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Measurement of unfrozen water content and relative permittivity of frozen unsaturated soil using NMR and TDR

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ABSTRACT

The soil-freezing characteristic, the relationship between unfrozen water content and temperature, is relevant for any mass transfer processes in frozen porous media. To determine the soil-freezing characteristic, we simultaneously measured liquid water content and relative permittivity of various unsaturated soils at above-zero and subzero temperatures by using pulsed nuclear magnetic resonance (NMR) and time-domain reflectometry (TDR). The dielectric permittivity of frozen soil decreased with a decrease in temperature, which was accompanied by a decrease in liquid (unfrozen) water content. Frozen soils with different total water content had the same amount of unfrozen water at below −1 °C; however, the permittivity of frozen soil depended on the total water content. A dielectric mixing model without considering reduced dielectric permittivity due to surface forces and ice formation could only describe the data for sandy soils. We expanded the mixing model by including reduced dielectric permittivity due to surface forces and ice formation. The estimations of liquid water content using the expanded mixing model were in agreement with the values measured by NMR at any soil type, total water content, ice content, and temperature.

1. Introduction

In soil, a certain amount of water remains unfrozen at subzero temperatures because of a decrease in the free energy of soil water due to surface forces of soil particles and the pore geometry among soil particles (Cannel and Gardner, 1959; Miller, 1980; Dash et al., 1995; Watanabe and Mizoguchi, 2002). During ground freezing, unfrozen water flows along a temperature gradient in partially frozen soil (Hoekstra, 1966; Fukuda et al., 1980), creating an uneven distribution of not only water but also solute and colloidal particles (Konrad and McCammon, 1990; Watanabe et al., 2001; Gay and Azouni, 2003). The change in unfrozen water content alters the hydraulic properties of frozen soil (Watanabe and Wake, 2008; Watanabe and Flury, 2008), which in turn affect soil water flow. The soil freezing characteristic (SFC), which is the relationship between temperature and unfrozen water content, is a fundamental property for simulating soil water dynamics (Spaans and Baker, 1996; Flerchinger et al., 2006). The SFC has been determined and examined mostly for saturated soils, but to estimate soil water storage and distribution in frozen ground, it is important to quickly and precisely measure the unfrozen water content of soils at various saturation levels.

The soil water content, θ, is commonly estimated based on the relative dielectric permittivity of soil, ε_r (e.g., Hoekstra and Delaney, 1974; Topp et al., 1980). Patterson and Smith (1981) assumed that liquid water content in frozen soil, θ_u, can be expressed from ε_r by using the same relationships as that used for soils at temperatures above freezing, because the measurement of ε_r for dry soil and pure ice were similar in magnitude. Smith and Tice (1988) showed a mismatch between Topp's equation and the $\theta_u-\varepsilon_r$ relationship in frozen soil, and they proposed a calibration equation based on nuclear magnetic resonance (NMR) measurements. Spaans and Baker (1995) suggested, based on experimental results using a gas dilatometer, that the calibration equation would change depending on the total water content. Many studies have attempted to clarify the effect of temperature changes and absorbed water content on ε_r (e.g., Herkelrath et al., 1991; Dirksen and Dasberg, 1993; Pepin et al., 1995; Warith and Or, 1999a,b). Birchak et al. (1974), for example, incorporated the low permittivity of absorbed water into the $\theta-\varepsilon_r$ relationship by using a dielectric mixing model. Yamanaka and Kaihotsu (2003) emphasized that the temperature dependence of ε_r could not be ignored, especially for frozen soil. Yoshikawa and Overduin (2005) reported that θ_u of unfrozen soil was often overestimated when determined by the $\theta-\varepsilon_r$ relationship due to the permittivity of ice in frozen soil. Bittelli et al. (2003, 2004) estimated the SFC and the ice content of frozen soils based on a dielectric mixing model including both ε_r and the permittivity of ice. Several other studies have taken a similar approach (e.g., Seyfried and Murdock, 1996; Stähli and Stadler, 1997; Flerchinger et al., 2006). In this study, we determined the SFC (θ_u-T relationships) of unsaturated soils using pulsed NMR. Then, to examine the applicability of the mixing model to frozen soils, we compared the

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SFCs to the $\varepsilon_r - T$ relationships measured by time-domain reflectometry (TDR).

2. Materials and methods

2.1. Sample and procedure

We examined Toyoura sand (Sand1), Tottori dune sand (Sand2), Fujinomori silt (Silt loam), and Mie Andisol (Loam) from Japan. Sand2 had a slightly larger mean diameter and a broader particle size distribution than Sand1 (Table 1). Silt loam is a mineral subsoil and known as a frost-susceptible soil that had more than 60% silt and about 24% clay. The Sand1 and Sand2 were washed in deionized water, and the Silt loam was passed through a 2-mm screen. These soils had relatively low electric conductivity (Table 1). Loam (Andisol), which had an aggregated structure and contained organic matters, was collected from a depth of 50 mm in fallow farmland. Two samples of Loam were prepared: one air-dried and filtered through a 2-mm screen, and the other crushed using a mortar and pestle.

Air-dried samples were mixed with distilled and deionized water in a plastic bag, equilibrated for 1 day, and then packed in a metal cell with an internal diameter of 17.3 mm and a height of 25 mm at a given bulk dry density. Table 1 lists the bulk dry density, initial water content, and soil particle density of the samples. The samples were frozen with liquid nitrogen immediately after the packing procedure to prevent water redistribution, placed in a freezer at −20 °C for 1 day, and then repacked under frozen conditions into a Teflon cell (sealed with a Teflon stopper to prevent water evaporation) for NMR measurements. The NMR cell was soaked in a coolant in a temperature-controlled bath together with an identical cell containing thermocouples to monitor temperature.

For TDR measurement, we conducted preliminary trials using several cells, and selected a cylindrical metal cell with an internal diameter of 45 mm and height of 120 mm in which the cell wall had no influence on TDR waveforms. Meanwhile, the cell containing a soil sample with 14 thermocouples was set under several temperature changes in the temperature-controlled bath, and selected an experimental temperature condition in which temperature profile in the soil sample became almost uniform within 1-hour and water movement during temperature change was negligible. Samples were mixed with water as same procedure as NMR samples and packed into the TDR cells with each 20-mm depth to make uniform initial water content and bulk density (same as the NMR experiment shown in Table 1), which was rechecked at the end of experiment. A three-pronged TDR probe (length × diameter × spacing = 74 × 1.5 × 5 mm) was vertically inserted into each sample. This TDR cell was equipped with thermocouples and sealed with a rubber stopper and soaked in the coolant in the constant-temperature bath.

The coolant was set to a specific temperature, and the samples were first maintained at that constant temperature for at least 2 h. The NMR cell was relatively small (diameter × height = 17.3 × 25 mm), so we assumed the sample was in thermal equilibrium and regarded as the same temperature as monitored temperature. Then, they were removed from the bath, and the amount of unfrozen water, θ_u, was measured by NMR, and the samples were back to the bath. Simultaneously, the relative permittivity of frozen soil, ε_r, was measured by TDR and the temperature in the TDR cell was measured by the thermocouples. The $\theta_u - T$ and $\varepsilon_r - T$ relationships of each sample were determined by repeating this procedure at temperatures from −20 °C to above 0 °C.

Table 1
Soil properties and experimental conditions.

	Sand1	Sand2	Silt loam	Loam (Andisol)
Mean particle diameter (mm)	0.21	0.35	0.014	–
Uniformity coefficient	1.44	1.70	200	–
Bulk density (Mg m^{-3})	1.43	1.46	1.13	1.045
Solid fraction (m^{3}m^{-3})	0.55	0.55	0.45	0.4
Total water content (m^{3}m^{-3})	0.17–0.37	0.16–0.26	0.2–0.49	0.26–0.51
Specific surface area[a] (m^2 kg^{-1})	1600	900	29900	26000
1:5 EC (mS m^{-1})	3.8	4.7	8.5	20.0
Ignition loss (kg kg^{-1})	1.34	0.50	6.85	17.83

[a] Specific surface area was measured by Brunauer, Emmett and Teller (BET) method (Soil Science Society of America, 2002).

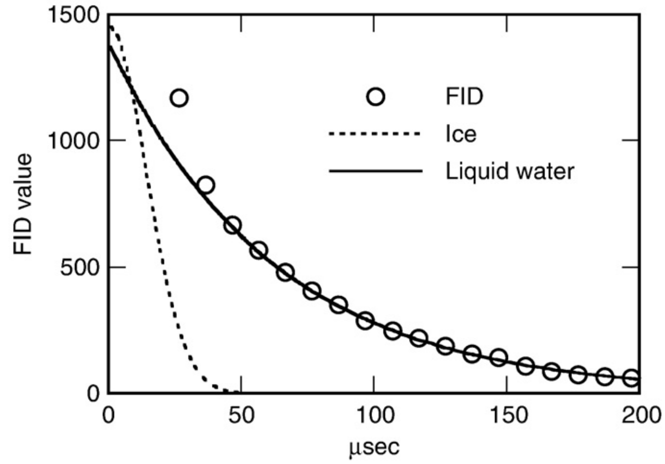

Fig. 1. Example of a FID curve. Open circles are measured FID values for Silt loam at −5 °C by pulsed NMR method. Dotted and solid lines are FID signals for ice and liquid water, respectively, separated by Eq. (1).

2.2. Pulsed NMR

NMR is a fast and accurate method for obtaining unfrozen water content in frozen soil in the laboratory (Smith and Tice, 1988; Ishizaki et al., 1996; Watanabe and Mizoguchi, 2002). NMR measures the free-induction decay (FID) of protons in a magnetic field. The FID value of water is proportional to the amount of water in the sample. Fig. 1 shows an example of a FID signal of frozen silt. As the FID signal of ice decreases more rapidly than that of liquid water, the amount of unfrozen water in a sample, θ_u, can be determined from its FID value, when the signals can be separated. We measured FID signals using a Maran Ultola (Resonance Ltd., USA) with a frequency of 40.3 MHz and fitted the following equation by the least-squares method to the data:

$$\mathrm{FID} = C_1 exp\left(\frac{-t}{C_2}\right) + C_3 exp\left(\frac{-t^2}{C_4^2}\right), \tag{1}$$

where t is the induction time, and C_1, C_2, C_3, and C_4 are fitting parameters. The first and second terms on the right side of Eq. (1) hypothetically represent the signals of water and ice, respectively (Farrar and Becker, 1971). Using the value obtained from the first term C_1 exp $(-t/C_2)$, the unfrozen water content was calculated following Ishizaki et al. (1996).

2.3. TDR method

The relative permittivity of soil, ε_r, can be calculated from the TDR waveform as follows (e.g., Topp et al., 1980):

$$\varepsilon_r = \left(\frac{ct_s}{2L}\right)^2 = \left(\frac{c\,|\mathrm{FP}-\mathrm{RP}|}{Lv}\right)^2, \tag{2}$$

where c and v are the propagation speeds of an electromagnetic wave in a vacuum and the porous media, respectively; L is the probe length; and $t_s = 2|\mathrm{FP} - \mathrm{RP}|$ is travel time of the wave deduced from the first