Resistivity and SIP response of rocks during freezing and thawing

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The use of electrical imaging methods for characterizing the frozen state of the subsurface requires the understanding of the low-frequency electrical soil/rock conduction and polarization properties as dependent on temperature in frozen and unfrozen regimes. While electrical resistivity tomography, measuring the quasi-static electrical conduction properties, is increasingly used in permafrost monitoring studies, relatively little attention has been paid so far to the measurement of electrical polarization properties for thermal state characterization. In particular, integrative models describing the low-frequency behaviour of both conduction and polarization properties below $T_0 = 0$ °C, where ice and water coexist in stable equilibrium, are still lacking.

We here present results from laboratory studies on sandstone and limestone samples with different pore size distributions, whose electrical conduction and polarization properties were measured in controlled freeze-thaw cycles (−30 to 10 °C) by means of impedance spectroscopy in the frequency range 1 mHz to 45 kHz. We used five different cylindrical samples, four sandstones and one limestone, of 10 cm length and 3 cm diameter. After saturating the samples with deionized water and waiting for chemical equilibrium, associated with some increase of pore water conductivity due to the dissolution of ions from the rock matrix, the samples were placed in a freezing chamber where they were cooled down from 10 to −20 °C by successively changing the temperature in steps between 0.2 and 1 °C; afterwards the samples were heated up again following the same procedure. After adjusting a new temperature, impedance measurements were only taken after thermal gradients in the sample had equilibrated, where waiting for two hours was found to be sufficient for the given size and thermal properties of the samples.

In Fig. 1, the spectral complex resistivity behaviour as a function of temperature for one of the sandstones, which was qualitatively observed for all sandstones, is shown. Resistivity magnitude varied only insignificantly with frequency below 1 kHz over the investigated temperature range, and thus the temperature dependence shown in Fig. 1a at 1 Hz is representative. The results reveal a distinct hysteretic behaviour, which is characterized by a critical transition of the sample at some temperature T_f during freezing, indicating abrupt ice crystallization in the pore space of the sample, and a gradually decreasing resistivity during thawing, suggesting continuous melting of the ice in the pores up to a temperature T_m. The observed resistivity hysteresis is qualitatively in agreement with the well-known hysteretic behaviour of other properties of porous media during freezing and thawing. Given the melting point depression of ice constrained in a pore according to the Gibbs-Thomson relationship, i.e., $T - T_m$ being inversely proportional to the pore size, the resistivity melting curve can be considered as reflecting the cumulative pore size distribution of the sample, similar to the concept of differential scanning calorimetry and NMR cryoporometry. Accordingly, the freezing curve clearly indicates that liquid water in the sample is supercooled down to the critical temperature T_f where sudden ice formation occurs.

The polarization properties likewise show a hysteretic behaviour as a function of temperature, which, however, is strongly frequency dependent. Inspecting the observed resistivity phase spectra, as exemplarily shown in Fig. 1b, one can identify the well-known relaxation of ice between 10^3 Hz and 10^5 Hz above −20 °C. The occurrence of this relaxation response during freezing and thawing is in correspondence with the above interpretation of the resistivity magnitude behaviour in terms of thermal state of the sample. In addition, polarization is observed in the lower frequency range, which in the unfrozen regime exhibits relatively weak frequency dependence between 1 Hz and 1 kHz. We attribute the response below 100 Hz to electrochemical polarization associated with electrical double layers (EDL) at the water-mineral interfaces. The observed phase spectra suggest that upon ice crystallization this polarization mechanism partly breaks down, in particular at lowest frequencies representative of largest pores. This could be explained by ice formation in corresponding pores hindering the build-up of membrane polarization. However, water in smaller pores and films of premelted water around grains as well as ice nuclei could still maintain electrochemical polarization (in particular Stern layer polarization), potentially explaining the residual phase response with ongoing cooling.

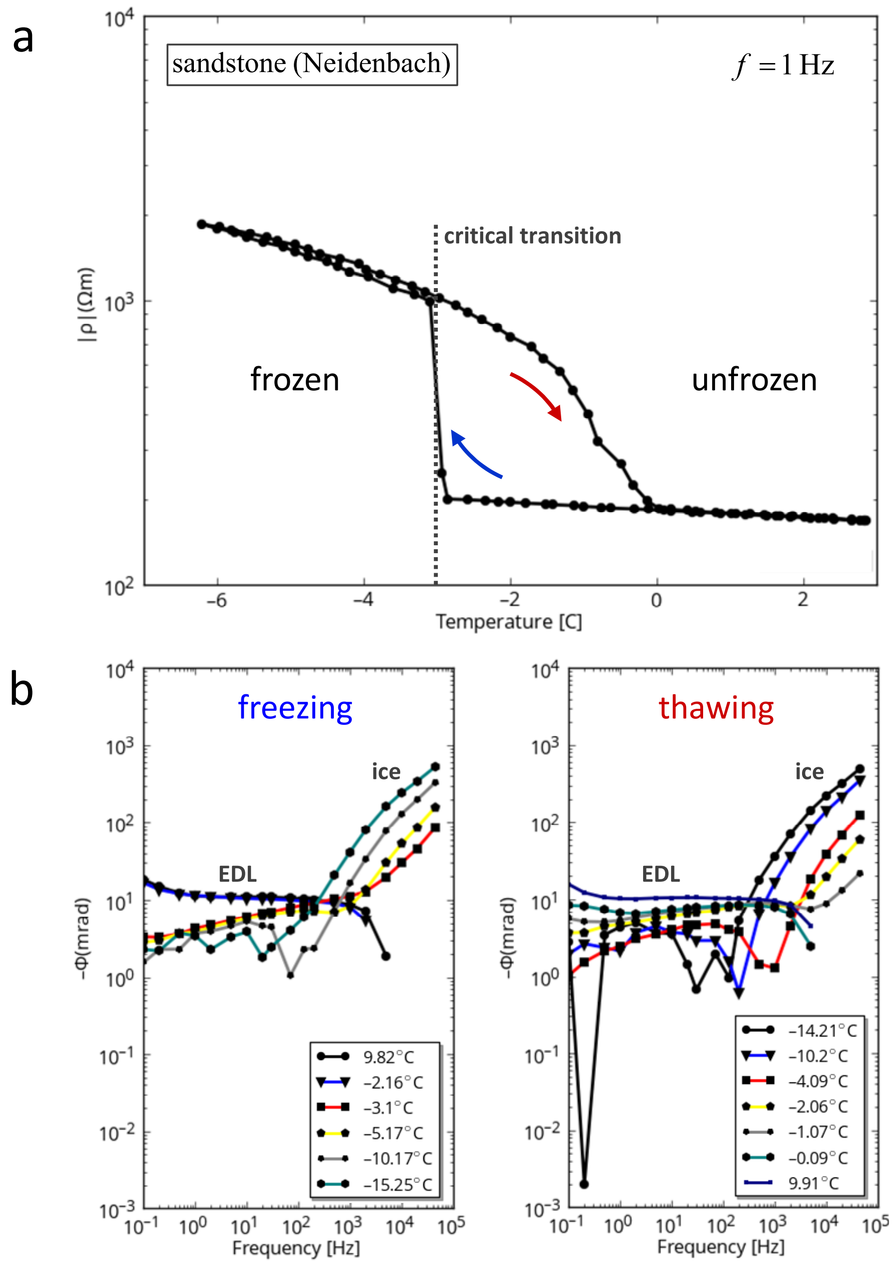

Fig. 1: Complex resistivity response measured on a sandstone sample during freezing and thawing. At each temperature step, measurements were taken after thermal equilibrium had re-established. a) Resistivity magnitude at 1 Hz as a function of temperature during freezing (bottom curve in hysteretic region) and thawing (top curve in hysteretic region). b) Resistivity phase spectra at selected temperatures during freezing (left) and thawing (right).

We conclude that low-frequency electrical properties measured in SIP offer direct access to the presence and amount of ice as well as liquid water (films) in frozen rocks. The hysteretic behaviour must be taken into account when electrical properties are, for example, to be used for monitoring seasonal temperature changes in permafrost environments. Studying the relationship between the critical temperature at which nucleation is triggered and the pore size distribution might provide improved insight into the mechanisms governing ice formation in porous rocks.