jayasuriya and Dr Rizmi for reading the manuscript.

*Date of receipt* October 1999

*Date of acceptance* July 2000

#### REFERENCES

- 1 TILLEKERATNE, L M K, SCOTT, G AND AMIN, A (1975) *Eur Poly J*, **2**, 85
- 2 BOON, A (1988) Hock Cleavage – The Cause of Main-chain Scission in Natural Rubber Autoxidation *J nat Rubb Res*, **3**(2), 90–91
- 3 TILLEKERATNE, L M K, NUGAWELA, A, JAYASURIYA, M, WEERAMAN, S AND SIRIWARDENA, T A S (1966) Utilisation of Sunlight for Drying of Rubber *J nat Rubb Res*, **10**(2), 77–81
- 4 ALEX, R, KURIAKOSE, B AND KURUPPU, W C M (1996) *Symposium on the Technology and Uses of Natural Rubber, Beruwala Sri Lanka* Rubber Research Institute of Sri Lanka
- 5 BRISTOW, G M (1974) Comparison of the ACS-1 and ISO Black-filled Test Mixes *NR Technology*, **5**, 75–81
- 6 GEE, G AND MORREL S H (1952) *Trans Rub Ind* **28**(2), 102–104
- 7 HOFFMAN, W (1967) *Vulcanization and Vulcanizing Agents* London MacLarene and Sons Ltd, 151–153