

## Velocity and Attenuation of Compressional Waves in Brines

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### Summary

We conducted an acoustic pulse transmission experiment in brine as its temperature decreased from 25 °C to – 21°C. The purpose was to understand how the transmitted wave changes with the onset of freezing. The main practical reason for this experiment was to use partially frozen brine as an analogue for a mixture of methane hydrate and water present in the pore space of a gas hydrate reservoir. The results indicate that as the brine cools, the P-wave velocity decreases from 1500 m/s to 1480 m/s. With the onset of freezing, the velocity begins to increase until it reaches a maximum of 3700 m/s. Counter to our intuition, this increase in the velocity is accompanied by a strong reduction of the signal's amplitude and frequency content. The signal quality does not recover even after a brine sample was kept frozen at – 21°C for over two months. This result may partly explain the reported effect that the attenuation in methane hydrate sediment is relatively large although the hydrate in the pore space acts to dramatically increase the velocity.

### Introduction

Brine reacts to freezing very differently than pure water. While the latter becomes solid ice as the temperature falls below freezing, the former remains slush even at very low temperatures. The reason for this behavior is that salty water does not freeze and the ice generated from brine is always fresh. As larger and larger portions of brine turn into fresh ice, the remaining liquid pockets become more and more concentrated which further inhibits the freezing.

We have decided to investigate the effect of this behavior on the acoustic properties. One reason was scientific curiosity – to the best of our knowledge, such experimental data are nonexistent.

The other reason was practical. Unexpectedly large attenuation in sediments with gas hydrates has been recently observed at different geographical locations, in different depositional environments, and at different frequency (Guerin et al., 1999; Sakai, 1999; Wood et al., 2000; Guerin and Goldberg, 2002; Pratt et al., 2003).

Experimenting with methane hydrate is expensive and difficult. This is why we decided to use frozen brine where ice and salty water coexist at the pore-scale level as an elastic analogy to methane hydrate and seawater present in the pore space of sediment. These experiments are not intended to entirely explain the mechanism of elastic-wave attenuation in methane hydrate reservoir but rather partially

unveil it.

### Experimental Setup

The experimental setup consisted of a Plexiglas container that held the brine. Immersion transducers (1 MHz Panametrics Transducers V303-SU) were positioned on two sides of the container such that their distance was fixed during the experiment (Figures 1 and 2).

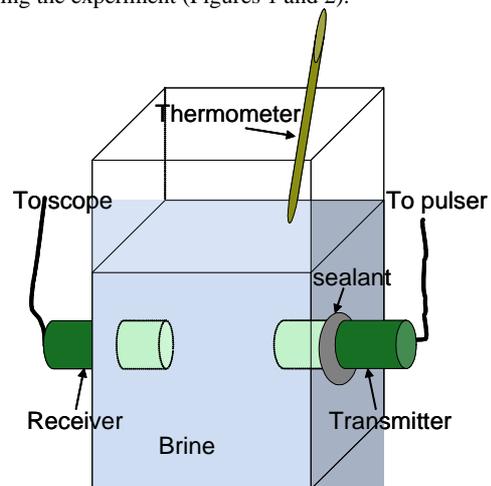


Figure 1. Experimental setup (a scheme) used in the study. The transducers are immersed in the brine at all times and freeze together with it.

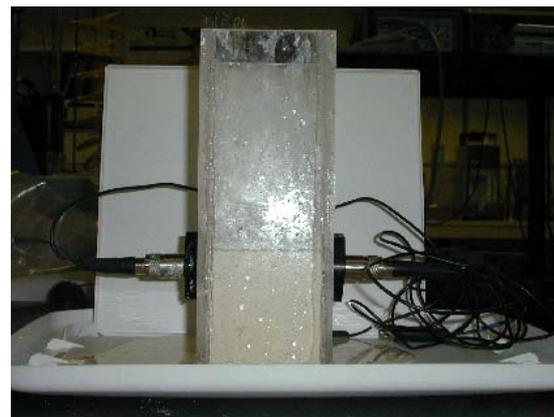


Figure 2. Experimental setup (photograph)

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The transducers were excited using a Panametrics pulser. The signals through the brine were received and digitized by a Tektronix oscilloscope.

The brine was a solution of NaCl in distilled water with concentration 50,000 parts per million. It was de-aired in vacuum.

The whole setup was placed in the freezer. With the onset of freezing, the slush was mechanically stirred to ensure homogeneous distribution of ice crystals in the brine.

### Results

Figure 3 shows the waveforms as the brine cools. Above freezing, the waveforms do not noticeably react to temperature variations, except for a small delay in the arrival time and reduction in the peak-to-peak amplitude as the brine becomes cooler.

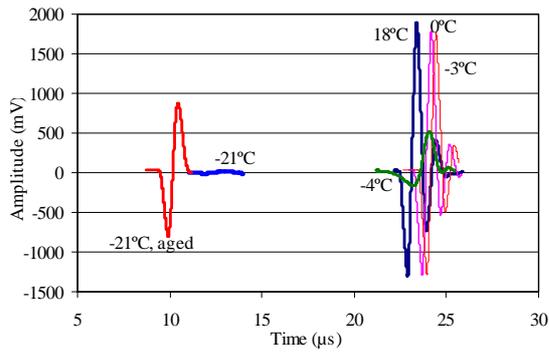


Figure 3. Waveforms registered in the cooling and freezing brine with temperature marked. There is a significant reduction in signal amplitude and frequency as the brine freezes. At  $-21^{\circ}\text{C}$  the amplitude is reduced by almost a factor of 100 (bold blue waveform on the left side of the graph), while the velocity is almost doubled. After two months in the freezer, the velocity increases slightly (red waveform on the left side of the graph). However, the amplitude does not fully recover.

However, there is a significant reduction in signal amplitude and frequency as the brine freezes. At  $-21^{\circ}\text{C}$  the signal amplitude is reduced by about two orders of magnitude, while the velocity is almost doubled.

After two months in the freezer, as the frozen brine matures, the velocity increases slightly. At the same time, the amplitude recovers but only to half of its original value at  $18^{\circ}\text{C}$  (Figure 3 and Table 1).

Figure 4 shows normalized amplitude frequency spectra of the waveforms versus temperature. The spectra were calculated from the first cycle of the corresponding

waveforms.

Above freezing, the frequency content of the waveforms does not change. However, there is a significant reduction in signal frequency as the brine freezes. At  $-21^{\circ}\text{C}$  the principal frequency of the signal is reduced to almost one-third of its original value (from about 740 kHz to 200 kHz). After two months at  $-21^{\circ}\text{C}$ , the signal recovers almost 85% of its original frequency content at  $18^{\circ}\text{C}$ .

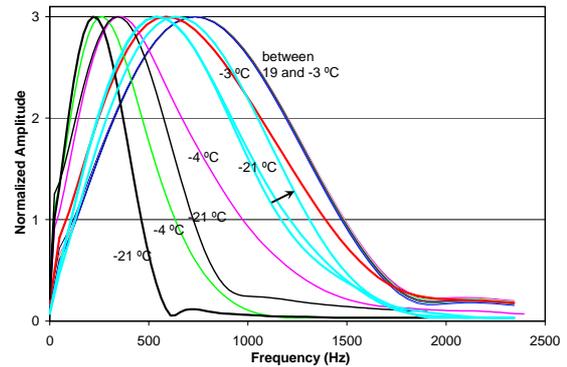


Figure 4. Normalized waveform spectra. The thick black and cyan colored lines show spectra at  $-21^{\circ}\text{C}$ . The black colored spectra was measured in the frozen brine after only a few hours. The cyan colored spectra show changes during the two months that the setup remained at  $-21^{\circ}\text{C}$ . The small arrow marks increasing time.

Table 1: Velocity, frequency, and amplitude versus temperature.

Temperature ( $^{\circ}\text{C}$ )	Velocity (m/s)	Principal Frequency (kHz)	Peak-Peak amplitude (mV)
18	1499	732	3188
9	1491	732	3139
5	1482	732	3143
3	1463	732	3104
0	1454	732	3090
0	1477	732	3082
-3	1431	732	3059
-3	1435	732	3064
-3	1624	586	1770
-4	1655	341	683
-4	1738	268	389
-20	3082	219	158
-21	3236	342	33
-21 aged	3686	537	1214
-21 aged	3679	537	1197
-21 aged	3679	586	1383
-21 aged	3709	634	1670

In Figure 5, we summarize the observed changes in the acoustic properties of the brine as a function of temperature.

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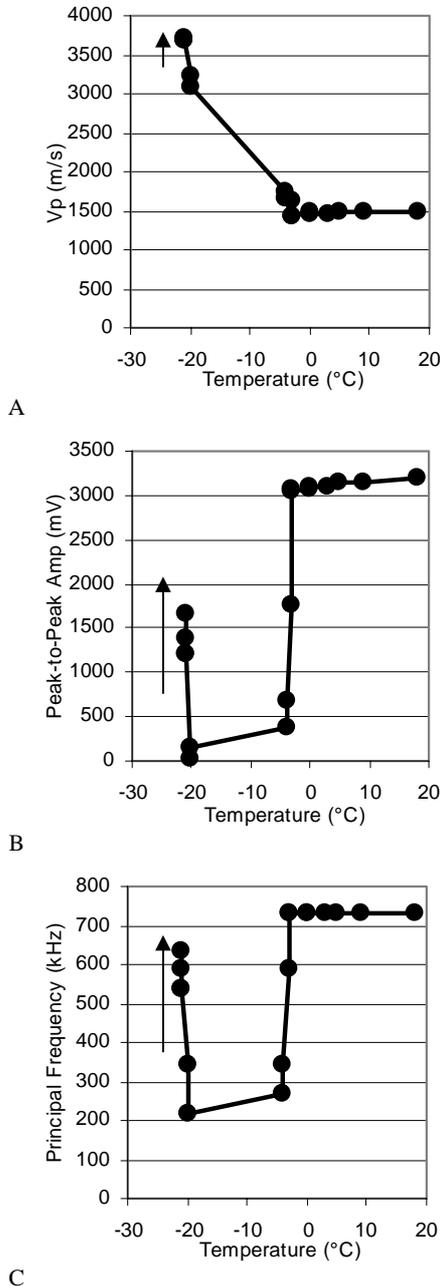


Figure 5. Velocity (A), peak-to-peak amplitude (B), and frequency (C) in the brine as a function of temperature. The arrow marks changes observed with increasing time as the brine matures in the freezer at a constant temperature.

Finally, in Figure 6, we show how the velocity and frequency vary with time at  $-21^{\circ}\text{C}$  as the frozen sample matures. The increase in the velocity is not large as compared to the velocity increase that occurred during freezing. However, the frequency recovers and so does the amplitude (Table 1), although not quite to their original values in liquid brine at room conditions.

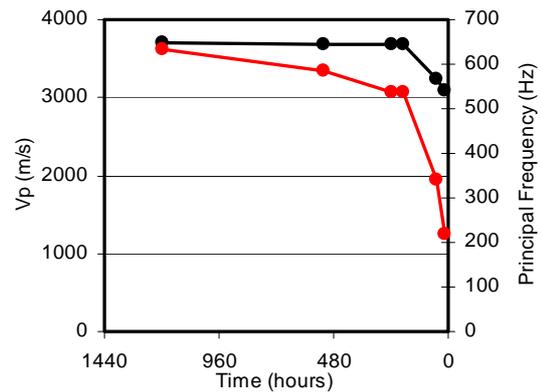


Figure 6. Velocity (black, left axis) and principal frequency (red, right axis) versus elapsed time as the frozen brine matures for about two months at  $-21^{\circ}\text{C}$ .

## Conclusions

The main result from our experiment that as the brine freezes the velocity increases and so does the attenuation is somewhat unexpected. The intuition tells us that the faster the material the smaller the attenuation. However, the results presented here counter this common assumption.

At this point we avoid any speculations about the nature of the observed phenomenon – several reasons for it may exist, including artifacts of ultrasonic pulse transmission – or any generalization of these results. Also, we do not resort to complicated theory that would explain these results.

In order to confidently generalize and mathematically describe the phenomenon, more experiments have to be and will be conducted and their practical and fundamental significance established.

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