

## Elastic constants of sodium chlorate from 77 K to 350 K

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**Abstract.** The elastic constants  $C_{11}$ ,  $C_{12}$  and  $C_{44}$  of sodium chlorate single crystal have been evaluated using 10 MHz ultrasonic pulse echo superposition technique. The values are  $C_{11} = 4.90$ ,  $C_{12} = 1.39$ ,  $C_{44} = 1.17$  ( $\times 10^{10} N/m^2$ ) at 298 K and 6.15, 2.16, 1.32 ( $\times 10^{10} N/m^2$ ) at 77 K. The data agree well with the values measured earlier up to 223 K. Brief mention is also made of the low temperature bonding problems in these soft crystals.

**Keywords.** Elastic constant; sodium chlorate; ultrasonic velocity.

### 1. Introduction

The measurement of elastic wave velocities of solids by ultrasonic methods has now become a well established technique. The pulse methods have generally proved to be accurate, versatile and adaptable to a variety of environmental conditions. In precision high resolution measurement, one has to take into account the diffraction effects and above all the effect of the coupling between the transducer and the specimen. Even though the dimensions, etc. of the transducer and the specimen may be known precisely, the dimensions of the bonding material are known only approximately and further the properties of the bond (transducer to specimen) change appreciably with temperature, pressure, etc. A second important problem is the location of the start of the echo signal. Because of the finite bandwidth of the acoustic system, the signals have a finite rise time. Thus, the location of the leading edge leaves some uncertainty. The detailed procedure necessary to take these effects into account have been discussed in the literature (Mason 1964, Eros and Reitz 1958).

A method originally suggested by McSkimin (1961, 1962) based on the principle of pulse superposition takes into account all these effects and is capable of great accuracy and precision. The measurement of the pulse-echo time is converted to the measurement of the repetition rate frequency which causes constructive interference among the echo signals. We have used a 10 MHz ultrasonic pulse interferometer developed by Srinivasan *et al* (1974) for the velocity measurements on sodium chlorate single crystals up to 77 K. The elastic constants  $C_{11}$ ,  $C_{12}$  and  $C_{44}$  are evaluated and presented in this note. Earlier measurements extending up to 223 K are also discussed. It is found that great care has to be taken in the selection of proper bond materials for different temperature regions.

## 2. Experimental procedure

Single crystals of laboratory reagent grade sodium chlorate were grown by slow evaporation of the saturated aqueous solution. Clear samples cut along the [100] and [110] crystallographic directions were used. Coaxial gold-plated X and Y cut 10 MHz quartz transducers obtained from Bharat Electronics Ltd. were used to generate the longitudinal and transverse waves respectively in the specimen. The velocities were measured to an accuracy of about 10 ppm using the pulse interferometer developed for the purpose.

For the longitudinal waves along the [100] direction,  $v_L = [C_{11}/\rho]^{1/2}$ ,  $\rho$  being the density of the specimen, while the transverse waves give  $v_T = [C_{44}/\rho]^{1/2}$ . The [110] direction yields  $v_L = [(C_{11} + C_{12} + 2C_{44})/2\rho]^{1/2}$  for the propagation of longitudinal wave and  $v_{T_1} = [C_{44}/\rho]^{1/2}$  and  $v_{T_2} = [(C_{11} - C_{12})/2\rho]^{1/2}$  for shear wave velocities. The corrections due to the piezoelectric effects are negligible at the present accuracy of the measurements.

Having obtained  $C_{11}$  and  $C_{44}$  directly from longitudinal and shear wave velocities along the [100] direction,  $C_{12}$  was evaluated from the longitudinal velocity along the [110] direction. The shear wave measurement along [110] was used to check the data. Using a simple cryostat, velocities were measured down to liquid nitrogen temperature. The specimen was cooled at the rate of 10 K/hr in order to avoid cracking due to thermal shocks.

## 3. Sample preparation and selection of bond material

In experiments using pulse-echo interferometer, great care has to be taken in the preparation of the sample and the selection of proper seal materials (Fuller *et al* 1974) if accuracies of 0.1% or better are aimed at. If the specimen end faces are not parallel, wedge effects and wall reflections affect the shape of the echoes and it will be difficult to locate the precise maxima while superposing the echoes which is the main criterion in the velocity measurement using the pulse-superposition technique. Hence the end faces of the specimen should be plane and parallel to better than about 100 ppm. A hand lapping tool has been fabricated to achieve the above result.

It has been experimentally found that the quartz transducer must be physically bonded to the specimen in order to obtain good echoes suitable for precise velocity measurements. The choice of the bond material used to bind the quartz transducer to the specimen must be made according to the temperature range over which the measurements are to be made. The reason for this is that as the temperature is lowered, differential thermal contraction between the quartz, the seal material and the specimen may result in many failures. The foremost is the cracking of the bond material so that the quartz transducer is no longer in contact with the specimen. The second is that stresses introduced in the quartz crystal may deform it and hence change its piezoelectric properties with the result that good echoes are no longer received. Sometimes the thermal stresses may be of sufficient amplitude to cleave the specimen to some extent. It is also imperative to have a very thin bond formed in order to locate the maxima precisely.

It is found experimentally that no one bond material could be used over the entire temperature range with NaClO<sub>3</sub>. Phenyl salicylate (Salol) has been found satisfactory from 315 K to about 280 K. At lower temperatures the bond gives

way. Nonaq stopcock grease is one of the most popular bond materials. With nonaq stopcock grease the bond fails at about 190 K. This has been observed in many other soft crystals also (Overton and Swim 1951, Briscoe and Squire 1957, Norwood and Briscoe 1958, Garland and Renard 1966, Ho and Ruoff 1967, Lewis *et al* 1967, Garland and Novotny 1969, Lin and Wong 1972). For temperatures below 190 K and down to liquid nitrogen temperature, several materials have been used, like 2.5 M centistoke DC-200 Dow Corning silicone oil (Chang and Graham 1966), a mixture of 50% isopentane and 50% rubber cement (Fugate and Schule 1966) and an organic mixture of ethyl ether, ethyl alcohol and pentane (Cleavolin *et al* 1972, Marshall *et al* 1967, Marshall and Miller 1967).

In the present experiments an organic mixture consisting of 5 parts (by volume) of ethyl ether, 6 parts of isopentane and 2 parts of ethyl alcohol has been used. This mixture is applied at a temperature of about 150 K and was satisfactory down to 77 K. For measurements above room temperature, dihydroxybenzene is used as the bond material. In all the experiments cooling/heating rate did not exceed 10K/hr.

#### 4. Results and discussion

Assuming the density of sodium chlorate to be  $2.485 \times 10^3 \text{ kg/m}^3$ , at 25° C the elastic constants are calculated from the velocity measurements. In table 1, the elastic constants measured by earlier workers by different methods are presented along with our own values. It is seen that the agreement is excellent especially when compared with the other pulse echo and resonance measurements.

The elastic constants at low temperatures are evaluated taking into account the thermal expansion data reported by Ganesan (1959). The low temperature varia-

**Table 1.** Comparison of the values of elastic constants of sodium chlorate crystal at 25° C

Source	Method	$C_{11}$	$C_{12}$ (in $10^{10} \text{ N/m}^2$ units)	$C_{44}$
Mason (1946)	Resonance	4.89	1.38	1.17
Bhagavantam and Suryanarayana (1947)	Wedge	5.09	1.55	1.18
Sundara Rao (1949)	Wedge	4.92	1.45	1.19
Jona (1950)	Shaeffer-Bergmann	4.89	1.39	1.17
Bechman (1951)	Resonance	4.99	1.41	1.17
Ramachandran and Wooster (1951)	Diffuse x-ray	4.80	1.50	1.20
Haussühl (1964)	Shaeffer-Bergmann	5.00	1.55	1.16
Viswanathan (1966)	Pulse echo	4.92	1.42	1.16
Radha <i>et al</i> (1968)	Pulse echo	4.99	1.45	1.13
Present values (1974)	Pulse echo superposition	4.89 <sub>9</sub>	1.38 <sub>9</sub>	1.17 <sub>4</sub>

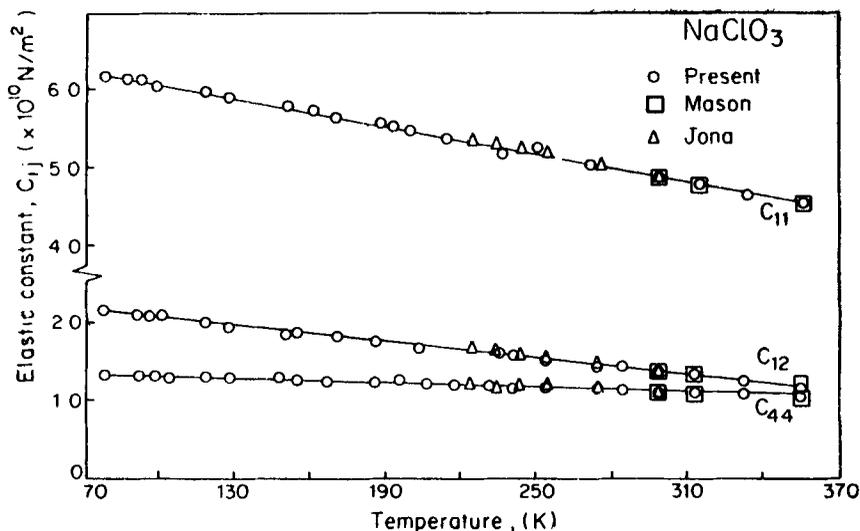


Figure 1. Temperature variation of elastic constants of NaClO<sub>3</sub>.

tion of the elastic constants is given in figure 1 along with the results obtained by Jona (1950) who has evaluated the three constants up to 223 K using a Schaefer-Bergman method. The agreement is seen to be quite good. The values of  $C_{11}$ ,  $C_{12}$  and  $C_{44}$  obtained at 77 K are 6.15, 2.16 and 1.32 ( $\times 10^{10}$  N/m<sup>2</sup>).

It is found that our values of elastic constants above room temperature agree well with Mason's (1946) reported values (elastic constants are calculated from the reported elastic compliances). However, from Mason's results one observes a small peak in the elastic constant around 343 K. In the present investigation  $C_{12}$  has been evaluated from a careful measurement of both shear and longitudinal velocities along [110] direction and no peak was observed.  $C_{12}$  gradually decreases with temperature as indicated in figure 1. Hence, we conclude that the peak observed in Mason's measurement is the result of scatter in the measurement.

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