Mill Behaviour of Rubber on Two Roll Mill with Temperature

Hiroyuki Ontsuka[1] and Michiharu Toh[2]

[1]*Chemicals Evaluation and Research Institute (Inc.) Nagoya Establishment, 1-4-chome, Orito-cho, Showa-ku, Nagoya, Aichi 466-0858, Japan*
[2]*R&D Department Kurume Research Park Co., Ltd., 1-1 Hyakunen Kouen, Kurume, Fukuoka 839-0864, Japan*

Translated by K. Halpin

INTRODUCTION

Open rolls, invented by Chaffee in 1833 and since improved, remain in use today. However, few reports are to be found on the thermal behaviour of rubber on open rolls, which has been looked at purely in the context of rubber milling and mixing operations. This review outlines what has hitherto been reported on rubber behaviour in milling on open rolls in relation to temperature.

HEAT TRANSFER CHANNELS IN OPEN ROLLS

Open rolls are of two kinds. The bored roll has a hollow interior as in Figure 1 and transfers heat via a thick wall. The drilled roll has heat transfer channels extending close to the roll surface as in **Figure 1** and presents a relatively uniform surface temperature [1]. A century ago, a year before Banbury's invention of the Banbury internal mixer in 1916, Pearson noted in the book "Rubber Machinery"

Drilled roll

Bored roll

Figure 1. *Drilled roll and bored roll*

[2] that open rolls, which were then the dominant form of rubber milling machinery, increased their output two-fold when the milling length of the rolls and speed of rotation were doubled, and gave an account of open rolls using various modes of heat transfer in which steam or coolant was passed either to raise the temperature or prevent rubber scorch due to self-heating during milling.

Drilled rolls reviewed are the Cowen-Bragg cooling roll, which has eight linear channels in the axial direction (i.e. lengthwise) close to the roll surface; the Bragg cooling roll, which is provided with a helical channel close to the roll surface; and the Bragg built-up roll, an improved form of these rolls. Bored rolls reviewed are the Brewster cooling roll, whose internal wall is provided with ribs between which eight perforated heat-transfer tubes are mounted; an improved form of this; and the Norris cooling roll, provided with channels formed by subdividing the hollow cylinder with six lengthwise partitions.
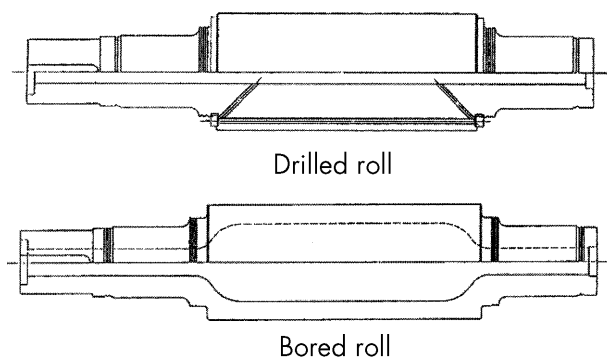
CHANGES IN RUBBER TEMPERATURE ON ROLLS

Heat transfer between rubber and rolls, etc.

In 1930 the journal *Gomu* carried a Japanese translation by Ishiguro of a report by Young [3] entitled "Rubber mixing machinery and mixing processes" [4]. The report notes that the mean temperature in a two-roll mill (since termed an open roll mill) is lower than in a closed mixer (since termed an internal mixer) but that there is a temperature distribution, the temperature

varying as shown in **Figure 2**. The gauge expressed as the nip (the minimum roll gap) N was 7/32 inch (5.5 mm) and the calender sheet had a band thickness of 12/32 inch (9.5 mm). The rubber temperature on the front roll was 105°C at the nip point N and 86°C at the exit 1 from the nip, thereafter reaching 82°C at the roll bottom point 2, 79°C at the level point 3, and 76°C at the top point 4. Where the front roll is exposed to the air, namely between the 90° (2) and 270° (4) positions, the rubber temperature falls 3°C every 90° owing to heat radiation; in contrast the temperature rises 30°C, almost 10 times as much, from the roll top point 4 to N and falls by about 20°C from N to 1.

The temperature rise observed approaching the nip is attributable to evolution of heat due to mixing with compounding agents in the rotating bank, heat due to friction between the rubber band and mill bank as the nip is approached, and heat due to rapid stretching of the rubber.

The account notes that the critical temperature in the nip (the temperature limit for scorching) is held down by cooling on the back roll, and the rubber should thus remain below the temperature at which scorch occurs. In this case, the back roll that pulls the rubber in contact with the mill bank through the nip would be constantly cooled by heat convection and radiation to the air from the roll surface and by heat conduction from the roll surface to the internal wall surface, heat travelling from the internal wall surface to the cooling water by convection.

The account states that the band rubber is cooled on the front roll by the coolant inside the roll, and also by convection and radiation. The heat in the band in contact with the roll is transferred by conduction from the external roll surface to the internal wall surface, from which heat then travels by convective transfer in the coolant. Cooling from the rotating rubber band surface,

in contact with air, is attributed to radiative transfer via the air and convective exchange with the air.

The original paper does not record the milling conditions in **Figure 2**, but appends the principal dimensions of five different open rolls. The rolls in **Figure 2** are very large, with a 36-40 inch diameter and 84 inch milling length; a back roll speed of around 15 rpm and a friction ratio of about 2 may be inferred. With the speed set at 16 rpm, for example, the peripheral speed of a 40 inch back roll works out to 51 m/min (0.85 m/s). Although questions surround rate of incorporation of the batch and compounding agents, amounts loaded, etc, it may be inferred from the scorch temperature that the rubber is natural rubber.

Critical temperature in the nip

The most obvious difference between open rolls and internal mixer for milling rubber is said to be that whereas in open rolls the rubber is passed through the nip subject to a critical temperature, this is not an issue in the case of an internal mixer. The following observations are also made. In open rolls, only the rubber layer in the nip comes into contact with compounding agents, whereas contact in an internal mixer is wholesale. The mean rubber temperature during mixing on open rolls is 90-95°C, whereas in an internal mixer the mean rubber temperature is generally 100-105°C, occasionally 110°C. It must not be forgotten that milling time is a major factor in scorch. In an internal mixer, the rubber is only exposed to temperatures from ambient to at most 105°C and milling is complete in 6-7 min. In the case of open rolls, the rubber temperature averages up to 95°C, but in milling that takes 35 min or more the rubber passes 500 times or more through the nip at up to the 120°C critical temperature. From the standpoint of scorch, therefore, the risk would seem to lie with open rolls.

Decrease in rubber temperature and back pressure after passage through the nip

After the rubber passing through the bank emerges from the nip, the rubber peels from the surface of the cooling back roll surface, heated through contact with the hot bank, and is exposed to the air. The pressure immediately the two rolls part beyond the nip would be expected to drop below atmospheric pressure, constituting a back pressure and resulting in cavitation. Any low boiling point matter incorporated would therefore volatilise into the atmosphere from the rubber band on the front roll, with a substantial decrease in the surface temperature of the band rubber by about 20°C in the interval from N in **Figure 2** to the exit zone 1. The reports reviewed below, though not relating to open rolls, substantiate the view that back pressure arises.
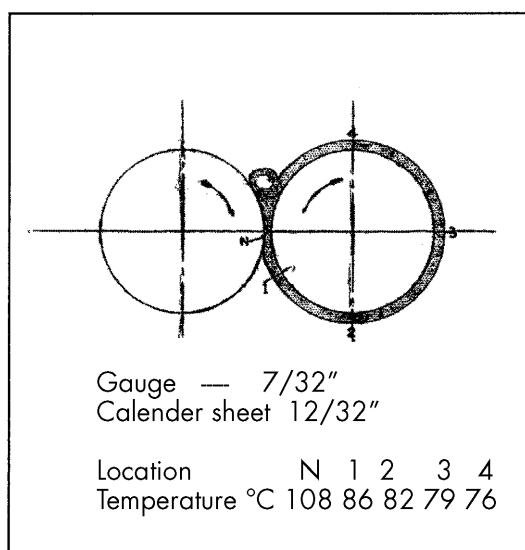

Gauge — 7/32"
Calender sheet 12/32"

Location	N	1	2	3	4
Temperature °C	108	86	82	79	76

Figure 2. Temperature distribution of rubber in a two-roll mill

Using oil as a Newtonian fluid, Banks et al. [5] and Myers et al. [6,7] made detailed observations of flow in the nip of a roll coater and divided flow into four regions: a bank region exhibiting secondary flow, a shear region at the nip, a cavitation region due to pressure drop beyond the nip, and a filamentation region where the two rolls part. Chung [8] measured the pressure change at the nip (gap 70-780 μm) in a stainless steel two-roll coating mill (diameter 16.4 cm, roll length 20 cm) in the mixing of polymerised linseed oil, a Newtonian fluid. The lower speed roll was immobilised, pressure sensors were attached from the interior close to the roll surface at the nip, and the pressure change in the nip was measured as the higher speed roll was rotated. The pressure was found to shift from positive to negative across the nip, consistent with theory.

Toh et al. [9] made a video recording of the motion of the rotor, rubber and voids in a two-dimensional model mixer and presented the results in **Figure 3a**. A void-free sickle-shaped zone filled with rubber is formed ahead of the rotor, while a void lies behind the rotor. Rubber in sheet form emerges into the void zone beyond the clearance between the rotor tip and inner wall of the vessel. **Figures 3b** and **c** are enlarged views of the clearance: the rubber on the rotor tip side is promptly peeled away on the trailing edge of the clearance, developing surface undulations, and this peeling invariably seems to occur ahead of peeling from the internal wall of the vessel, behaviour also observed in the actual video pictures.

Gondoh [10] attached a pressure sensor to the vessel wall of a miniature mixer and measured the internal wall pressure of the tank during mixing; a back pressure was reported to develop directly after the rotor tip had passed the sensor, resembling the change in pressure across the scraper blade in **Figure 3d** [11].

Figure 3e is an enlarged view of the nip section in **Figure 2**. It is supposed that in open rolls back pressure develops when the back roll separates from the band rubber on the front roll directly beyond the nip, and just like the above rubber peeled on the rotor tip side, the band rubber surface peeling from the back roll undergoes undulating shrinkage.

THE RUBBER MIXING PROCESS AND BANK FORMATION

In 1943 Mori [12] represented the state of mixing of rubber in the mixing operation (rubber milling) as shown in **Figure 4**, and observed that, generally speaking, care should be taken in two respects: (1) the various compounding materials must be blended (mixed) evenly across the roll in the shortest possible time; and (2) the rubber must be prevented from reaching a semi-vulcanised state, known as scorching. Moreover, unless

the roll temperature and gap and other operations are appropriately adjusted, the compounding chemicals will consolidate into small lumps instead of being completely distributed through the rubber. No explanation is offered for a, b, c, d, e and f in the diagram. However, Macbeth [13] presented the same drawing for the rubber mixing process in 1935, and although the rubber temperature is not given, a-f are explained as follows:

a) A zone in which the rubber mass (the bank) promptly begins to move, drawn towards the nip.

b) A zone into which the back roll draws the added compounding agents along with the rubber in zone a.
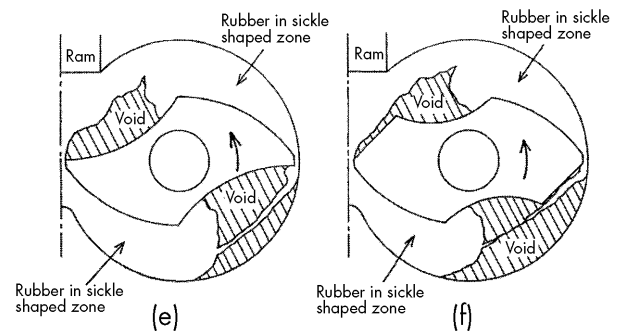

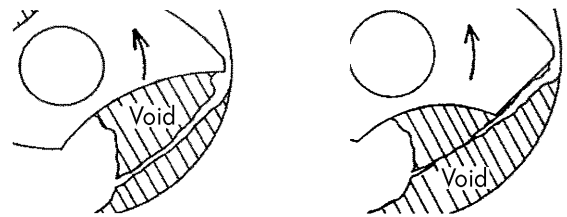*Figure 3a. Behaviour of rubber and void in rotors (e) and (f)*

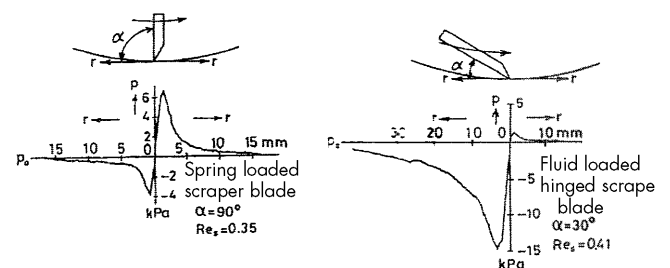*Figure 3bc. (b) Magnified view of clearance zone in rotor (e) (c) Magnified view of clearance zone in rotor (f)*

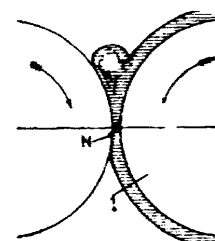*Figure 3d. Internal wall pressure distribution in front and behind scraper blade*

Figure 3e. Magnified view of nip environment

c) A zone in which the rubber reaches its greatest speed, a large temperature rise occurs, and heat passes to the roll.

d-e) A zone in which the temperature of the rubber gradually decreases.

f) A zone in which the rubber temperature starts to rise in contact with the bank of compounding agents.

Figure 4 seems to reference this drawing.

ROLL TEMPERATURE AND MILL BEHAVIOUR

In 1966, Tokita et al. [14, 15] and White [16] observed and filmed the milling of BR, SBR, etc, on 3 inch open rolls at different roll temperatures and discovered that mill behaviour can be divided into four regions, Region 1 to Region 4, according to temperature, as shown in **Figure 5**. They also made a theoretical analysis of the four regions. The roll speed was set at 16.2 rpm for the front roll and 22.7 rpm for the back roll, with a nip of 0.055 inch (1.4 mm). In Region 1 the rubber is at room temperature, elasticity is dominant, and the material does not penetrate the nip. In Region 2 the temperature is 66-71°C and rubber that has passed through the nip winds onto the front roll, forming a transparent viscoelastic band; as milling proceeds, a rotating bank forms above the nip. In Region 3, where the roll temperature has risen to 100°C, the rubber becomes turbid and hangs sack-like from the roll. In Region 4, the roll temperature has risen further to 121°C, viscosity is dominant and the rubber clings to the roll as a transparent liquid. The Mooney viscosity of the rubber (ML-4) in the experiments was 35-50.

Figure 6 corresponds to Region 2; it appears in texts published by the Society of Rubber Science and Technology Japan and others as a model of milling.

BANK TEMPERATURE AND RUBBER PROPERTIES

Bank temperature and Mooney viscosity in milling

In an evaluation of carbon black filler conducted by the Rubber Industry Technologists Association (Gomu Kougyou Gijutsuin-Kai) Committee 4 (Carbon Black), Aoe [18] reported the relation between the conditions in milling on open rolls and the physical properties of the rubber. The Committee member organisations used open rolls of different diameter, namely 3.5 inch, 6 inch, 7.5 inch, 8.9 inch, 10 inch, 12 inch and so on; dimensions such as roll length were also different. The Mooney viscosity in the milling of natural rubber (NR, RSS1) was therefore measured with the temperature of the bank rubber set constant.

Taking a 10 inch diameter, 20 inch long, double-geared variable speed open roll as an example, the front roll was set at 18 rpm and the temperature at 70±1°C, and milling was commenced when the back roll reached 70±5°C. The friction ratio was varied from 1:1 to 1:1.6, and a 900 g initial charge was milled for
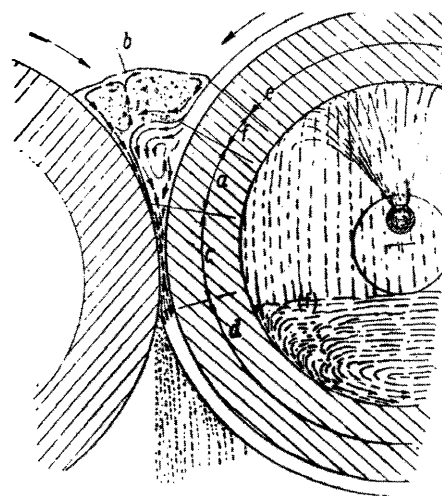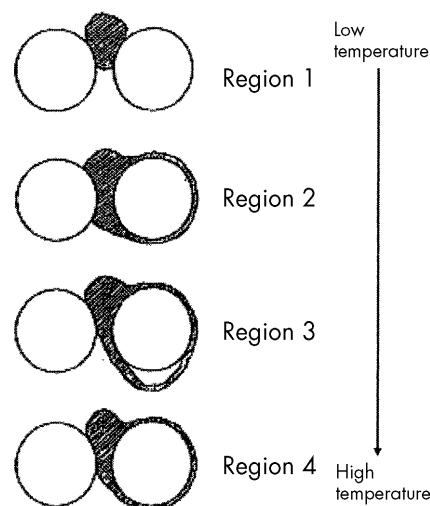

Figure 4. Mixing of rubber

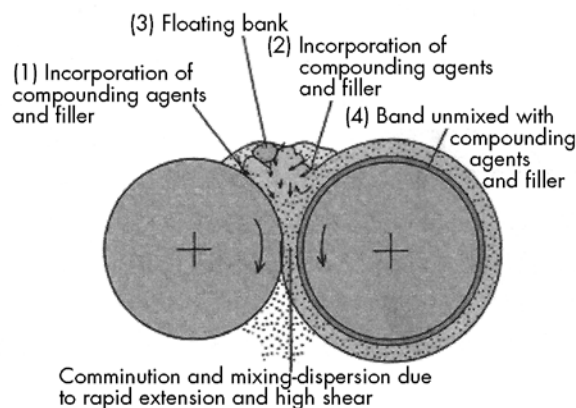*Figure 5.* Open roll temperature and milling behaviour

Figure 6. Model of mixing on open rolls

5, 15, 25 and 40 min at a roll guide spacing of 36 cm and gap at the nip of 1.4 mm. To measure the Mooney viscosity, 100 g was collected after each milling time, and to maintain the time taken to pass between the rolls the same as the initial time, the guide spacing was narrowed 4 cm at a time. The temperature of the bank was measured by thermistor.

The results in **Figure 7a** and **b** show that, with the rolls rotating at the same speed, the bank rubber temperature decreased with milling time from 92°C (5 min) to 82°C (40 min) while the Mooney viscosity decreased greatly from 63.5 (5 min) to 32 (40 min); at the high friction ratio of 1:1.6, however, while the bank temperature again decreased from 98°C (5 min) to 94°C (40 min) and the Mooney viscosity decreased from 66.0 (5 min) to 40.0 (40 min), the decreases were much smaller than at equal roll speed. Referring to **Figure 8**, in a similar experiment with the rolls cooled to hold the bank rubber temperature at 85±2°C, the Mooney viscosity ratio decreased from 59.0 (5 min) to 33.5 (40 min) at 1:1 and from 62.0 (5 min) to 32.0 (40 min) at 1:1.6, showing agreement regardless of friction ratio within a range of variation attributable primarily to experimental error. Given that bank rotation and the size of the rotating bank were essentially kept constant in the experiments, it is evident that the behaviour of the rotating bank is important.
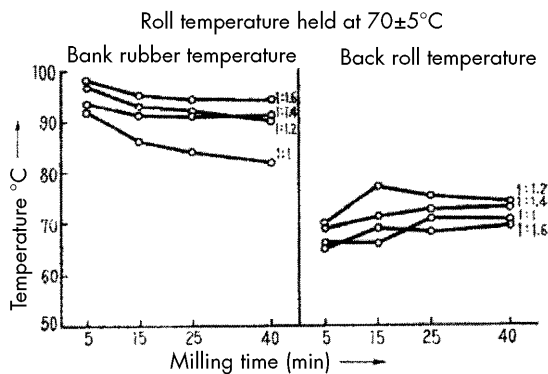
A further milling experiment was carried out with natural rubber using 8 inch and 10 inch rolls of different roll dimensions and speeds in which the bank temperature and number of passes (milling time) were controlled, affording the results in **Figure 9a** and **b**. Despite variation of the 10 inch and 8 inch roll sets and various other conditions, the curves for the two sets agree within the range of error in measurement at both 85°C and 65°C. It is concluded that, regardless of difference in roll diameter
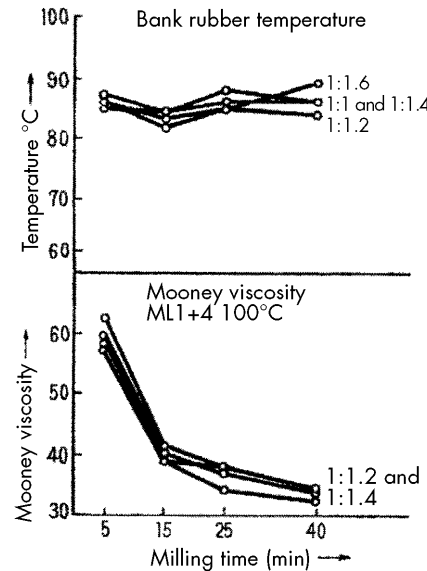
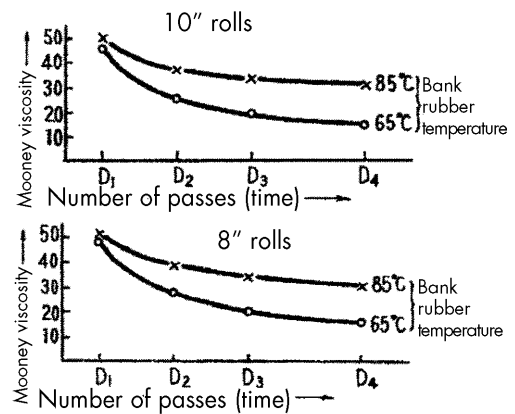**Figure 8.** Bank rubber temperature and Mooney viscosity at different milling times

Figure 7a. Rubber temperature in mill bank and back roll temperature
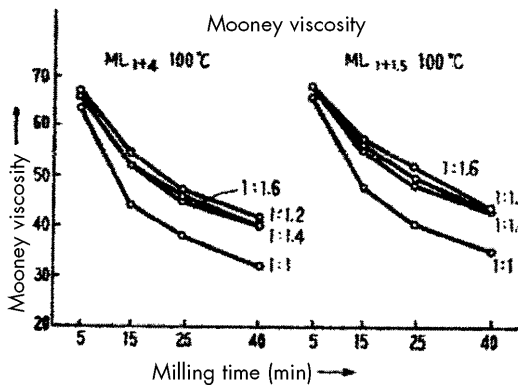
Figure 7b. Roll friction ratio and Mooney viscosity

Figure 9a. Change in Mooney viscosity with mill time on 10 inch and 8 inch rolls at fixed bank rubber temperature
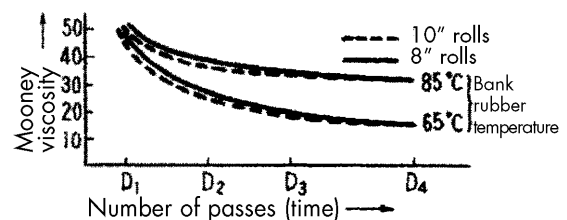
Figure 9b. Change in Mooney viscosity with mill time for two different roll sets at fixed bank rubber temperature

and conditions, the same milling is possible provided allowance is made for the other conditions, the main aim being to maintain a consistent bank temperature.

Bank temperature in roll mixing and vulcanisate properties

Eight members of the Carbon Black Committee conducted roll mixing experiments with carbon black using 6, 8 and 10 inch open rolls. Based on the ASTM mixing roll standard of roll diameter 6±0.1 inch (150-155 cm), roll guide spacing (band width) 10-11 inch (23-28 cm), front roll speed 24±0.5 rpm and friction ratio 1:1.4, the rubber was mixed at a bank temperature of 80±5°C with the number of passes through the nip kept the same. Mixing was carried out 3 times under the same mill conditions: the nip of 1.4 mm at commencement of carbon black addition was opened as appropriate during mixing, the mixing rate, etc, being set by adjustment of the nip to 1.9 mm by the time mixing was complete (20.5 min).

The compounding recipe was: constant viscosity NR 100.0 parts, carbon black (HAF) 50.0 parts, sulphur 2.5 parts, stearic acid 3.0 parts, DM accelerator 0.6 part. The compound was press-cured for 30 min at 145°C and the physical properties of the vulcanisate, namely tensile strength T_B [kg/cm^2], elongation E_B [%], 300% modulus M300 [kg/cm^2], and hardness H_S [JIS], were measured.

The bank temperatures recorded were 76-89°C (4 min) and 75-84°C (18 min), for which variations in vulcanisate properties of T_B 237-294 (mean 258), E_B 505-537 (mean 517), M300 119-139 (mean 133), and H_S 61-68 (mean 65) were reported.

It is clear from the reports that in mixing on open rolls the rubber properties are greatly influenced by the behaviour of the roll bank, specifically the bank temperature, and the roll nip as it relates to bank size.

CONCLUSIONS

On open rolls the rubber makes contact with compounding agents in just one place - at the bank over the nip, and the rubber is milled as it passes through the nip; the behaviour of the bank formed above the nip is hence paramount. Moreover, milling behaviour on open rolls is directly related to the temperature of the rubber, not the temperature of the rolls; the important factors are thus the temperature of the bank over the nip and the decrease in rubber temperature directly beyond the nip.

REFERENCES

1. Fujitani I., In "Gomu Kougyou Binran (4th ed.)". Nippon Gomu Kyoukai Ed. Nippon Gomu Kyoukai, Tokyo, p.l070 (1994).

2. Pearson H.C., In "Rubber Machinery", The India Rubber World, New York, p.59 (1915).

3. Young H.C., *India-Rubber Journal*, **78**, 621 (1929).

4. Young H.C., Isiguro K., Trans.: *Gomu*, **2**, 113 (1929-1931).

5. Banks W.H., Mill C.C., *Proc. Roy. Soc. (London)*, A223, 414 (1954).

6. Myers R.R., Miller J.C., T*rans. Soc. Rheol.*, **2**, 77 (1958).

7. Myers R.R., Hoffman R.D., *Trans. Soc. Rheol.*, **5**, 317 (1961).

8. Chung G.: " Roll Mill no Sousa Tokusei to Bunsan Tokusei ni Kansuru Kenkyu ", Doctoral Thesis. Kyushu University (1986).

9. Toh M., Gondoh T., Murakami Y., Nishimi H., Mishima M., In "Handbook of Applied Polymer Processing Technology", Marcel Dekker Inc., New York, p.523 (1996).

10. Gondoh T., 112th Gomuneri Kenkyu Bunkakai Siryou (2014).

11. Toh M., Murakami Y., *Kagaku Kogaku Runbunshu*, **6**, 416 (1980).

12. Mori T., Gomuseizouhou, Kagakukougyokai, p.7l (1943).

13. Macbeth C., "Kautschu kmaschinen", In "Handbuch der gesamten Kautschuktechnologie". (Union Deutsche Verlagsgesellschaft), Berlin. p.155 (1935).

14. Tokita N., White J.L., *J. Appl. Polym. Sci.*, **10**. 1011 (1966).

15. Tokita N., White J.L., *Rubber Chem. Technol.*, **40**, 1126 (1967).

16. White J. L., *Rubber Chem. Technol.*, **50**, 163 (1977).

17. Mizumoto K., In "Gomu Gijyutsu no Kiso (6th imp.)", Nippon Gomu Kyoukai Ed., Tokyo, p.l46 (1987).

18. Aoe I., *Nippon Gomu Kyoukaishi*, **41**, 314 (1968).