EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal:
https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Deadlines for submission of Experimental Reports

Experimental reports must be submitted within the period of 3 months after the end of the experiment.

Experiment Report supporting a new proposal (“relevant report”)

If you are submitting a proposal for a new project, or to continue a project for which you have previously been allocated beam time, you must submit a report on each of your previous measurement(s):

- even on those carried out close to the proposal submission deadline (it can be a *“preliminary report”),*
- even for experiments whose scientific area is different form the scientific area of the new proposal,
- carried out on CRG beamlines.

You must then register the report(s) as “relevant report(s)” in the new application form for beam time.

Deadlines for submitting a report supporting a new proposal

- 1st March Proposal Round - **5th March**
- 10th September Proposal Round - **13th September**

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report in English.
- include the experiment number to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** **Acoustic velocites of $(Mg_xFe_{1-x})_2SiO_4$ ringwoodite**	**Experiment number**: ES-1135
Beamline: ID06-LVP	**Date of experiment**: from: 28/06/2022 to: 04/07/2022	**Date of report**: 12/09/2022
Shifts: 18	**Local contact(s)**: Dr A. Rosenthal	*Received at ESRF:*

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Report:

Observations of seismic waves travelling throughout Earth's interior provide one of the few direct, and arguably the strongest, constraints on the current state of the mantle (e.g. Dziewonski & Anderson, 1981). Careful comparison with experimental and computational measurements of the elastic properties of minerals allows this data to be interpreted in terms of mantle mineralogy, chemistry and temperature; information that is not otherwise directly attainable. However, accurate interpretation of seimological data requires high-quality measurements of elastic properties for appropriate minerals, otherwise the information maybe lost or incorrectly deciphered.

The trasition zone (TZ) is a key region of the Earth located between the upper and lower mantle, and exerts important controls on global recycling, heat flow, mantle convection and mixing. However, it remains unclear whether it possesses a pyrolytic composition whilst its water content is also controversial (e.g. Fei et al., 2017; Irifune et al., 2008; Mao et al., 2012; Schulze et al., 2018). The elastic properties of wadsleyite and ringwoodite (Rw), which account for > 50 wt.% of the TZ, are only sparsely measured at appropriate mantle *PT* conditions. There has been only one single experiment that reports the acoustic properties of Fo90 Rw up to 19 GPa and 1400 °C and all other studies are limited to temperatures < 1000 °C and pressures < 15 GPa, outside TZ conditions (17.9-23.8 GPa, 1770-1873 K) and the Rw stability field, not to mention the effect of Fe content on the elastic properties of Rw. The lack of accurate knowledge of these properties is one of the major reasons for our limited knowledge of TZ composition.

ES-1135 was performed in June-July, 2022. The polycrystalline Rw samples with different Fe concentrations (X_{Fe}=0, 0.1, 0.2, 0.3, 0.5, 1) were successfully pre-synthesized at UCL using multi-anvil press at 19 GPa and 1473 K and double polished as the starting materials for the ultrasonic measurements at ESRF. During this beam time, eight multi-anvil runs were conducted using the 10/4 assembly with a newly developed X-ray transparent TiC-MgO composite heater. The composition of samples and the maximum *PT* conditions for each run are listed in Table 1. In each run, we first compressed the sample to the targeted pressure (~13-22 GPa) depending on its composition, and then heated the sample to 800-1000 °C to release the stress in the sample during compression. After that, the temperature was decreased and increased in cycles and X-ray diffraction (XRD), imaging and MHz frequency ultrasonic measurements were made during the heating loops to determine the structure, length and acoustic velocities. The temperature was monitored by the thermocouple and pressure was determined using the peaks of NaCl by real-time XRD to make our best not to go out of the Rw stability field. The new diffraction free gaskets were used through the X-ray windows in all experiments.

In all runs, the TiC heater worked well. In run-1 to run-4, graphite lids were used at the end of the heater which caused instability of the heater above 1200 °C, maybe due to the tranformation of graphite to diamond, therefore, we only used TiC in run-5 to run-8, which proved to be a more stable heater to more than 1400 °C. In all experiments the heater underwent reactions to first form, and subsequently dehydrate, brucite (identified using XRD) at 550 and ~ 900 °C which caused temporal instabilities and in one instance led to a blowout. This was presumed to originate from atmospheric water sorbed to the fine-grained heater in a period of high humidity and not experienced during prior testing at UCL. The thermocouple contacted with the heater in some experiments and could not read correct temperatures (run-1, run-2, run-4) influenced by the direct current through the heater but we managed to avoid such contacts in run-5 to run-8. The ultrasonic signal was not good at the beginning in run-1 to run-4 due to some connection problems (impendance contrasts in wiring and final cable mounting on the press being deformed at high pressure conditions) and quality of the connection cables between the transducer and oscilloscope. However, after several trials, good ultrasonic signals were obtained in later runs (run-5 to run-8, Figure 1 shows an example of our ultrasonic data). At the same time, the sample was observed to be in the middle of the X-ray window after compression, which enables the measurement of sample length through X-ray imaging. Although we did not have good ultrasonic signals in run-1 to run-4, we collected the XRD pattern through several heating loops and therefore by refinement and fitting, we can get high pressure high temperature equation of state parameters of the corresponding phases. In run-5 and run-6, we made ultrasonic measurements at 22.4 GPa and 18.7 GPa respectively up to 1400 °C. In run-7, we made ultrasonic measurements at 20 GPa up to ~ 1000 °C.

In some experiments, surrounding mica peaks came in at certain high tempratures which was misidentified as wadsleyite due to their simlar peak positions at low 2-theta angles, therefore we tried to decrease the pressure during heating to make it fully transform into wadsleyite, however, blowout happened during such hot decompression at above 900 °C (run-5 and run-7). Another blowout happened in run-8 due to the failure of the diffraction free gaskets in the X-ray window.

Table 1: summary of experimental runs performed in ES-1135.

Experiments	Starting material	Max *PT* conditions	Notes
Run_1	Fe_2SiO_4 Rw	13.2 GPa, 280 W (> 923 °C)	-TC not work -No ultrasonics signal -three heating cycles, XRD recorded -collected diffraction to 1 bar
Run_2	$(Mg_{0.8}Fe_{0.2})_2SiO_4$ Rw	17.6 GPa, 400 W	-TC not work -No ultrasonics signal -one heating cycle, XRD recorded -new low angle peak appears at 244 W -beam off during decompression
Run_3	$(Mg_{0.9}Fe_{0.1})_2SiO_4$ Rw	21.2 GPa, 450 W (~1205 °C)	-TC works well

			-No ultrasonics signal - two heating cycles, XRD recorded -heater not stable in the 2nd heating cycle at 450 W, quench -collected diffraction to 1 bar
Run_4	$(Mg_{0.9}Fe_{0.1})_2SiO_4$ Rw	22.4 GPa, 366 W	-TC not work -No ultrasonics signal -two heating cycles, XRD recorded -massive blow out at 366 W
Run_5	Mg_2SiO_4 Rw	22.4 GPa, 550 W (~1429 °C)	-Max temperature is high enough -Good ultrasonic signal -TC works well -two heating cycles, XRD recorded -Blowout during decompression at high tempreature
Run_6	$(Mg_{0.7}Fe_{0.3})_2SiO_4$ Rw	18.7 GPa, 600 W (~1400 °C)	-Good ultrasonic signal -TC works well -three heating cycles, XRD recorded -collected diffraction to 1 bar
Run_7	$(Mg_{0.8}Fe_{0.2})_2SiO_4$ Rw	20 GPa, 444 W (~1100 °C)	-Good ultrasonic signal -TC works well -1.5 heating cycles, XRD recorded - Blowout during decompression at high tempreature
Run_8	$(Mg_{0.5}Fe_{0.5})_2SiO_4$ Rw	16.97 GPa, 343 W (~962 °C)	-TC works well -Blow out in the 1st heating cycle at 962° due to the failure of diffraction free gasket

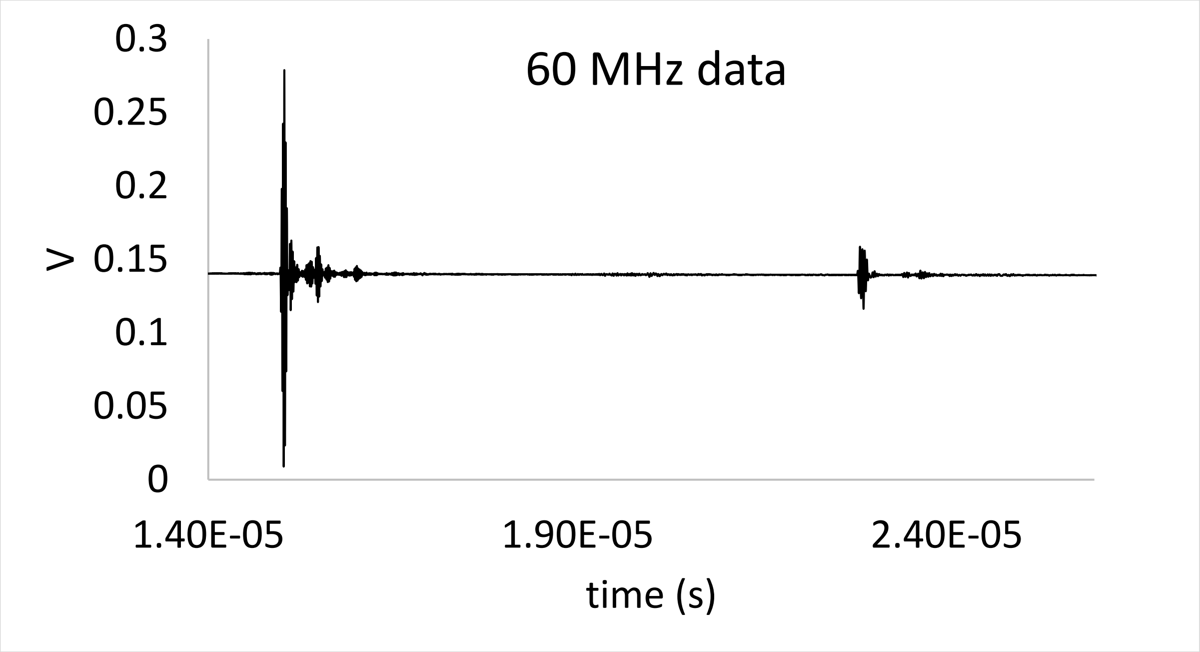

Figure 1: exemplar ultrasonic data signal from run_5.

Experiments all produced large volumes of XRD and ultrasonic data (typically ~ 8000 diffraction patterns per experiment). In order to deal with this data quantity, new analyses procedures have been under development and have now been successfully implemented for 1 experimental run. Whilst analyses is ongoing, due to experimental difficulties during beamtime it is already known there are gaps in the collected datasets that additional beamtime is required to fill.

To continue and complete our study we will request further beamtime. It proves that the new TiC works well up to at least 1400 °C and we managed to make good starting polycrystaline samples. By adjusting the Al_2O_3 buffer rod length, the samples are in the middle of the X-ray window which allows its length to be determined. After a few test experiements, we are now also able to get good ultrasonic signals of our samples at high pressure and high temperature conditions. So we believe we find the suitable way to measure the acoustic velocites of ringwoodite. The Mg-endmember sample (run-5) and Fo70 sample (run-6) have been successfully measured up to 1400 °C and we would like more beam time to further study other Fe-bearing compositions also to such high temperatures corresponding to the mantle transition zone conditions.

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