Effect of Temperature on the Sticking of Low Melting Point Materials[1]

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Sticking is a common phenomenon in the manufacture of tablets in which the top of a tablet becomes pilled during compression.

The effects of tableting machine temperatures on the sticking of butyl *p*-hydroxybenzoate were examined. Sticking was estimated by measuring the pressure placed on a scraper (scraper pressure, or SCR). The pressure reached a maximum value at a homologous temperature of around $T/T_m = 0.90$, where the probability of sticking is high. It was found that sticking occurs easily in materials having a low melting point and in fine particles when compression stress and temperature are constant.

We also found that the $\bar{x}$–R control charts obtained by measuring SCR help to detect sticking. Moreover, a shear test of tablets on a temperature-controlled metal plate was carried out, and the results were similar to those of the SCR; that is, the shear strength (C) and friction coefficient (μ) reached a maximum value at $T/T_m = 0.90$.

Keywords sticking; scraper pressure; control chart; shear strength; low melting point

Tableting troubles such as sticking and picking are phenomena that occur during the manufacture of tablets, in which particles adhere to the punch during compression. These problems can be major obstacles in the production of high-quality tablets. The followings are potential causes of sticking and picking.

The factors relating to the materials are melting point, size and distribution, surface condition and the hardness of particles. Factors that relate to machinery and the environment are the surface condition of the punch, compression force and speed, as well as the temperature and humidity around the machine.

Naito[2—4] has reported a means of measuring the slipping force of a tablet surface and the passive pressure of the lower punch during compression. Schmidt *et al.*[5] developed a simple method with which to measure the force necessary to remove the tablet from the punch surface, using a new transducer. Toyoshima *et al.*[6] studied the relationship between sticking and tablet surface roughness. Sugimori *et al.*[7] found that capping occurs when a tablet is cracked by high residual die wall pressure at the final stage of the decompression process. These studies however, failed to establish a method with which to assess sticking and picking.

Therefore, the measurement of scraper pressure, which is a kind of shear stress, would be the most direct means of evaluating sticking and picking.

The purpose of this study was to quantify sticking and picking on the basis of scraper pressure. We focused upon the effects of temperature on sticking during compression.

Experimental

Materials Butyl *p*-hydroxybenzoate (Kanto Kagaku Co., Ltd.) was passed through an air-jet sieve (Alpine Co., Ltd.). The samples are listed with their physical properties in Table I.

True density was measured with a helium-air pycnometer (Shimadzu-Micromeritics, type 1302). The melting point was taken as the intersection of the endothermic peak maximum gradient with the base line and was measured using a DSC apparatus (MAC Science DSC 3200). The size of at least 500 particles was determined using an image analyzer (Nireco, Luzex 500).

Control of Tableting Machine Temperature The temperature of the tableting machine was controlled by means of a thermocouple placed in close contact with the surface of each part of the machine (Kikusui Co., Ltd., type Correct 12 HUK-AW), and then connected to an air dryer.

Measurement of Scraper Pressure (SCR) Figure 1 shows the device used to measure SCR. The shear stress between the tablet and lower punch surfaces was determined using a strain gauge placed on the scraper, as shown in Fig. 1. Here, scraper 4 was set on arm 6 and the tablet collided vertically with the scraper. A strain gauge was placed on the scraper where the output value is maximum, and the values were calibrated with weights.

Detection of Sticking The surfaces of 100 tablets were visually observed and the sticking ratio was estimated. Tablets containing holes were regarded as "sticking," without consideration of the size and number of holes.

Preparation of Model Tablets Model tablets were compressed using a universal tension and compression tester (Shimadzu, type Autograph AG 5000D). Butyl *p*-hydroxybenzoate was compressed under 49 MPa pressure.

Measurement of Shear Strength The shear strength between the model tablet and metal plate surfaces was measured using the device shown in Fig. 2.

Shear strength was detected when it occurred between the model tablet (2.0 cm in diameter) surface and the metal plate, under constant normal

TABLE I. Physical Properties of Samples and Their Mixing

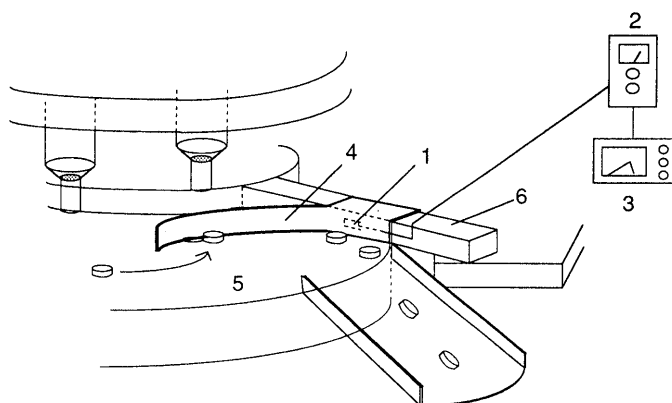| Sample	True density (g/cm³)	Melting point (°C)	Mean particle diameter (μm)
I	1.28	68	271
II	1.28	68	191
III	1.28	68	98
	Mixing ratio		
IV	I/III = 2/1		
V	I/III = 1/1		
VI	I/III = 1/2		

Fig. 1. Schematic Diagram of the Device for Measuring SCR

1, strain gauge; 2, amplifier; 3, *X*–*Y* recorder; 4, scraper; 5, turn table.

TABLE II. Surface Roughness of Metal Plates

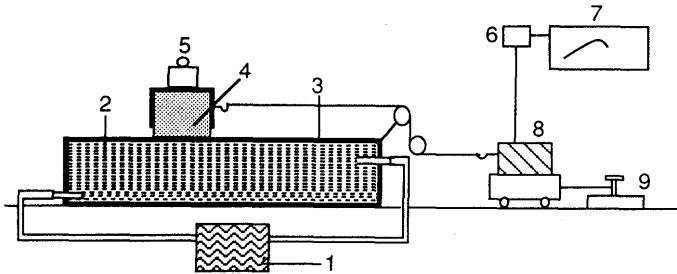|	Tutanaga steel plate	Stainless steel plate I	Stainless steel Plate II
Surface roughness R_{max} (μm)	64	7.2	3.8

Fig. 2. Schematic Diagram of Tester for Measuring Shear Strength

1, temperature controller; 2, constant temperature water bath; 3, metal plate; 4, model tablet; 5, weight; 6, amplifier; 7, X–Y recorder; 8, strain gauge; 9, motor.

stress. The model tablet to be tested was placed on a metal plate, which was controlled at a constant temperature and tensiled with the motor at a speed of 0.69 mm/s. The load was measured by a strain gauge fixed on the moving box, and led through a strain meter to a recorder. Each tablet was measured five times under specific conditions, and the values of the shear strength were averaged.

Measurement of Surface Roughness The surface roughness of the metal plates was measured by means of a SEM photograph (JEOL, type JSM-T-20). The sample metal plate was placed on the sample stage and photographed at right angles. The surface roughness was determined with SEM photographs and evaluated by an average of the maximum concavo-convex, R_{max}, as shown in Table II. A model tablet (2.0 cm in diameter and 0.5 cm in thickness) was made by using a universal tension and compression tester (Shimadzu, type Autograph AG-500D) for samples I and III. The model tablets were placed on the metal plate controlled at a constant temperature and slipped with shear stress.

Results and Discussion

Relationship between SCR and Sticking Table III shows that the tablet machine temperature was regulated. The scraper pressure was measured as the shear stress between the tablet surface and the lower-punch surface, thus it was necessary to control the lower punch temperature. Since the
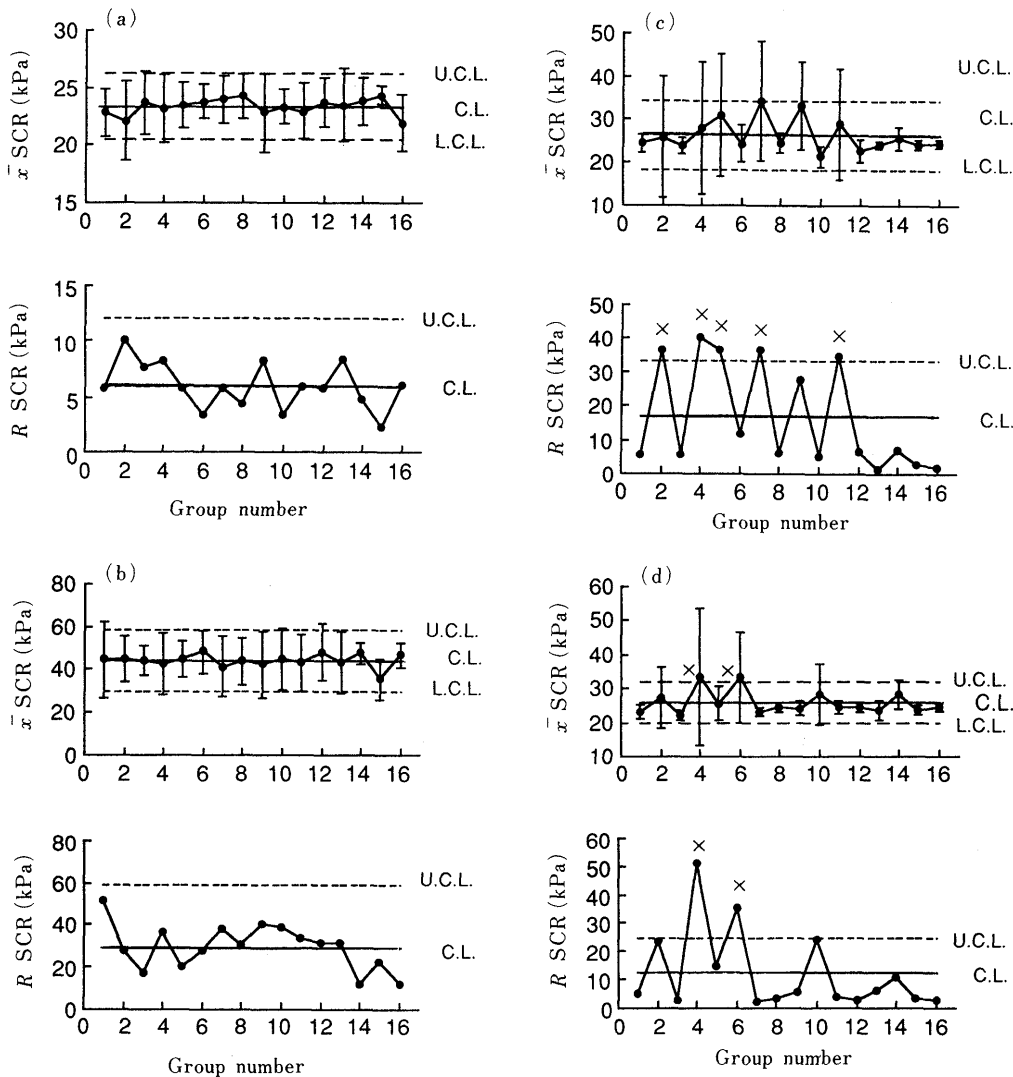

Fig. 3. x–R Control Charts for SCR (n = 6)

(a) Sample V: upper punch pressure = 108.6 MPa, lower punch temperature = 35 °C; (b) sample III: upper punch pressure = 16.9 MPa, lower punch temperature = 35 °C; (c) sample VI: upper punch pressure = 35.8 MPa, lower punch temperature = 35 °C; (d) sample VI; upper punch pressure = 23.1 MPa, lower punch temperature = 39.5 °C. × shows the occurrence of sticking.

temperature of the lower punch is difficult to determine during compression, we measured that of the upper punch instead, which adequately indicated the temperature of the lower punch, as shown in Table III.

The $\bar{x}$-R control chart was represented by using about 96 data after statistically evaluating the SCR, as shown in Fig. 3.

The control chart, which is useful for process control, is based upon a standard deviation or range.[8] Control charts using range, R, as a measure of spread, were drawn up with the following relationships:

upper limit of average, U.C.L. $= \bar{x} + A_2 \bar{R}$ (1)

lower limit of average, L.C.L. $= \bar{x} - A_2 \bar{R}$ (2)

TABLE III. Temperatures of Component Parts of Tableting Machine after Heating

Tablet machine parts	Temperature (°C)					
Upper punch	25.0	30.0	35.0	40.0	45.0	50.0
Lower punch	25.0	29.0	35.0	39.5	44.0	45.0
Turn table	24.5	28.0	35.0	41.0	45.0	48.0
Scraper	25.0	29.5	34.5	42.5	49.0	54.0
Upper-punch holder	23.5	29.0	37.0	44.0	50.5	55.0

where $\bar{x}$ is the average of total samples, $\bar{R}$ is the average of the range values, and A_2 is the factor for control limits, which consisted of six units at each sample interval. The value for A_2 has been calculated to be 0.483.[9]

Each point in Fig. 3a—d represents the average SCR of six tablets (x) taken at 1 to 16-cycle intervals. Thus, the SCR of 96 tablets were obtained. The average for the 96 (x) is represented by the solid horizontal line. The horizontal broken lines above and below the solid line represent three standard deviations from the mean. For the normal curve distribution of SCR, this means that 99.7% of the tablets in the batch will fall within the SCR range represented by the upper and lower limits. Figures 3a and b show the $\bar{x}$ and R control charts for examples of each point within the range of U.C.L. and L.C.L. However, in Figs. 3c and d, each point deviated from the upper and lower limit lines.

As shown in Figs. 3c and d, the abnormal SCR values suggest that the adhesive force between the tablet surface and the lower punch surface is very high. We then observed the tablet surface and estimated the sticking ratio.

In the $\bar{x}$-R control charts for sample V, each data point is within the range of U.C.L. and L.C.L., as shown in Fig. 3a. In these samples, sticking was not detected. The control charts of sample III showed almost the same pro-
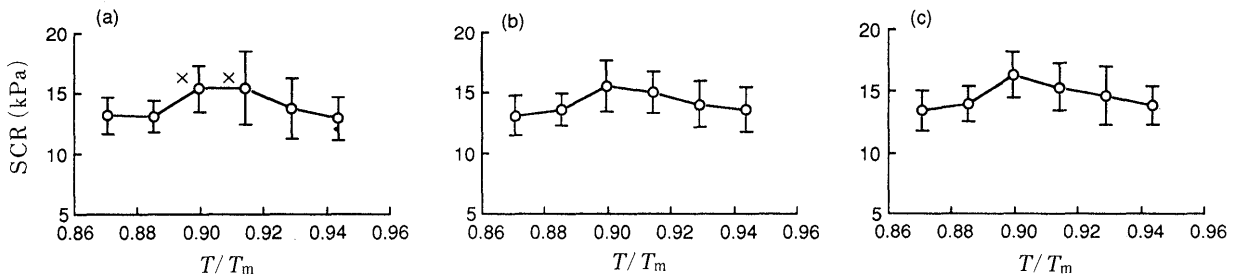

Fig. 4. Relationship between Homologous Temperature (T/T_m) and SCR for Sample I

(a) Upper punch pressure = 44.5 MPa, (b) upper punch pressure = 102.0 MPa, (c) upper punch pressure = 185.8 MPa. × shows the occurrence of sticking.
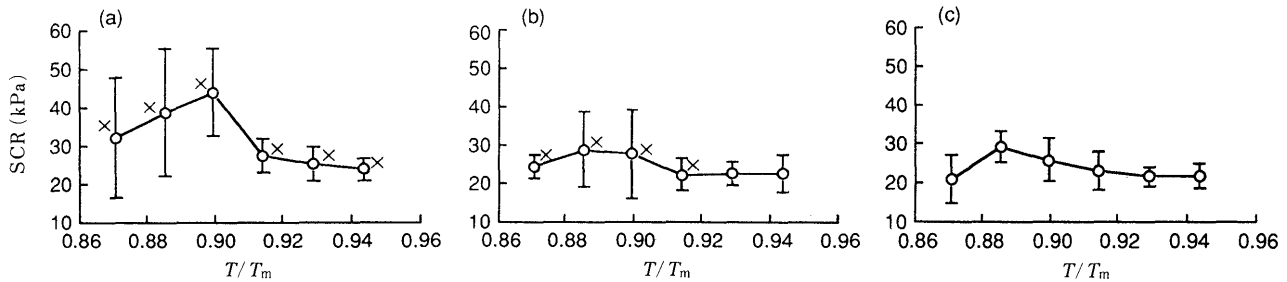

Fig. 5. Relationship between Homologous Temperature (T/T_m) and SCR in Sample III

(a) Upper punch pressure = 25.5 MPa, (b) upper punch pressure = 41.6 MPa, (c) upper punch pressure = 111.0 MPa. × shows the occurrence of sticking.
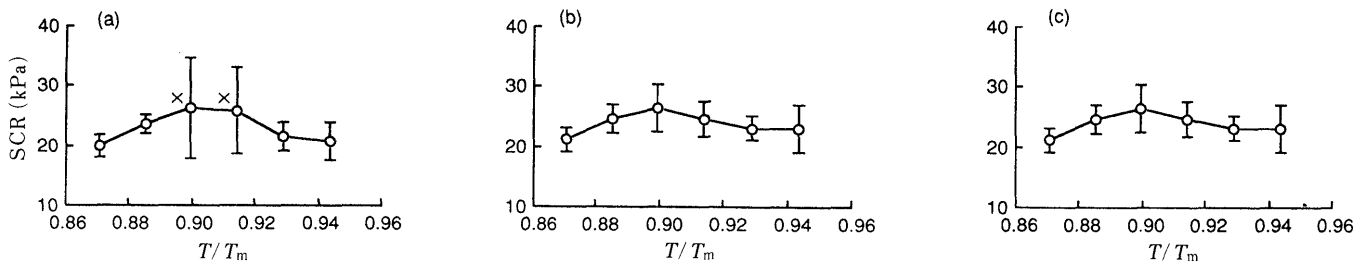

Fig. 6. Relationship between Homologous Temperature (T/T_m) and SCR in Sample VI

(a) Upper punch pressure = 38.5 MPa, (b) upper punch pressure = 90.8 MPa, (c) upper punch pressure = 158.4 MPa. × shows the occurrence of sticking.

files as sample V (see Fig. 3a), however, the sticking ratio was 69%. The sticking ratio evaluated from visual observation was 25% and 32%, respectively, for sample VI at 35 and at 39.5 °C as shown in Figs. 3c and d. These results suggested that sticking occurred at either abnormally high SCR values or beyond the range of U.C.L. and L.C.L., as shown in Figs. 3b, c and d.

Effect of the Lower Punch Temperature on SCR The SCR of each sample was measured and the effect of the lower punch temperature was examined. Figures 4—6 show the results obtained using samples I, III and VI. These figures represent the relationship between homologous temperature, T/T_m (abscissa), and SCR (ordinate) for each sample. The abscissa T/T_m denotes the ratio of lower punch temperature to the melting point of butyl p-hydroxybenzoate in terms of absolute temperature. Each plot in Figs. 4—6 represents the average and standard deviations of SCR in 100 tablets. The SCR indicated a maximum value at $T/T_m = 0.90$ for every sample. Mechanical properties such as flowability and compressibility of powders are influenced by the elevation of temperature during handling of the powders, even below the melting point. York and Pilpel[10] and Otsuka et al.[11] have studied the tensile strength of an organic powder bed as a function of temperature, and they found that the tensile strength reached a maximal value when the homologous temperature, T/T_m is 0.60 to 0.90. We statistically analyzed the data by applying the significant difference test. There was a significant difference between maximum SCR values when T/T_m is 0.90, and these values occurred at a confidence limit of 95%. These changes indicate that sintering occurred during contact, as reported elsewhere.[12] The occurrence of sintering is still uncharted, because the tablets were compacted very quickly in this study. However, it is satisfactory to consider that the contact area between the lower punch surface and powder particles is maximal at about $T/T_m = 0.90$. This suggests that the adhesive force is the strongest between the tablet surface and the lower punch surface. These phenomena were remarkable and occurred
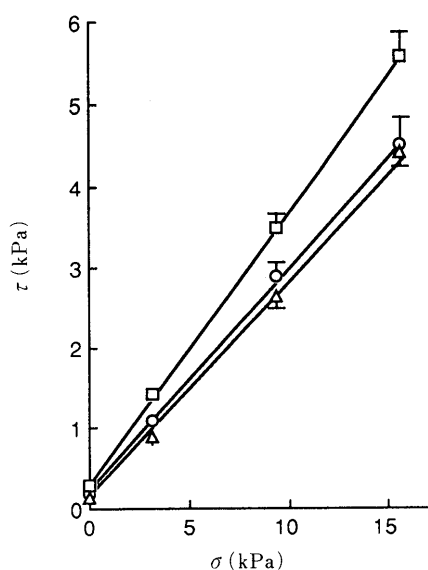
in samples III (Fig. 5) and VI (Fig. 6), including large amounts of small particles. Sticking occurred under lower compression pressure, and the probability of sticking was high. A precise solution to the problem was not obtained. However, if the adhesive force between particles was less under low rather than high compression, then the adhesive force between the punch and the particles would be high.

Sticking occurred in samples I, II and VI, when $T/T_m = 0.90$ under conditions of low compression. On the other hand, the SCR values were maximal when T/T_m was 0.90 for samples IV and V, but there was no sticking. Sticking can be prevented by mixing small amounts of small particles with large particles; however, this solution is ineffective when large amounts of small particles are added, as in sample VI. It is considered that the large standard deviation of SCR is caused by the occurrence of sticking.
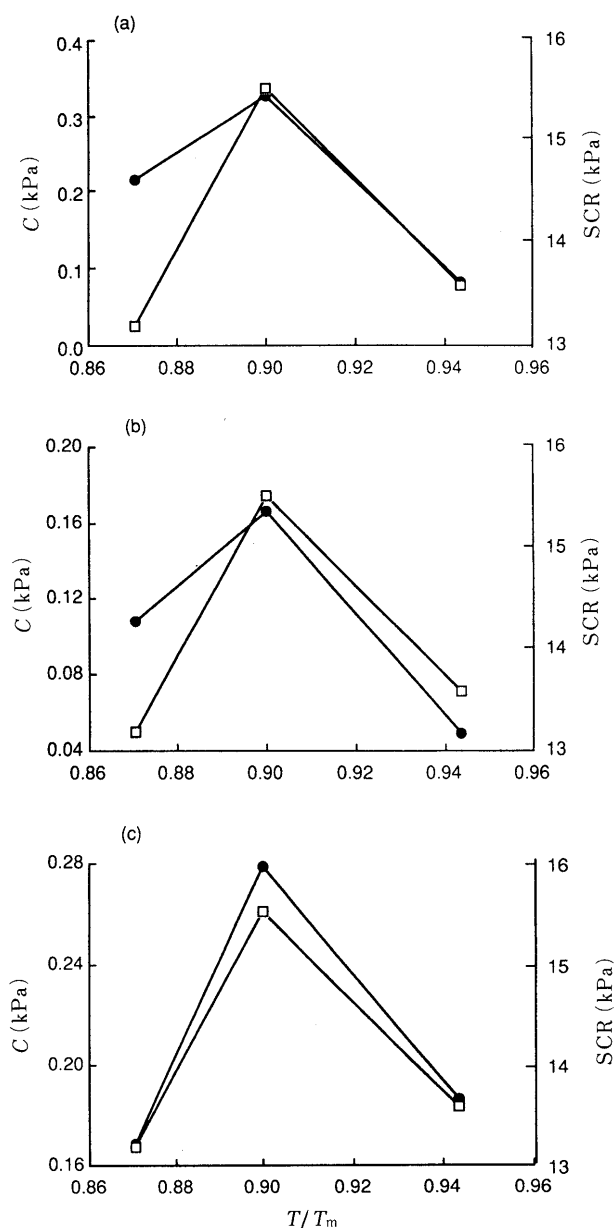

Fig. 7. Plots of Normal Stress (σ) against Shear Stress (τ) for Sample I and Tutanaga Plate

○, 25 °C; □, 35 °C; △, 50 °C.

Fig. 8. Relationship between Shear Strength (C) and SCR as a Function of Homologous Temperature (T/T_m)

(a) $R_{max} = 64 \mu$m, (b) $R_{max} = 7.2 \mu$m, (c) $R_{max} = 3.8 \mu$m. ●, C; □, SCR.

Effects of Temperature and Surface Roughness of the Metal Plate on Shear Strength Sticking was caused by the smoothness of the punch surface. Toyoshima et al.[6] reported the relationship between sticking and tablet surface roughness. Thus, we considered that the smoothness of the punch surface may play an important role in the sticking.

This phenomenon was observed by detecting SCR during compression at a homologous temperature $(T/T_m = 0.90)$. We considered that the SCR represented the shear stress between the tablet surface and lower punch surface. Therefore, we examined the relationship between SCR and shear strength by performing a shear test for each sample at each temperatute.

Figure 7 shows the relationship between normal stress σ and shear stress τ. These plots provide a linear relationship for each sample and are fitted to the following modified Coulomb's equation.

$$\tau = \mu\sigma + C \tag{3}$$

where μ is the friction coefficient and C is the shear strength.

Figure 8 shows the relationship between shear strength, C, and SCR, as a function of homologous temperature, T/T_m. The shear strength, C, and SCR values of all samples were maximal at a T/T_m of 0.90. Though not indicated here, the friction coefficient, μ, showed a similar tendency at shear strength, C.

We assumed that the shear strength and friction coefficient values would increase with increasing surface roughness of the metal plate. However, the shear strength and friction coefficients were minimal when using a stainless steel plate I with a 7.2 μm R_{max} value. The contact area between the model tablet and the stainless steel plate II $(R_{max} = 3.8 \mu m)$ was larger than with stainless plate I $(R_{max} = 7.2 \mu m)$ under a small amount of normal stress. On the other hand, for tutanaga steel plate $(R_{max} = 64 \mu m)$, the large frictional resistance due to its large concavo-convex texture can increase the shear strength and friction coefficient as a matter of course.

These results indicate a tendency towards SCR, similar to the effect of temperature.

Conclusion

Under constant conditions, such as temperature and humidity, tableting troubles such as sticking and picking arise easily in samples that have low melting points and a small particle diameter. Moreover, the temperature of the tableting machine and the compression stress have an influence on sticking.

SCR and $\bar{x}$–R control charts are an effective means of detecting sticking.

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