

## **ON PREDICTION OF CONCENTRATION OF LIQUID FOOD BY ACOUSTIC NON-LINEAR PARAMETER B/A**

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### **ABSTRACT**

The purpose of this study is to investigate the possibility of the non-destructive quality evaluation for food by the acoustic non-linear parameter B/A which is a measure of the non-linearity of the state equation of the medium in terms of pressure and density.

The B/A of water, corn oil O/W(oil in water) emulsion and milk were measured by using a sound velocity measuring system. The B/A value of water was measured for ascertaining reliability of our experimental system. Corn oil O/W emulsion was prepared as a model of milk. It was proved that the B/A value of O/W emulsion was related to the oil concentration by a law of mixture. We applied this result to milk and obtained satisfactory results for predicting the milk fat concentration.

**Key Word :** Non-destructive evaluation, Quality, Liquid food, Emulsion, Milk, Acoustic non-linear parameter, B/A, Fat, Solids-non-fat, Ultrasonic

### **INTRODUCTION**

Recently the quality evaluation of agricultural products has been rising in importance, because the quality is one of the most important indices for the determination of the market price. And in the near future the necessity of the quality evaluation will increase more and more on account of trade liberalization for agricultural products.

The quality of agricultural products must be inspected non-destructively and quickly. Various reports have been given on non-destructive methods over a long time. Among many non-destructive methods, the ultrasonic method can be put to actual easily because it doesn't need the expensive instruments and the complicated system.

The sound velocity and the absorption coefficient are well-known as the ultrasonic characteristics of the agricultural products. Since those acoustic parameters reflect the static properties of medium, e.g. macro or micro structure and the elasticity, the non-destructive inspection using the sound velocity and the absorption coefficient seems to have its limit. In medical engineering field, therefore, attention has come to be paid to an acoustic non-linear parameter recently. The B/A is a degree which expresses an non-linear interaction between the medium and the sound wave and is defined as the pressure dependency of sound velocity. Depending on such a definition, it can be said that the B/A reflects the pressure dependency of the elasticity. Hence, the B/A may have more information than the static properties of the medium, e.g. medium composition, maturity and existence of decay parts in the medium.

In the present report we describe an experimental study of the relationship between concentration and the B/A for two-phase dilute liquid (corn oil O/W emulsion) and examine the possibility of predicting the fat concentration of milk non-destructively by measuring the B/A of milk.

## PRINCIPLE

### Acoustic non-linear parameter

The non-linear parameter of the liquid medium is obtained from the adiabatic equation of state, i.e. the relationship between the pressure and the density of the medium for an adiabatic process. If this relation is expanded in a Taylor's series, the following equation is obtained :

$$P = P(\rho_0) + \rho_0 \left( \frac{\partial P}{\partial \rho} \right)_{S, \rho_0} \left( \frac{\rho - \rho_0}{\rho_0} \right) + \rho_0^2 \left( \frac{\partial^2 P}{\partial \rho^2} \right) + \dots \quad (1)$$

where P, P( $\rho_0$ ),  $\rho$  and S are the instantaneous pressure, the hydrostatic pressure, the density and entropy and the suffix "0" indicates the equilibrium value. Those derivatives in Eq. 1 can be limited to terms through the quadratic because the higher order is too small. On one hand, according to the wave theory, the following equation for the sound velocity in the liquid medium is well-known :

$$c_0^2 = \frac{\partial P}{\partial \rho} \quad (2)$$

Consequently Eq. 1 can be rewritten as

$$P = P_0 + A \left( \frac{\rho - \rho_0}{\rho_0} \right) + B \left( \frac{\rho - \rho_0}{\rho_0} \right)^2 \quad (3)$$

where  $A = \rho_0 c_0^2$  and  $B = 2\rho_0^2 c_0^3 \left( \frac{\partial c}{\partial P} \right)_{S, \rho_0}$

The acoustic non-linear parameter B/A is defined as the ratio B/A ;

$$\frac{B}{A} = 2\rho_0 c_0 \left( \frac{\partial c}{\partial P} \right)_{S, \rho_0} \quad (4)$$

and this parameter indicates the degree of the distortion of finite amplitude waves in the liquid medium.

### Measurement

A lot of measuring methods of B/A have been proposed. They can be almost classified by two methods; a thermodynamic method and a finite amplitude method.

The thermodynamic method requires the measurement of the change in the sound velocity with the adiabatic (or isentropic) change in pressure. This method is based on Eq. 4 and has the high accuracy. On the other hand, the finite amplitude method uses the distortion of the wave in the liquid medium. Since the theory of the distortion needs to include the diffraction and the attenuation, it is complicated to measure the B/A using this method and it is said that the accuracy is lower than the thermodynamic method. This tiwhere T,  $\varepsilon$  and  $C_p$  indicate temperature, thermal expansion coefficient and specific heat at constant pressure. In Eq. 5 the first term and the second term represent isothermal processme we made use of the thermodynamic method because this one doesn't need the complicated measuring system.

According to Eq. 4 the isentropic change is required in order to get the B/A, but it is hard to realize the isentropic process.

Eq. 4 can be rewritten as follows :

$$\frac{B}{A} = 2\rho_0 c_0 \left( \frac{\partial c}{\partial P} \right)_{T, \rho_0} + \frac{2c_0 T \epsilon}{C_P} \left( \frac{\partial P}{\partial T} \right)_{P, \rho_0}, \quad (5)$$

and equal-pressure process respectively, and it is possible to measure each term experimentally.

It is well-known theoretically that the second term in Eq. 5 is much smaller than the first one. Therefore we neglected the second order and used the following equation for the calculation of the B/A :

$$\frac{B}{A} = 2\rho_0 c_0 \left( \frac{\partial c}{\partial P} \right)_{T, \rho_0} \quad (6)$$

We only measured the change in the sound velocity versus the change in pressure under isothermal condition.

## MATERIALS AND METHODS

### Materials

The corn oil O/W emulsions were prepared according to the inversion emulsification method. The corn oil (Ajinomoto, Co.) containing 5 wt% of the lecithin powder from soybean (Nacalai Tesque) was homogenized in water at 3000rpm for 3 min.

The homogenized milk (Snow Brand, Co.) was obtained from the market. In order to change the percentages of milk fat, the skim milk (Snow Brand, Co.) and distilled water were added to the 1% or 3.5% or 4.5%-fat milk.

### Method of Measuring B/A

A block diagram of the experimental setup is shown in fig. 1. The pressure chamber contains two piezoelectric ceramic discs (Dia.=15mm, Resonance Frequency=0.5MHz) separated by 100mm. The transmitting transducer was excited by 0.5MHz burst pulses that were obtained by gating a sinusoidal wave generated by a function generator. Longitudinal ultrasonic pulses are radiated into the liquid medium which exists between those two ceramic discs, and then the ultrasonic waves transmitted through the medium are transformed into electric signals by the receiver. These signals are transferred to a computer through a digital storage oscilloscope.

The pressure vessel of the inside diameter 20mm and the inside length 100mm is made of stainless steel. The pressure is generated by a piston which is operated manually. The static pressure in the chamber was measured by a pressure gage. Because too high pressure causes degeneration of the medium, we decided to limit the applied pressure to about 1MPa.

The pressure vessel was immersed in a water bath. In the bath water temperature was held at a fixed temperature within 0.1K by a temperature control equipment. The temperature in the chamber was observed by a copper-constantan thermo couple.

First we filled the chamber with sample liquid (about 30ml) and put the vessel in the water bath where the temperature was kept at the setting value. Ultrasonic waves were input to the sample medium at the fixed temperature and

atmospheric pressure, and the received signals were sent to the computer via the digital storage oscilloscope. Then the inside pressure was slowly increased by the setting value. Since during this process the inside temperature was slightly changed, after equilibrating to the setting value the above-mentioned measurement was done.

The sound velocity are determined by the relation between the time required for transmission and the distance between the two transducers. In our experimental system the sound velocity can be calculated from the distance 100mm and the propagation time between the trigger signal and the received signal. Fig. 2 shows the received signals both before and after applying pressure into the chamber. The value of sound velocity difference between those two waves is needed to determine the B/A . But this difference was very small because the inside pressure was limited in order to prevent degeneration of the medium and the distance between the transducers cannot be too long because of wave attenuation. Therefore one of those waveforms was shifted along time base until the Lissajous figure about those waveforms became linear line (Fig. 3), and the sound velocity difference was obtained by the value of the phase shift.

## RESULTS AND DISCUSSION

Fig. 4 shows that the sound velocity of water increases linearly with hydrostatic pressure. We obtained the B/A by substituting the slope into Eq. 6. The slope was calculated by the least squared error method. The B/A of water was 5.28 at 283K. The B/A of water reported by Beyer et al. was 4.83 at 293K, and most researchers reported that the B/A of water was about 5 at about 290K. Therefore we considered that our measuring system was reasonable for getting the B/A.

Values of density, sound speed and acoustic non-linear parameter for water, corn oil and market milk are shown in Tab. 1.

The O/W emulsion is classified into a two-phase disperse system, as shown in Fig. 5a. Water is a matrix (continuous phase) and oil particles are dispersed in the matrix. If we consider that the ultrasonic wave is propagated straight on one line in the medium, the total distance propagated through oil phase by sound wave should be proportional to the volume fraction of oil in the emulsion. Therefore we can consider that this system is made up of the oil layer and the water layer which are placed perpendicularly to the direction where the ultrasonic waves propagate, as shown in Fig. 5b. Such an approximation is called one-particle approximation.

Assuming that the propagation time of the ultrasonic waves through the whole system equals the sum of the propagation time through the oil layer and the water layer, we obtain the following equation :

$$\frac{1}{c} = \left( \frac{1}{c_d} - \frac{1}{c_c} \right) \varnothing_d + \frac{1}{c_c} \quad (7)$$

where  $\rho$ ,  $c$  and  $\varnothing$  are the density, the sound velocity and the volume fraction respectively and the suffix "d" and "c" indicate the disperse phase (oil) and the continuous phase (water). Eq. 7 means that the inverse of the sound velocity is proportional to the oil volume fraction by a law of mixture. Fig. 6a shows the plot of  $1/c$  as a function of corn oil concentration in the corn oil O/W emulsion at 283K. There is a linear dependence on corn oil concentration as predicted by theory. In Fig. 6a the solid line represents the theoretical data calculated by substituting the

data in Tab. 1 into Eq. 7. The experimentally-determined  $1/c$  agree approximately with the theoretically-determined data.

Now assuming that the total volume change of the whole system by applying pressure equals the sum of the volume change of the oil phase and of the water phase, we obtain the following equation for the B/A :

$$\left(\frac{1}{\rho c^2}\right)^2 \left(\frac{B}{A}\right) = \left(\frac{1}{\rho_c c_c^2}\right)^2 \left(\frac{B}{A}\right)_c (1 - \phi_d) + \left(\frac{1}{\rho_d c_d^2}\right)^2 \left(\frac{B}{A}\right)_d \phi_d \quad (8)$$

According to Eq. 7 and Eq. 8, the effective B/A is not proportional to the oil volume fraction. In such a case as

$$\rho = \rho_c = \rho_d \quad \text{and} \quad c = c_c = c_d \quad ,$$

Eq. 8 simplifies to the following form :

$$\frac{B}{A} = \left(\left(\frac{B}{A}\right)_d - \left(\frac{B}{A}\right)_c\right) \phi_d + \left(\frac{B}{A}\right)_c \quad (9)$$

In this case a law of mixture can be applied for relationship between the effective B/A and the oil phase concentration. Fig. 6b shows the plot of the B/A as a function of corn oil concentration at 283K. In Fig. 6b the solid line and the broken line represent the data calculated by Eq. 8 and ones calculated by Eq. 9 respectively. Below 10 percent of oil concentration the difference between the former data and the latter is below 0.35 percent. Therefore we can use a law of mixture on B/A for the dilute O/W emulsion. While the difference between the experimentally-determined B/A data and the theoretical data becomes more significant above 10 percent of oil concentration, both data are approximately agreed below 8 percent concentration.

In the discussion that follows, we investigate the relationships between acoustic measurements and milk components. Assuming that milk is two-phase emulsified system, separating the milk fat part and the non-fat solution, we can expect that the results of the corn oil O/W emulsion are applied to milk.

Fig. 7a shows the relationship between the inverse of the sound velocity and milk fat concentration at 283K. The milk fat content had no effect on the inverse of the sound velocity. We believe that it was caused by no difference between the sound velocity in milk fat phase and the sound velocity in non-fat continuous phase. Therefore it is impossible to predict the fat content of milk by determining the sound velocity. Fig. 7b shows the relationship between the B/A of milk and milk fat concentration at 283K. The B/A of milk increased linearly with the milk fat concentration and the correlation coefficient was 0.974. The fat concentration of non-processed raw milk changes between 3 and 6 percent, and within this region the B/A is more effective in predicting the fat content of milk.

The other representative component is solids-non-fat (SNF). Generally the changes of SNF content of raw milk are less significant than those of fat content. In Japan SNF contents of raw milk change between 8 and 9 percent. Next we observed the relationship between the SNF content and the acoustic measurements for milk. Fig. 8a shows that the inverse of the sound velocity decreases linearly with the SNF content. The changes of the sound velocity by changing the SNF content are more significant than by changing the fat concentration. Therefore it can be said that the sound velocity in the non-fat solution part is very different from one in the milk fat part. Fig. 9a shows the  $1/c$ -fat content relation's dependence on the SNF content. The intercept of each line decreases gradually when increasing the SNF content of milk. We think this is because  $c_c$  in Eq. 7 increases. Fig. 8b shows

the relationship between the B/A and the SNF content and in this figure the solid line and the broken line represent the non-fat milk and 3.5%-fat milk respectively. The linear relationship between the milk fat concentration and the B/A is influenced by the SNF content. From this result we can say that the B/A of the non-fat continuous phase depends on the SNF content. Fig. 9b shows the B/A-fat content relation's dependence on the SNF content. When the SNF content does not change, the relationship between the B/A and the fat concentration is linear. The slope of each line should decrease gradually when increasing the SNF content of milk if we apply the Eq. 9 apply for these cases, but in Fig. 9b the slope increases. From this result the Eq. 9 is not effective for milk because the density and the sound velocity of the non-fat solution are larger than those of milk fat part and the conditions for simplifying Eq. 8 are not given. In view of the argument presented above, we need to treat milk as fat-SNF-water system for our research.

## CONCLUSIONS

We got the following conclusions :

- (1) An one-particle approximation for the dilute corn oil O/W emulsion was effective, and the corn oil concentration was related with the inverse of the sound velocity and the B/A by a law of mixture.
- (2) The relationship between the B/A of the homogenized milk and its fat concentration was linear.
- (3) The milk fat content had no effect on the inverse of the sound velocity. Therefore we should not predict the fat concentration of the homogenized milk by the sound velocity, but the B/A.
- (4) For applying the B/A on predicting the fat concentration of the non-processed raw milk we should regard the milk as three-phase (milk fat, SNF and water) system.

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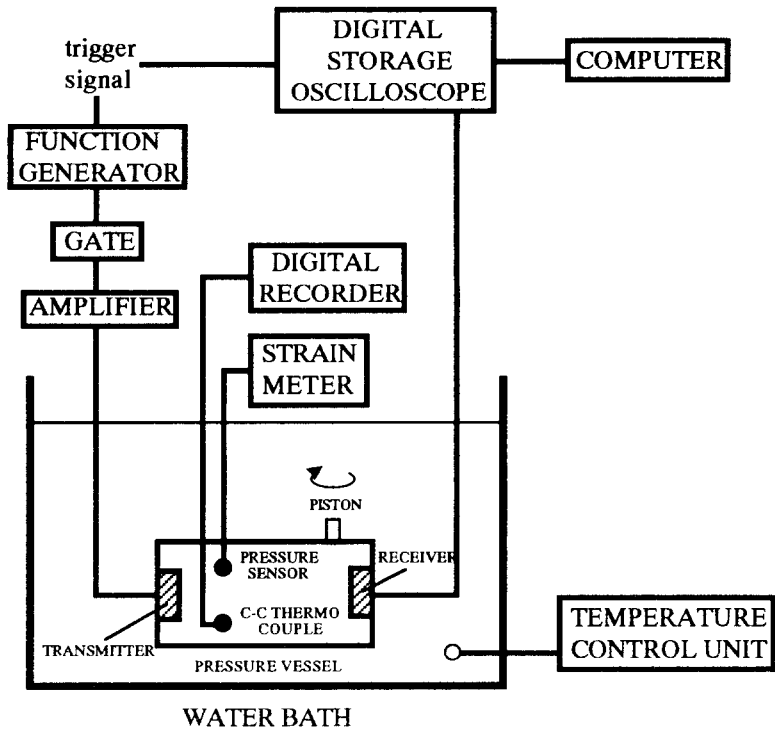


Figure 1 : Block diagram of the experimental setup for measuring acoustic parameters

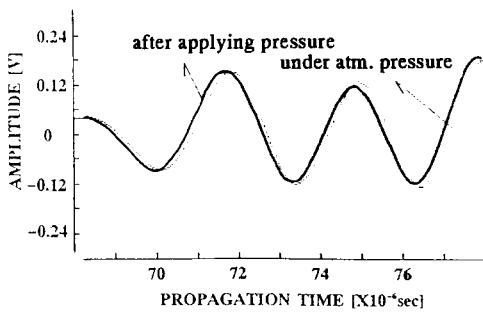


Figure 3 : Received waveforms before and after applying pressure

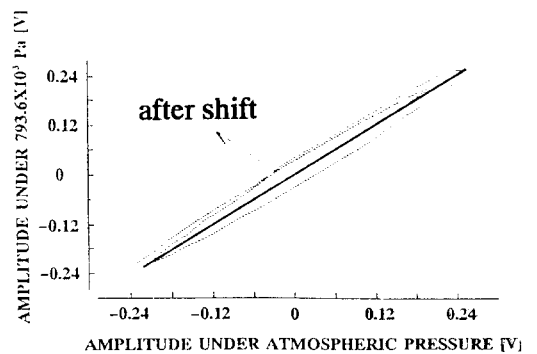


Figure 4 : Lissajous figures before and after shift

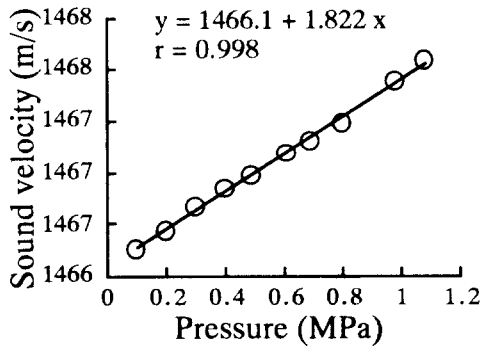


Table 1 : Specific gravity, sound velocity and B/A of experimental samples at 283K

	Specific Gravity	Sound velocity (m/s)	B/A
Water	1.0	1447.59	5.28
Corn oil	0.9	1542.7	11.14
Milk	1.03	1486.9	6.07

Figure 4 : Relationship between sound velocity and pressure for water at 283K

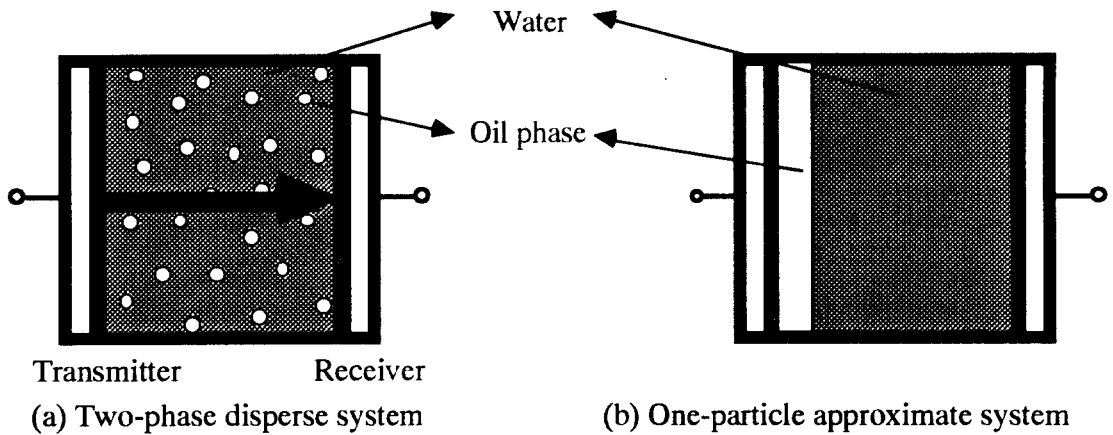
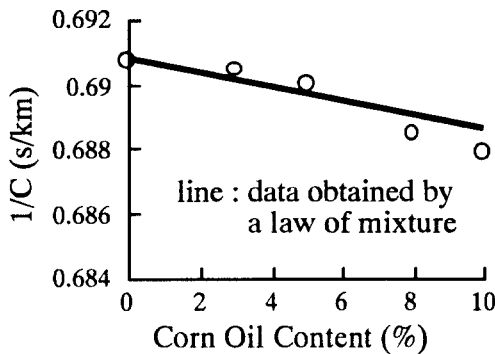
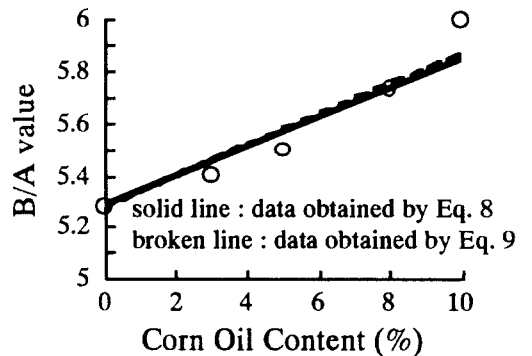


Figure 5 : Schematic diagrams for O/W emulsion



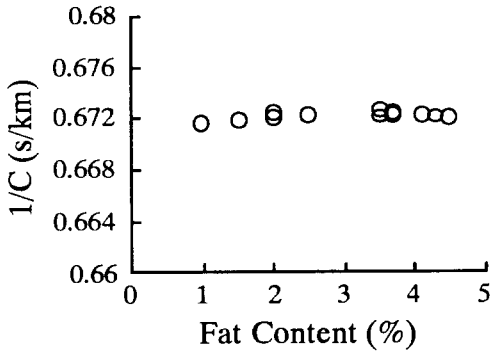
(a) 1/c-oil content relationship



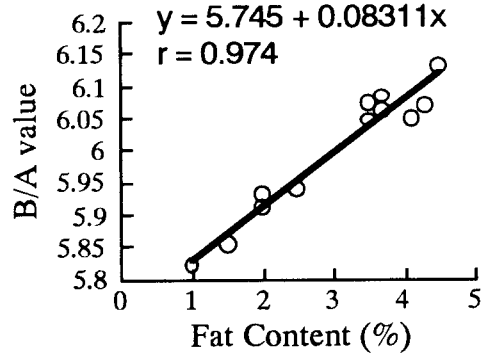
(b) B/A-oil content relationship

Figure 6 : 1/c and B/A as functions of oil content in corn oil O/W emulsion at 283K



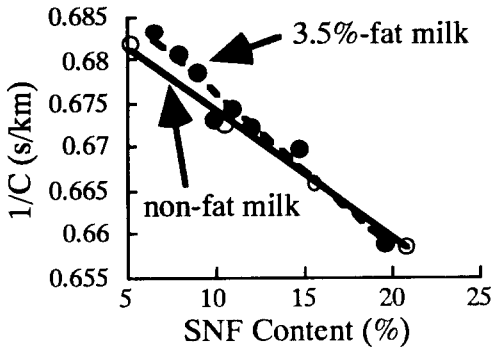


(a) 1/c-fat content relationship

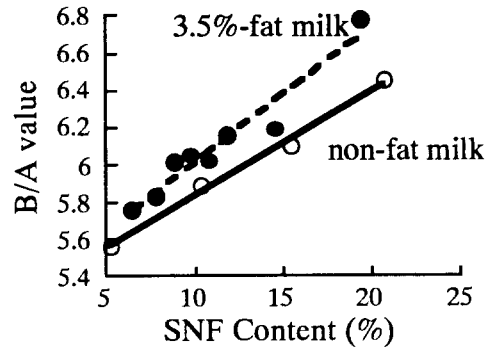


(b) B/A-fat content relationship

Figure7 : 1/c and B/A as functions of fat content in 8.5%-SNF milk at 283K

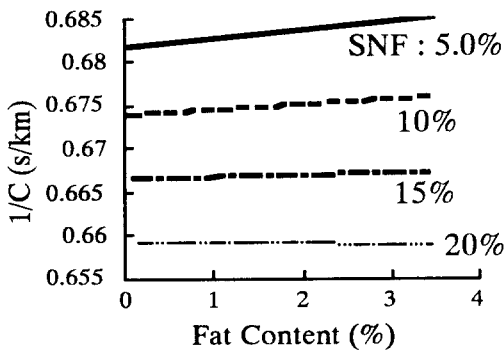


(a) 1/c-SNF content relationship

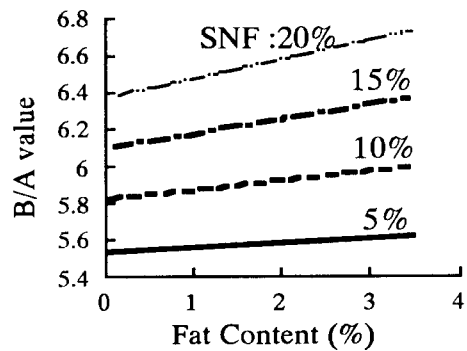


(b) B/A-SNF content relationship

Figure8 : 1/c and B/A as functions of SNF content in non-fat and 3.5%-fat milk at 283K



(a)



(b)

Figure8 : 1/c- and B/A-fat content relation's dependences on SNF content for milk at 283K