



# Determination of elastic constants of a single-crystal topaz and their temperature dependence via sphere resonance method



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## ABSTRACT

Water and halogens in ocean floor sediments transported by a descending slab might play important roles in geodynamic processes. Imaging subducted sediments through seismological observations requires a thorough understanding of elastic properties of sediment origin hydrous minerals. Topaz is a sediment origin hydrous mineral, which is formed at the depth of 250–350 km on a cold subducting slab. We determined elastic constants and their temperature derivatives of a natural single-crystal of topaz ( $\text{Al}_{1.97}\text{SiO}_4(\text{F}_{1.56}, \text{OH}_{0.42})$ ) at the temperature from 271.5 to 312.7 °K by using the sphere-resonance method. Elastic constants at an ambient temperature ( $T = 291.9$  °K) are  $C_{11} = 281.21(1)$  GPa,  $C_{22} = 346.23(9)$  GPa,  $C_{33} = 294.99(9)$  GPa,  $C_{44} = 108.49(1)$  GPa,  $C_{55} = 132.47(1)$  GPa,  $C_{66} = 130.32(1)$  GPa,  $C_{12} = 121.48(3)$  GPa,  $C_{13} = 80.94(3)$  GPa and  $C_{23} = 81.77(2)$  GPa. Since our sample  $[\text{Al}_2\text{SiO}_4(\text{F}_{1.56}, \text{OH}_{0.42})]$  was relatively rich in fluorine, only small differences in elastic constants can be seen between our sample and fluorine end member. Elastic constants of OH-rich topaz should be experimentally investigated to understand the influence of F-OH substitution on elasticity of topaz. All the elastic constants decrease linearly with increasing temperature. The temperature derivatives are  $dC_{11}/dT = -0.014(3)$  GPa/°K,  $dC_{22}/dT = -0.010(7)$  GPa/°K,  $dC_{33}/dT = -0.021(5)$  GPa/°K,  $dC_{44}/dT = -0.011(1)$  GPa/°K,  $dC_{55}/dT = -0.016(2)$  GPa/°K,  $dC_{66}/dT = -0.010(2)$  GPa/°K,  $dC_{12}/dT = -0.0041(6)$  GPa/°K,  $dC_{13}/dT = -0.001(2)$  GPa/°K and  $dC_{23}/dT = -0.002(1)$  GPa/°K. The isotropic seismic velocities in topaz are distinctly higher than those in olivine at 10 GPa and 300–1400 °K. There should be a strong velocity contrast between the overlying mantle and the thin sediment-origin layer at the depth around 300 km. A seismological technique like the receiver function technique should be applied to detect a thin layer of topaz in a cold subduction zone.

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## 1. Introduction

Subduction zones are important tectonic sites, where water is brought into the Earth's interior by descending hydrated materials in oceanic sediments, crust and lithospheric mantle. Water brought by hydrated materials plays important roles in geodynamic processes. Most of water is released by dehydration reactions to cause slab earthquakes (e.g., Seno and Yamazaki, 2003) and magma generation in the overlying wedge mantle (e.g., Iwamori, 1998). Some part of water might be carried by hydrous phases into the mantle transition zone (at depths of 400–660 km), where  $\beta$  and  $\gamma$  phases of olivine can hold up to 3 wt.% of water (Kohlstedt et al., 1996). Imaging hydrated materials through geophysical observations gives us insights into the water transport in the Earth. Seismological inferences of hydrated materials have been reported for subducted oceanic crust (e.g., Kita et al., 2006) and lithospheric mantle (e.g., Seno and Yamazaki, 2003). Though oceanic sediments

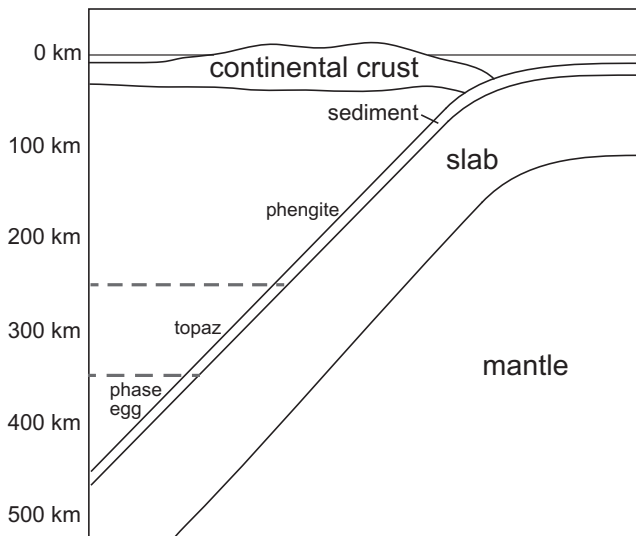
are suggested to carry water into the Earth's deep interior (e.g., Nakamura and Iwamori, 2009), sediment-origin hydrated materials have not been seismologically detected. A thorough understanding of elastic properties of sediment-origin hydrous minerals is essential for seismological detection.

Phase relations of subducted oceanic sediments have been experimentally studied. Domanik and Holloway (1996) conducted high pressure and temperature experiments on mixtures of minerals, the bulk chemical composition of which is between muscovite and phlogopite, and suggested that phengitic muscovite and topaz-OH be stable at the depth greater than 360 km if the temperature is lower than 1273 °K. Ono (1998) conducted high pressure and temperature experiments on a synthetic oceanic sediment, and showed that topaz-OH ( $\text{Al}_2\text{SiO}_4(\text{OH})_2$ ) is stable at 8–12 GPa and 1123–1473 °K. At higher pressures, phase egg ( $\text{AlSiO}_3(\text{OH})$ ) appears as the hydrous mineral with a large stability field (Fig. 1).

It should be noted that halogens in oceanic sediments have been overlooked in previous studies. Halogens, especially fluorine (520 ppm) and chlorine (850 ppm), are relatively abundant in

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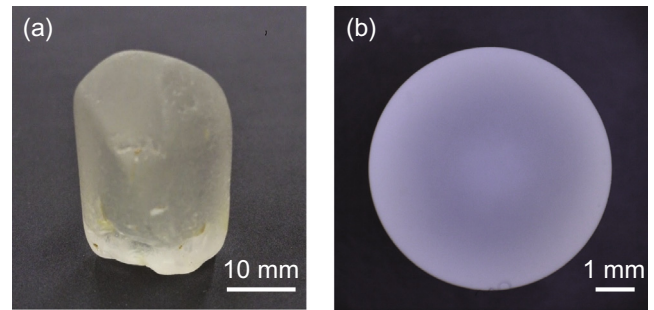
**Fig. 1.** A schematic illustration of a subducting cold slab. The major hydrous phase in the sediment layer changes from phengite to topaz and phase egg with increasing depth.

oceanic sediments (Carpenter, 1969; Muramatsu and Wedepohl, 1998). Because the ionic charge and radius of  $F^-$  resemble those of  $OH^-$ ,  $F^-$  is capable of substituting for  $OH^-$  into many hydrous minerals (Pyle and Mather, 2009). F-OH substitution can affect the mechanical strength of layered hydrous minerals. In micas, F-OH substitution eliminates the electrostatic repulsion between the interlayer cation and hydroxyls, thus enhancing the mechanical strength (Dahl and Dorais, 1996). The reduced electrostatic repulsion force is likely to be responsible for the enhanced thermal and pressure stability of fluorine-bearing hydrous phases (Mookherjee and Bezacier, 2012). Fluorine released by the breakdown of hydrous phases will also exert a significant impact on the genesis and evolution of magmas, as well as their viscosity (Beyer et al., 2012). The stability field should thus be investigated on fluorine-bearing hydrous minerals, as well as elastic properties. The starting material of Ono (1998) was free from fluorine, while that of Domanik and Holloway (1996) contained it.

Topaz is one of sediment-origin hydrous minerals. A pioneering work of Voigt (1888) determined elastic constants of topaz through bending and twisting of single-crystal prisms, though the chemical composition of the sample was unfortunately not shown. Later, Haussühl (1993) determined elastic constants of fluorine end member topaz ( $Al_2SiO_4F_2$ ) by the rectangular-parallelepiped resonance method. More recently, Mookherjee et al. (2016) determined elastic constants of topaz-OH in the case of monoclinic and orthorhombic symmetries by the first principle calculation. Since no measurements have been made on topaz containing  $OH^-$ , the influence of F-OH substitution on elastic properties has been poorly understood. In order to study the influence of F-OH substitution, we determined elastic constants of a natural single crystal topaz, which contained  $F^-$  and  $OH^-$ , and their temperature dependence through the sphere resonance method. The seismological detectability of topaz in subduction zones is also discussed on the basis of isotropic seismic velocities at high pressure and temperature.

## 2. Specimen

A transparent crystal (25 mm × 15 mm × 15 mm) of topaz (Fig. 2(a)), in which no cracks or inclusions were seen, was collected from Fukuoka, Nakatsugawa city, Gifu prefecture, Japan.



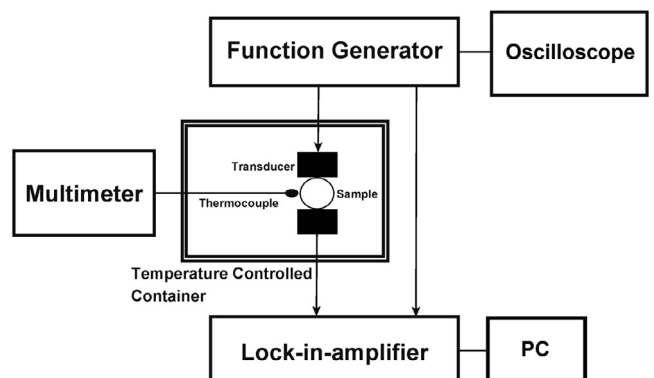
**Fig. 2.** Photographs of the topaz specimen before (a) and after (b) shaping.

The uniformity of crystallographic orientation was examined on a polished surface of the crystal by SEM-EBSD system (JEOL, JSM6300 with HKL Channel 5) at the Center for Instrumental Analysis, Shizuoka University. The chemical composition was determined by an EPMA (JEOL, JXA-8500F) at JAMSTEC Yokosuka Headquarters and the chemical formula is estimated to be  $Al_2SiO_4(F_{1.56}, OH_{0.42})$ . The orthorhombic symmetry was confirmed by the powder X-ray diffraction method, and the lattice parameters  $a$ ,  $b$  and  $c$  were determined to be 4.640(1), 8.794(3) and 8.356(3) Å, respectively.

The crystal was shaped into a sphere by the two-pipe method (Bond, 1954). The average and standard deviation of diameter were 6.483(1) mm and 0.001 mm, respectively for 30 measurements (Fig. 2(b)). The ratio of the standard deviation to the average was 0.02%. The density was calculated to be 3.575(2) g/cm<sup>3</sup> from the mass and volume of the sphere.

## 3. Sphere resonance method

The sphere resonance method (e.g., Soga and Anderson, 1967) was employed to determine elastic constants. A schematic diagram of the measurement system is shown in Fig. 3. The spherical specimen was held between two ultrasonic transducers (OLYMPUS, V156RM). A function generator (SONY Tektronix, AFG320) applied a sinusoidal signal to the driving transducer, and the excited vibration in the specimen was picked up by the receiving transducer. The lower transducer was placed on a balance to apply the specimen-holding force. A lock-in-amplifier (Stanford Research Systems, SR884) was used to measure the amplitude of the excited vibration. The frequency of the sinusoidal signal was changed from 0.6 MHz to 1.5 MHz with the frequency step of 100 Hz. The transducers, sample and balance were placed in a temperature-controlled container. The temperature was changed from



**Fig. 3.** A schematic diagram of the measurement system.

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