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High-precision neutron-diffraction measurements for the determination of low-level residual stresses in a sandstone

L. Pintschovius[a,*], M. Prem[b,c], A. Frischbutter[d]

[a]*Forschungszentrum Karlsruhe, INFP, P.O. Box 3640, D-76021 Karlsruhe, Germany*
[b]*Institut für Experimentalphysik, Universität Wien, Strudlhofgasse 4, A-1090 Wien, Austria*
[c]*Laboratoire Léon Brillouin, C.E.-Saclay, F-91191 Gif-sur-Yvette Cedex, France*
[d]*GeoForschungsZentrum Potsdam, Telegraphenberg C3, D-14473 Potsdam, Germany*

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Abstract

Strain analysis by means of neutron diffraction usually has an accuracy of about 10^{-4}, which is far too inaccurate in view of the very low level of residual strains to be expected in most geological samples. We report a method for an improvement of the accuracy by more than an order of magnitude. Using this technique, we were able to determine quantitatively a load stress as low as 1 MPa applied to a sandstone sample. The same technique allowed us to show that there are residual stresses in the order of 1 MPa in a Cretaceous sandstone from the Elbezone. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

So far, the available data on the stresses acting in the Earth's crust are fragmentary and often only qualitative in nature. Recently, it has been proposed to exploit the residual stresses frozen in geological samples as an additional source of information for the stresses which act in situ (Frischbutter, 1998). Neutron stress analysis seems particularly well suited for the determination of residual stresses in geological samples because the penetration depth of neutrons is several centimetres rather than about 0.01 mm as in the case of X-rays. This means that stresses determined by neutron diffraction are not influenced by surface effects, but can be considered as representative for the bulk. However, although neutron stress analysis is a well-established technique in materials science, its successful application for geoscientific problems, i.e. the study of geological samples, is still to be shown. We note that neutron stress analysis has been developed for technical components made of steel, aluminium or other metals, in which residual or load strains are typically in the order of 10^{-3} (Pintschovius et al., 1983). Hence, it was usually sufficient to measure strains with an accuracy of 10^{-4}. When neutron stress analysis was later applied to components made of ceramic materials such as aluminium oxide or silicon nitride, the precision of the measurements had to be substantially increased, because residual strains in ceramic samples are usually quite small, typically only a few times 10^{-4}. Using high-resolution neutron diffractometers, a precision as high as 2×10^{-5} has been achieved (Pintschovius et al., 1989). However, it is very unlikely that even this precision will be sufficient for the study of geological samples in general, because many geological samples are expected to show very low levels of residual strains. When a rock is brought to the Earth's surface, either by drilling (sampling) or by geological processes (uplifting), stress relaxation will wipe out the macrostresses and will instead induce microstresses (grain interaction stresses or phase-specific stresses) which are, however, only a small fraction of the original macrostresses. For this reason, geological samples studied in the laboratory will show stresses that are an order of magnitude smaller than those acting in situ. This means, for example, that a lithostatic pressure of about 25 MPa (corresponding to a depth of 1 km and a medium density of approximately 2.5 g/cm^3) will give rise to residual stresses in the order of only 1 MPa (or a few MPa at most). One might think to restrict application of neutron stress analysis—at least in the first instance—to samples which were subjected to very large in-situ stresses in which relatively large residual stresses may be expected after extraction from the Earth's crust. However, it was frequently observed that such samples develop cracks whereby the geological information contained in the residual stress state will be lost even

* Corresponding author. Fax: +49 7247 824 624.
E-mail address: pini@ifp.fzk.de (L. Pintschovius).

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when the residual stresses relax only partially on crack formation. As a consequence, we consider it as indispensable to demonstrate from the methodical point of view that the neutron diffraction technique can be brought to such a high precision that residual stresses of the 1 MPa level can be detected reliably. In this paper we will show that neutron diffraction experiments using a constant wavelength can indeed be conducted in a way so as to achieve a precision of the strain values of a few times 10^{-6}. Further, we report on the first measurements on a Cretaceous sandstone sample which yielded evidence of residual stresses of about 1 MPa. This indicates that the sensitivity of the improved neutron diffraction technique is sufficient for the detection of low-level residual stresses in geological samples.

2. Experimental technique

2.1. Sample preparation

The samples were prepared from a drilling core from a ground water well in the Elbezone (near Dresden, Germany). They represent marine Cretaceous, greyish-white sandstone with quartz as the main component (>90%), the rest being feldspar, calcite and mica. The samples were prepared from a drilling depth of about 250 m below surface level. The grain size is in the range of 200–500 μm. No preferred crystallographic orientation was found. Further details on the geological setting and on the composition of the samples can be found in Scheffzük et al. (1998).

The sample used for the residual stress investigation (labelled K2/2) was a cylinder of 100 mm length and 38 mm diameter, prepared within 24 h of core extraction. The K2/2-sample was pushed up to half of its length into a precision steel tube fabricated with practically the same inner diameter as that of the K2/2-sample. This was done to create two different (although only known with regard to its quality) stress states within the enclosed part (radial compression and axial extension) and the free part as a test case for the capability of the neutron diffraction technique to detect this difference. Pushing the sample into the steel tube resulted in squeezing some pore water out of the sandstone, which means that the stress state in the enclosed part had indeed been significantly altered (Scheffzük et al., 1998). The orientation of the cylinder axis relative to the drilling core can be seen in Fig. 1.

The sample used for the experiment under applied load was a cylinder of 50 mm length and 30 mm diameter, which was prepared from the same drilling core and is characterized by the same geological features.

2.2. Instrumental details

The test measurements were performed on the G4.3 triple-axis spectrometer located at a guide of cold neutrons at the Laboratoire Léon Brillouin, Saclay. The instrument

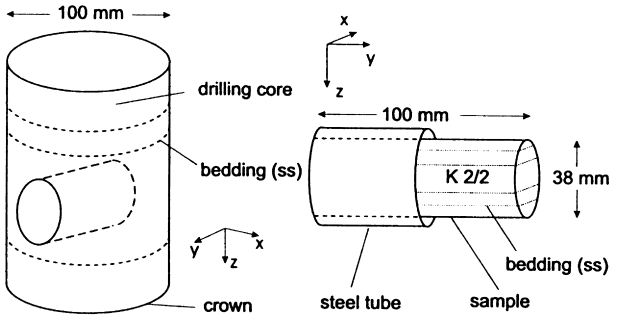

Fig. 1. Schematic representation of the K2/2-sample and its orientation in respect to the drilling core (from Scheffzük et al., 1998).

was operated in the diffractometer mode, i.e. the analyser was set to the same neutron wavelength as the monochromator. In this way, a triple-axis spectrometer acts as a normal neutron diffractometer, whereby the analyser somewhat improves the resolution compared with an instrument without an analyser. Tight collimation of 10 min was used before the sample and before the detector in order to achieve a high resolution, whereas the collimation between the sample and the analyser was more relaxed (30 min) for intensity reasons. The in-pile collimation was defined by the critical angle of the neutron guide, which corresponds to an effective collimation of about 50 min for a wavelength $\lambda \approx 0.5$ nm. The useful wavelength range of the instrument is $0.57 > \lambda > 0.2$ nm, whereby the lower limit of 0.2 nm is due to the fact that the instrument is fed by cold neutrons. We decided to use a neutron wavelength as long as possible (0.572 nm) in order to achieve a high resolution. Using sandstone samples with quartz as the main component, the strains were deduced from shifts of the (0111/1011)-peaks of quartz, which were observed at $2\theta \approx 116°$. The internal probe regions within the samples were defined by slits in the incoming and in the diffracted beam of dimensions 1×2 cm^2. Hence, the volume of the internal probe region was about 2 cm^3.

2.3. Experimental errors

Neutron stress analysis is based on the determination of interplanar distances d_{hkl} using Bragg's law

$$2d_{hkl} \sin \theta = n\lambda \quad (1)$$

with $\{hkl\}$ being the Miller indices, θ the scattering angle, λ the neutron wavelength and n a small integer number (mostly $n = 1$ in practice). Hence, the precision with which a strain can be determined is directly related to the precision with which the position of a diffraction peak can be determined experimentally. Errors of the peak position can be classed into statistical errors and systematic errors. Statistical errors are primarily related to the counting statistics, but small fluctuations in the settings of the spectrometer when stepping the angle 2θ can be considered as statistical errors as well. We found

that errors related to the counting statistics can be made fairly small in measurements on a geological sample. On our instrument, a total counting time of 2 h led to a statistical error of the peak position $\Delta 2\theta \approx \pm 0.0006°$ corresponding to a precision of the strain value of $\approx 3 \times 10^{-6}$. Therefore, if errors related to the counting statistics are the only ones, it should be easily possible to detect residual stresses well below 1 MPa.

Unfortunately, positioning of detector components of actually available diffractometers is not precise enough to achieve an error level much below 0.01° without further precautions. The angular movements of our diffractometer (and of practically all other available neutron diffractometers as well) are encoded with a stepwidth of 0.01°. Consequently, any target position closer than this value cannot be expected to be reached. This means in practice that the real values of the angular settings differ from the nominal ones by largely random amounts in the order of 0.01°. When evaluating the peak position, this angular scatter is partially averaged out, but an error $0.01°/\sqrt{n}$, with n equal to the number of steps, will remain. In order to reduce this contribution to well below 0.001° it will be necessary to conduct many scans each with a small stepwidth, so that n becomes sufficiently large. A more efficient strategy to eliminate this error source will be described in the next section.

When attempting to increase the precision of the strain measurements by repeating the scans many times we became aware of a drift of the 2θ-offset in the order of a few times 10^{-3} degree over a period of a day. This drift is certainly specific to the instrument used and may or may not occur on other instruments as well. In our case, this drift seriously limited the achievable precision. The optimized measurement technique described in the next section eliminates this error source, too.

A serious problem for the determination of highly precise strain values is related to absorption effects. In view of the fact that these absorption effects are rarely taken into consideration, we will describe them in some detail in the following. When the neutron beam enters into the sample it is attenuated by scattering and by absorption. For the effect under consideration it is irrelevant by what mechanism the beam is attenuated, so we will simply speak of absorption. In geological samples the mean free path will be 1–3 cm (for quartz, the mean free path is 2.3 cm).

As a consequence, different areas of the internal probe region will contribute with different weight to the total intensity depending on the total flight path of the neutrons in the sample. Fig. 2 shows a typical case for a bar-like sample investigated in reflection (Fig. 2, top) and in transmission geometry (Fig. 2, bottom). Absorption effects will shift the absorption-weighted centre of the internal probe region by about 1 mm from the geometrical centre for 1-cm-wide slits in front of and behind the sample for measurements in reflection geometry, whereas no such shift occurs in transmission geometry. The shift of the centre-of-gravity leads to an apparent shift in 2θ of the order of some 0.01°, i.e. a fairly large value when compared with the error due to the counting statistics. Using a tight collimation (10 min) will reduce this error by roughly a factor of three which is, however, not sufficient for our purpose. A possible remedy would be the use of a much tighter collimation (unavailable on present-day diffractometers) or the use of narrow slits before and behind the sample. Both measures would cause a very drastic loss in intensity and thereby cause a very significant increase in the statistical errors. Moreover, narrow slits before and after the sample will generate a very small internal probe region, so that the results might not be fully representative for the bulk of the sample. For these reasons, we adopted a different strategy as described below.

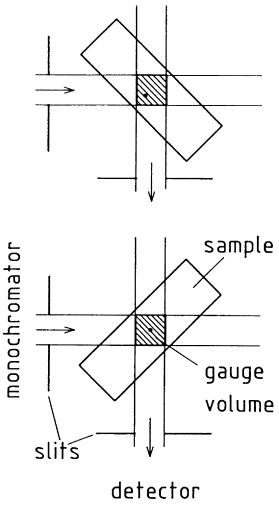

Fig. 2. Schematic of the neutron flight path in a bar-like sample investigated in reflection geometry (top) and in transmission geometry (bottom). The full points within the gauge volume denote the absorption-weighted centre-of-gravity.

2.4. Optimized measurement strategy

Errors due to the imprecision of the angular settings are eliminated when for each angular setting of the diffractometer the sample is exchanged with a reference sample and thus two scans are conducted for completely the same steps in 2θ. Errors associated with absorption effects will be eliminated at the same time if the reference sample has the same shape and is placed on exactly the same position on the sample table. The penalty to be paid is a fourfold increase in beam time to achieve the same statistical accuracy as in a single scan on the sample of interest. This increase in beam time can be somewhat reduced if more than one sample is interchanged with the reference sample. A systematic use of this measurement strategy needs an automated interchange of samples which, however, is easy to realize using computerized translation tables as are currently employed on neutron stress scanners.

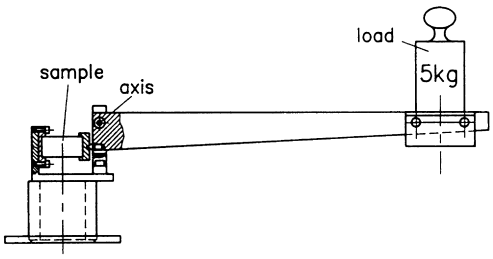

Fig. 3. Press for a uniaxial pressure of 1 MPa. The lever carrying the load had a length of 500 mm. The sample diameter was 35 mm.

3. Experimental results

3.1. Load stresses

Although the optimized measurement strategy described above promises to achieve an accuracy of the peak position limited by the counting statistics only, it is not obvious whether such a high precision can be achieved in practice. For a check, we performed a measurement in which a sandstone sample was loaded with a uniaxial pressure, which was chosen deliberately very low, i.e. about 1 MPa. The loading apparatus is depicted in Fig. 3. It was designed to apply the desired rather low load stress in a reliable and reproducible manner. The load was orientated perpendicular to the bedding plane (defined by the axes [x] and [y]).

A scan was performed with and without load for each angular setting. The results are depicted in Fig. 4. The observed peak shift is $\Delta 2\theta = (0.0027 \pm 0.0008)°$. This peak shift corresponds to a strain $\varepsilon = (1.5 \pm 0.45) \times 10^{-5}$. Under the assumption of a uniaxial stress-state (which is very reasonable in view of the loading conditions) and using a value of $E = 70$ GPa for Young's modulus as determined by Scheffzük et al. (1999) on a similar sample using much larger loads, we find a stress value of $\sigma = (1.05 \pm 0.3)$ MPa, which compares very well with the value calculated from the mechanical load, i.e. 1.1 MPa.

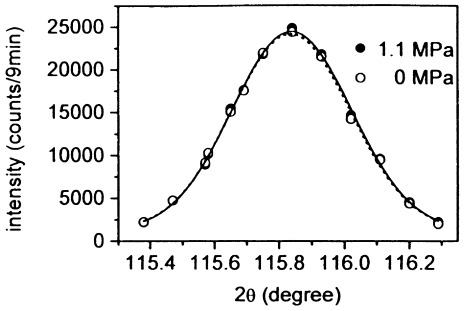

Fig. 4. Neutron diffraction profiles of the (0111/1011)-peak of quartz observed at ambient pressure (open circles, dotted line) and under a uniaxial pressure of 1.1 MPa (full circles, full line). Note that the scatter of the experimental points around the fit curves is largely due to positioning inaccuracies rather than statistical fluctuations which, however, cancel each other out in the evaluation of the peak-shift.

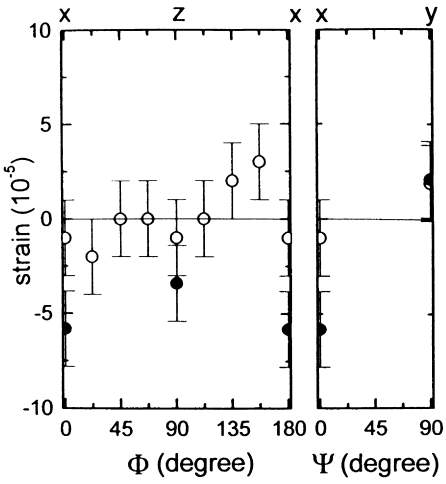

Fig. 5. Residual strains observed in the K2/2-sample in the x–z-plane and in the y-direction. Open circles and filled circles refer to the free-standing part and the enclosed part of the sample, respectively.

3.2. Residual stresses

In a first experiment, the residual stresses in the K2/2-sample were investigated by a series of scans sampling the strains in the directions x–y–z of the sample system (see Fig. 1) both in the free-standing and in the enclosed part. The value of the stress-free lattice parameter d_0 was deduced from measurements on a sample which was obtained by milling a piece of the same sandstone up to a grain size of <62 μm, heating the powder over 24 h at 500°C, followed by gradual cooling. The results are depicted in Fig. 5. We note that these results were not obtained using the technique described in Section 2.4 but are averages of 5–10 individual scans of rather short duration (≈10 min). Our experiment was carried out nearly one year after a first measurement of the same sample by Scheffzük et al. (1998). The experiment did not confirm the rather large strain values reported by Scheffzük et al. (1998) from measurements just five days after taking the sample from the drilling core. Rather, the residual strains observed in the free-standing part of the sample are practically zero within the error limits of this experiment, i.e. 2×10^{-5}. A non-zero stress level could be concluded only for the enclosed part, i.e. a stress of about 5 MPa in the radial direction of the sample. Since the residual strains in the free-standing part were obviously much smaller than that, the precision of the measurements had to be increased in the search for finite residual stresses.

Following the pressure experiment described above, we used the same technique to search for residual stresses in the K2/2-sample. In view of the fact that the interchange of the sample with the reference sample was not yet automated and that rather little beam time was allocated to this experiment—which was considered as a test only—the results obtained so far are rather incomplete. These results are displayed in Fig. 6. They confirm the presence of radial stresses of about 5 MPa in the enclosed part of the sample

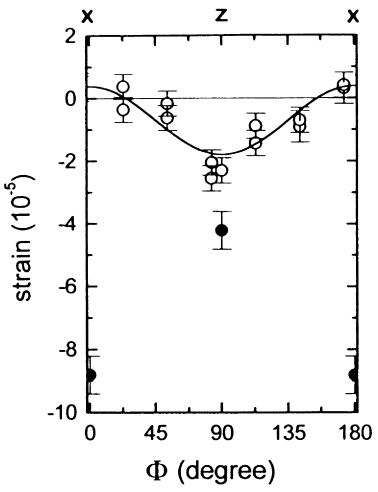

Fig. 6. Residual strains observed in the K2/2-sample using the method described in Section 2.4. Open and filled circles refer to the free-standing part and to the enclosed part of the sample, respectively. The line was obtained by a fit with a sine-function.

found in the previous experiment. More importantly, they provide evidence for non-zero residual stresses in the free-standing part of the sample. The observed anisotropy of the residual strains corresponds to an anisotropy of the residual stresses $\sigma_x - \sigma_z \approx 1$ MPa. This anisotropy is probably the signature of a non-hydrostatic in-situ stress in the geological setting. However, no quantitative conclusions concerning the in-situ stresses can be drawn until the relationship between in-situ stresses and residual stresses is explored by further investigations.

4. Conclusions and outlook

In this report, we have shown that neutron diffraction measurements using available diffractometers allow one to detect and even to determine quantitatively residual stresses in geological samples of very low level, i.e. in the order of 1 MPa. The use of the optimized measurement strategy described above is greatly facilitated by an automated interchange of the sample of interest with the reference sample. This is normally not implemented on general-purpose diffractometers, but technically not difficult, and neutron diffractometers dedicated to residual stress analysis generally allow this possibility. Our measurements of the residual strains in a Cretaceous sandstone indicate that such samples do contain residual strains of a level which are detectable by neutron diffraction measurements. These measurements were, of course, only a first step of a study which attempted to obtain information on the in-situ stresses from neutron stress analysis on geological samples. The next step is obvious, i.e. a complete characterization of the stress tensor of the K2/2-sample already studied in this report. The following step is also rather obvious, i.e. the study of related samples to search for a correlation between the observed strains and the geological setting. Further steps are less obvious, but are nevertheless indispensable in order to explore the relationship between the in-situ stresses and the residual stresses after extraction. This relationship will be sample specific in that it will depend on the composition and both the macro- and microstructure. It will need some modelling as well as some experiments designed to shed light on stress relaxation on cutting a sample out of its environment. At present, it cannot be said how far we will be able to understand the stress relaxation phenomena to allow us to draw quantitative conclusions concerning the in-situ stresses. Still, we consider the results obtained so far as sufficiently encouraging to pursue further investigations of this kind.

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