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— Report —

Experiments on interstitial water squeezing at high pressure using water-gathering plates

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A sediment squeezing tool for use in interstitial water extraction at the onboard laboratory of D/V *Chikyu* was modified for use at pressures up to 112.3 MPa. A pair of titanium water-gathering plate originally developed by Central Research Institute of Electric Power Industry (CRIEPI) replaces a thin mesh to allow squeezing of sediment at a higher pressure without jamming of sediments. The modified tool endured a longer period of squeezing at high pressure, and yielded greater volume of interstitial water in test experiments. The acquired fluid sample composition showed no major changes even at the pressure of 112.3 MPa, although further tests must be conducted to assess the potential dehydration of clay mineral at high pressures. However, squeezing of sandstone of approximately 12% porosity yielded no interstitial water at all, showing system limitations for low-porosity samples. The improved tool and the data from these experiments are expected to be useful when low-porosity sediments from a deep hole are obtained.

Keywords : interstitial water, squeezing, core sample, drilling, pressure

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1. Introduction

Analyses of interstitial water (IW) from core samples provide insights into chemical processes and migration of sub-seafloor fluid. Such analyses have been an important aspect of onboard analysis in scientific ocean drilling. Water is considered as playing principal roles in deep sub-seafloor environments, as a trigger of fault activity, an essential element of life, a lubricant of mantle movement, and so on. The analysis of IW has therefore become much more important for exploration to great depths. Onboard D/V *Chikyu*, IW has been collected from core samples using conventional squeezing with a hydraulic sediment press (Manheim, 1966) and has been collected in plastic syringes (Manheim and Sayles, 1974).

In most cases in previous expeditions, pressure up to 46.8 MPa produced sufficient volume of IW sample from a shallow sediment core from a borehole up to 1000 meters below seafloor (mbsf). The upper limit of the squeezing load of our standard protocol was determined mainly to avoid clay mineral dehydration, which occurs at > 80 MPa (Rieke and Chilingarian, 1974), with additional constraints imposed by technical considerations. However, the extraction of IW from core samples tends to be difficult with depth because porosity and water contents decrease in lithified sediments. Higher pressure often causes sediments to break through the paper filter and titanium mesh and jam in the fluid sample outlet. In Integrated Ocean Drilling Program (IODP) Exp 337, no IW was obtained by squeezing core samples below 2000 mbsf. At that depth the core sample porosity was about 30% (Exp. 337 scientists, 2013), which underscores the limitations of the original squeezing method used in the *Chikyu* laboratory.

However, some groups have reported IW sample recovery from low-porosity samples (see review of Sacchi et al., 2001). A research group of the Central Research Institute of Electric Power Industry (CRIEPI) developed a sediment squeezing system that can produce 512 MPa maximum pressure. They obtained an IW sample from shale with porosity of 10–20% (Oyama and Suzuki, 2006).

Although the 512 MPa maximum pressure is not achievable in the *Chikyu* laboratory without renewing the whole system, we modified some squeezing tools to enable squeezing at a higher pressure than under our standard protocol. This report describes the squeezer modification for high pressure squeezing and the preliminary results of test experiment.

2. Method

2.1 Modification of squeezing tools

Figure 1 and 2 show the IW squeezing system and schematic illustrations of the original and modified squeezing tools, respectively. The IW squeezing system in *Chikyu* laboratory is composed of automated press machine CARVER 3894 and a squeezer jacket containing sediment sample. In the original tool, sediment samples are contained in a titanium jacket, capped by the top piston which conveys the top load to the sediment. The sample is pressed to the bottom, which has a small outlet of fluid sample, through a paper filter (thickness: 0.18 mm, particle retention: 3 μm) and titanium mesh (40 mesh, wire diameter: 0.2 mm).

In the modified tool, the titanium mesh was replaced by a water-gathering plate (Fig. 3) and an additional sample collection port was made on the upper piston. The collection port of the upper piston is connected to a syringe by a ϕ 1/16 inch plastic tube.

The water-gathering plates were made from 3-mm-thick pure titanium (JIS Classes 4 and 2), as is the jacket, having radial ditches with small (ϕ 1 mm) holes (Fig. 3). The original tool has a hole of ϕ 3.6 mm at the center of the bottom for sample collection. In previous tests, the hole tended to be jammed at high pressures. The water-gathering plate, which is stronger than a thin mesh, reduces the risk

Fig. 1. Interstitial water squeezing system in *Chikyu* laboratory. The system is composed of a squeezer jacket in an automatic press machine.

of jamming caused by broken mesh, and allows squeezing at a higher pressure. The collection port of the upper piston presumably enhances the ability to collect IW from samples having low fluid mobility.

2.2 Test experiments

Test series 1: Squeezing of the low-porosity sandstone (cylinder)

Sample: Sandstone from India with porosity of 12.6–12.8% was used in this study. The samples were shaped into cylinder with diameter of 55 mm and length of 100 mm. They were soaked in seawater for four days in vacuumed desiccator before being wrapped with wet paper towels and storage in an aluminum bag. We prepared two representative samples for runs A and B, and their initial weight were 258.6 and 257.9 g, respectively.

A) Standard protocol up to 25,000 lbs (corresponding to 46.8 MPa) in the original squeezer

B) Higher pressure up to 60,000 lbs (112.3 MPa) with the modified squeezer

Test series 2: Squeezing of the low-porosity crushed sandstone

Sample: Samples were prepared in the same way as described for Test series 1. However, to increase the surface

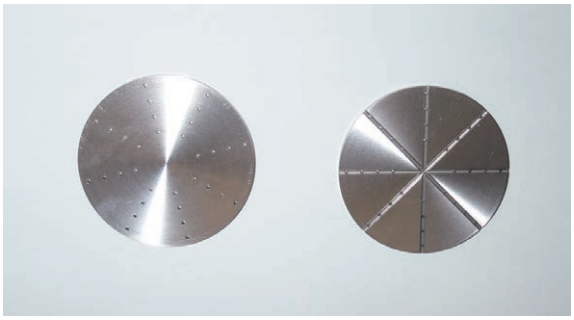

Fig. 3. Water-gathering plate (left, obverse; right, reverse).

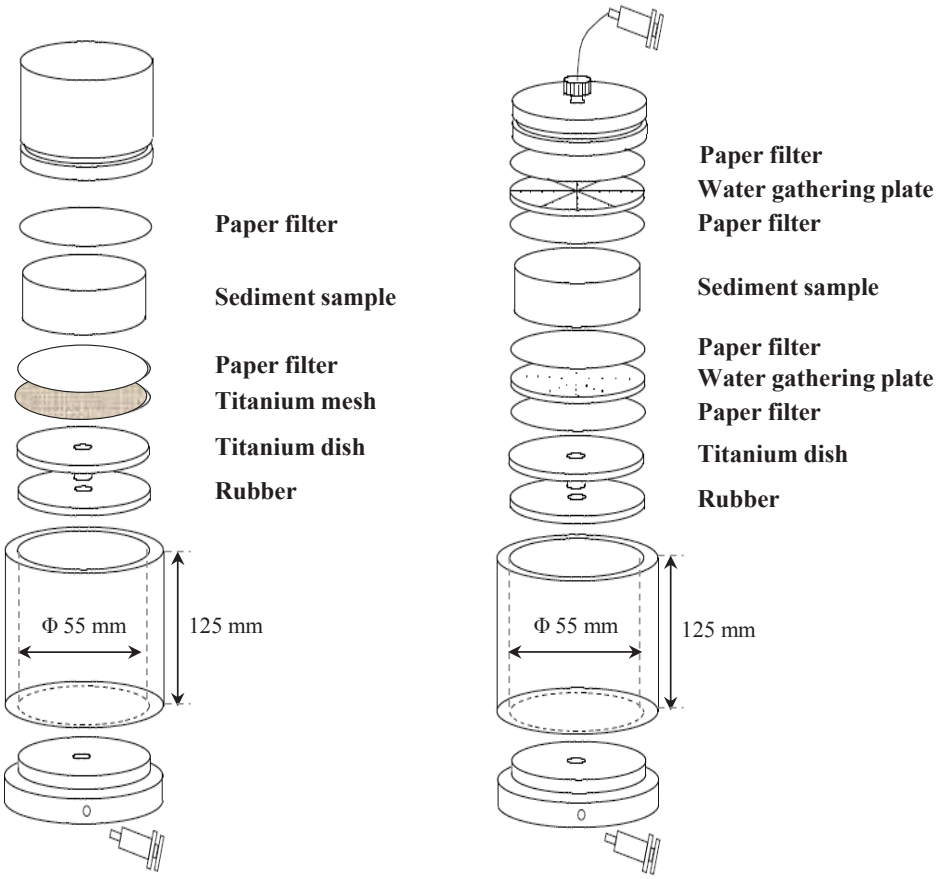

Fig. 2. Current (left) and modified (right) squeezing tools. The modified tool incorporates a water-gathering plate at the top and bottom of the sample, and a sample collection port through the upper piston, which is inserted into a squeezer jacket.

area, samples were crushed to small pieces with average size of 20 mm just before squeezing.
C) Higher pressure with the modified tool

Test series 3: Squeezing of the cutting sample from Exp. 337

Sample: Cutting sample 116SMW (mud stone) from approximately 1300 mbsf of Hole C0020A of Exp 337 was used. The samples had been washed in seawater during the expedition, and had been stored in the laboratory refrigerator for about a year. For this experiment, the samples were again washed and sieved in seawater. The 1–4 mm size fraction was selected before being soaked in seawater for 17 h. The samples were wiped on a paper towel until no moisture remained on the surface. Then the wet mass was measured before squeezing. The sample porosity was 48.4%. Water contents were 39.3 wt%. Samples were divided to three subsamples for runs D–F. The respective wet masses were 163.7, 162.1, and 155.0 g for Runs D, E, and F.
D) Standard protocol
E) Higher pressure with the modified tool
F) Higher pressure with the modified tool for a longer period

Runs D) and E) were conducted at the same time. Run F) was conducted the next day. The samples were finally collected at the end of standard protocol after waiting an additional 30 min. In run F, samples were also collected at 24 and 48 h. Each sample was weighed before chemical analysis.

The target force and pressure in the standard protocol are shown in Table 1. In the high-pressure runs, the target force was increased to 60,000 lbs (112.3 MPa) after the end of step 6.

2.3 Chemical analysis

The obtained IW sample in test series 3 was subjected to a series of measurements of refractive index, anion concentrations (Br^-, SO_4^{2-}), cation concentrations (Na^+, K^+, Ca^{2+}, Mg^{2+}), and chlorinity after filtration by membrane filter (0.45 μm pore size). Paper filter and titanium mesh were cleaned by ultrapure water and detergent soaking, respectively. Plastic syringes and sample bottles for sampling were washed with acid bath (HCl and HNO_3).

Table 1. Target force and the duration maintained at the force of the standard protocol.

Step No.	Target force (lb)	Target force (kg)	Pressure (MPa)	Time (min)
1	15,000	6804	28.1	5
2	17,000	7711	31.8	7
3	20,000	9072	37.4	10
4	21,500	9525	39.3	10
5	23,000	10,433	43.0	10
6	25,000	11,340	46.8	10

Refractive index

Refractive index was measured by digital refractometer (RX-5000α, ATAGO).

Anion (Br^- and SO_4^{2-}) concentrations

$Br^-$and SO_4^{2-} ion concentrations were measured by ion chromatography using an ion chromatograph (ICS-1500, DIONEX) and auto-sampler (AS-50, DIONEX), IONPAC AG12A for guard column, IOCPAC AS12A for analytical column and ASRS-300 for anion suppressor. The degassed 2.7 mM Na_2CO_3 / 0.3 mM $NaHCO_3$ solution was used for eluent. All sample were diluted into 1: 100 before measurement.

Cation (Na^+, K^+, Ca^{2+} and Mg^{2+}) concentrations

Na^+, K^+, Ca^{2+} and Mg^{2+} ion concentrations was measured by ion chromatography using an ion chromatograph (ICS-1500, DIONEX), auto-sampler (AS-50, DIONEX), IONPAC CG12A for guard column, IOCPAC CS12A for analytical column and CSRS-300 for cation suppressor. The degassed 20 mM methanesulfonic acid solution was used for eluent. All sample were diluted into 1: 200 before measurement.

Chlorinity

Chlorinity was measured by potentiometric titration using a titrator (794 Basic Titrino, Metrohm). IAPSO standard seawater (P-series) is used for standardization of the 0.01 M $AgNO_3$ titrant. Diluted 100 μL of IW sample with 15 mL of 0.2 M $NaNO_3$ was measured.

3. Results

3.1 Test Series 1 & 2

No water sample was obtained by squeezing for up to 48 h, irrespective of the pressures and sample shapes. The results suggest that pressure higher than 112.3 MPa was necessary to obtain IW by squeezing samples of approximately 12% porosity.

3.2 Test Series 3

The IW yields of Runs D, E, and F are presented in Table 2. Results show that the higher pressure yielded about 40% more volume of IW samples (Runs D and E). Run F showed that the sample yield continued after 24 h.

Although reproducibility was not tested, the results suggest that squeezing at a higher pressure yielded increased volume of the fluid sample. However, the modified tool might be less effective to collect IW sample than the original tools when used at the same pressure up to 25,000 lbs, possibly because the modified tool has more dead space through the sample passage.

The results of the chemical analyses of the obtained IW samples are shown in Table 3. No systematic change with the increased pressure or time was observed, suggesting that the new protocols had no major effects on the IW composition. Nevertheless, it is noteworthy that the seawater chemistry changed during soaking of the cutting samples. The increases in K^+ and Ca^{2+} after soaking suggest that drilling mud remained with the cutting samples. It dissolved during the soaking, thereby hindering observations of other effects such as dehydration of clay minerals at high pressure. The decreases in the refractive index and chlorinity at 24 h and 48 h might indicate the possible occurrence of dehydrated water from clay minerals. However, the data are insufficient to attempt any further interpretation.

Table 2. IW volume collected in test series 3. The target force of 25,000 lbs corresponds to 46.8 MPa and 60,000 lbs to 112.3 MPa, respectively.

Squeezing pressure and time	IW yield (g)		
	D)	E)	F)
Standard protocol (52 mins)	10.48	9.82	8.97
25,000 lbs (+30 mins)	1.01	–	–
60,000 lbs (+30 mins)	–	6.34	5.27
60,000 lbs (+2 h)	–	–	2.31
60,000 lbs (+12 h)	–	–	2.19
60,000 lbs (+24 h)			0.90
60,000 lbs (+48 h)	–	–	0.71
Total yield	11.49	16.16	20.34
Sample mass (g)	163.67	162.14	154.96
Yield/100 g cuttings	7.02	9.97	13.13

Table 3. Results of chemical analysis of the IW samples: RI, refractive index; Chl, chlorinity; B.D., below detection limit. Units are mM for the concentrations except for unitless RI. Cations of F (60,000 lbs, 48 h) were not measured due to short of obtained sample.

	RI	Chl	Br^-	SO_4^{2-}	Na^+	K^+	Ca^{2+}	Mg^{2+}
D (<25,000 lbs)	1.33947	524	0.70	10.1	394	58.3	51.2	B.D.
D (at 25,000 lbs)	1.33962	516	0.69	8.7	384	56.3	51.5	B.D.
E (<25,000 lbs)	1.33948	516	0.69	9.8	393	58.6	51.4	B.D.
E (60,000 lbs)	1.33979	528	0.74	9.6	367	52.9	53.8	B.D.
F (<25,000 lbs, 52 mins)	1.33938	519	0.68	10.7	395	49.2	55.9	B.D.
F (60,000 lbs, 15 mins)	1.33968	533	0.73	11.4	406	49.5	62.7	B.D.
F (60,000 lbs, 30 mins)	1.33976	540	0.71	11.8	401	48.0	63.2	B.D.
F (60,000 lbs, 24 h)	1.33971	529	0.71	10.7	406	46.0	62.0	B.D.
F (60,000 lbs, 48 h)	1.33957	507	0.69	9.8	–	–	–	–
Seawater	1.33921	537	0.82	27.9	461	10.0	10.3	52
Seawater after soak	1.33893	513	0.72	14.2	408	50.3	47.4	B.D.

4. Conclusion

A modified tool with water-gathering plates enabled squeezing of sediment samples at pressure up to 112.3 MPa. The tool produced more volume of IW samples than the original method did. When testing a higher pressure, longer squeezing than the standard protocol is recommended. The chemistry of the produced IW showed no significant change with pressure or time, although further tests must be conducted to assess the potential dehydration of clay mineral at high pressures. However, it is noteworthy that the contaminants were dominant in the tested sample and that the results might not reflect the IW chemistry.

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