The use of cold neutrons and monochromatic X-rays for NDT in geology

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Abstract. Both neutrons and X-rays have very interesting properties for tomography. Tunable monochromatic X-ray beams were used for element sensitive tomography of geological samples containing heavy elements using the K-edge dichromatic scanning technique. This research was done at the high-energy beam-line of the ESRF (France). A new set-up has been build for cold neutron tomography at the spallation source SINQ at PSI (Switzerland). The high flux cold neutron beam transmissions are observed with a CCD camera via a neutron-to-visible-light converter. Many samples have been investigated with parallel beam geometry. The possibilities of neutron NDT in the field of geology are discussed.

Introduction

Since a couple of years our research team has been working in the field of tomography [2],[3], [4]. The use of intense monochromatic X-ray beams led to the possibility of element sensitive 3D tomography. This research was done at the high-energy beam-line of the European Synchrotron Radiation facility (ESRF) in France and at the linear electron accelerator of the Ghent University in Belgium.
A Cold neutron beam tomography setup for non-destructive testing of samples has been installed at the spallation source SINQ at the Paul Scherrer Institute (PSI) in Switzerland.

X-ray tomography has been used for nondestructive analysis of samples for a long time. The same procedure can be applied using neutrons. By looking at the attenuation of the neutron beam in the sample from different angles, it is possible to do a neutron tomography. Because the properties of neutrons are so much different from X-rays, a complementary range of applications can be found. Due to the different nature of interaction and the complementary Z-dependent cross sections, X-rays mainly used to study samples containing light elements whereas neutrons are interesting probes for the investigation of samples containing heavy elements. There are some disadvantages with the use of neutrons like the activation of samples and equipment due to nuclear reactions.

Experimental Set-up used for computed tomography

The standard procedure for tomography is to measure the attenuation of the radiation through a sample for several orientations. The recorded patterns are used to reconstruct the three-dimensional attenuation distribution for the whole sample. A completely automated transportable measurement setup was developed, which can be used for X-ray and neutron tomography. A schematic presentation can be seen in figure 1. The X-rays or neutrons are converted to visible light after passing the through sample in an appropriate scintillator. To protect the CCD camera from the damaging radiation, the visible light is reflected out of the X-ray or neutron beam by a surface coated mirror. The entire system is shielded with lead and lead glass in the case of X-ray tomography and with lead, boron and lithium in the case of neutron tomography.
During the image acquisition the beam intensity is being monitored by a National Instruments PC counter card. In case of beam loss or other non-predictable events, the image acquisition is stopped.

The resolution of a standard X-ray intensifying screen, usually Gd_2O_2S:Tb, is of the same order as the scintillator thickness (>50 µm). For a parallel beam tomography configuration 50 µm is too much. Therefore we opted for a new type of scintillator, called Yag (Y3Al5O12:Ce) with a resolution of less than 1 µm.

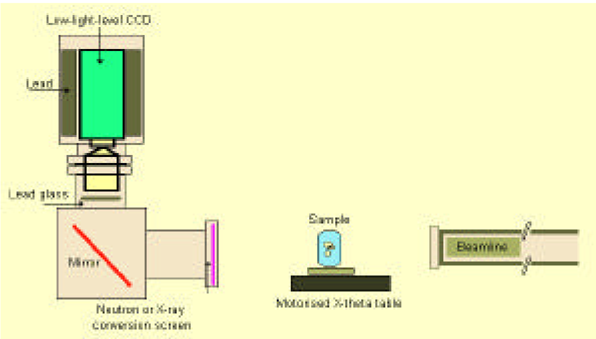

FIGURE 1. A schematic representation of the tomography set-up. In the case of neutrons a lens can be placed between the sample and the beam-line.

The tomography set-up is completely automated which allows the operator to leave the instrument. When the storage ring needs to be refilled or if there is an accelerator problem the tomography is automatically paused and restarted when the particle beam is back on-line.

The neutron-to-visible light converter, manufactured by Applied Scintillation Technologies, consists of a dispersion of ZnS(Ag) and ^{6}LiF in an acrylic binder. The detection mechanism is based on the ^{6}Li(n,α)^{3}H nuclear interaction. The α's and the tritons interact with the phosphor to create scintillation events that can be detected by the CCD camera. The conversion screen emits a spectrum in the blue region with a wavelength peak at 460 nm. The advantage of this converter is the relatively low sensitivity to background γ radiation. This ZnSLiF layer is deposited onto a reflecting Al plate to increase the intrinsic efficiency. Backwards emitted light is reflected by the plate into the forward direction. The thickness of the converter is 420 µm. To increase the resolution, thinner scintillators have been tested. The increase in resolution comes with lower conversion efficiency.

Monochromatic X-rays for Element Sensitive tomography

Principle:

Every heavy element has large photon absorption discontinuities around the energies corresponding with the binding energy of the atomic electrons with the nucleus. These discontinuities are called absorption edges. The abrupt change of the X-ray attenuation around the edge together with the fact that the K-shell electron binding energy is different for every element makes it possible to perform element sensitive radiographs.

High energy X-rays have two advantages. If we look at the X-ray absorption coefficient, we see that it decreases with energy for energies below 511 keV. Therefore hard X-rays can penetrate thicker or denser samples than low energy X-rays. Heavy elements have K-edges at high energy with large discontinuities, which makes them perfect for element sensitive tomography based on dual energy scanning.

Another advantage of the monochromatic beam in comparison with a white or polychromatic beam is the absence of beam hardening. Hardening of the beam leads to artifacts that are very difficult to correct.

By using a crystal monochromator it is the possible to change the photon energy in a continuous way by changing the Bragg angle.

Synchrotron radiation source ESRF:

For all experiments at the ESRF we used the superconducting wavelength shifter at ID15. There are permanent filters in the beam (0.7 mm C, 4.0 mm Be, 4.1 mm Al). The beam intensity is $5.13 \cdot 10^{13}$ ph s^{-1} $mrad^{-2}$, 0.1% bw, 0.1A at 95 keV. The energy resolution is 1keV. The polychromatic X-ray beam is converted into a monochromatic beam with two 5 mm thick asymmetrically cut Si crystals working in fixed Laue-Laue mode.

Element sensitive X-ray micro tomography applied to Oklo and Cinnabar stone:

In the figures below we present some results of tunable monochromatic X-ray tomography. A number of samples from the natural fission reactor Oklo in Gabon were studied to determine the uranium and lead distribution. With X-ray absorption tomography it is not possible to separate the isotope U235 from the isotope U238. Therefore the same Oklo samples were also scanned with position sensitive Prompt Gamma Activation Analysis (PGAA). Natural uranium is always of the same isotopic composition 99.27 percent Uranium-238, 0.72 percent of Uranium-235 and traces of Uranium-234. The uranium samples from the Oklo mine showed a lack of U235, which can only be explained by some process other than simple radioactive decay. In 1972, a French physicist Francis Perrin founded the theory of the first nuclear reactor.

In figure 2, we show a radiography of the Oklo stone with monochromatic X-rays at 87 keV. Next to this radiography we present the result of an element sensitive tomography reconstruction of the U-distribution (b) and the Pb-distribution (c). The lead in the sample comes from the radioactive decay of the U-238. In (d) we present the result from a Promt Gamma Activation Analysis scan of the same Oklo sample; the first bar shows the relative distribution of the U-235 and the second bar shows the relative distribution of the U-238 (both relative to the maximum concentration in the sample). In natural uranium containing samples both bars should look identical. This is clearly not the case for this Oklo sample and it therefore proves that this sample has taken part in the natural nuclear reactor.

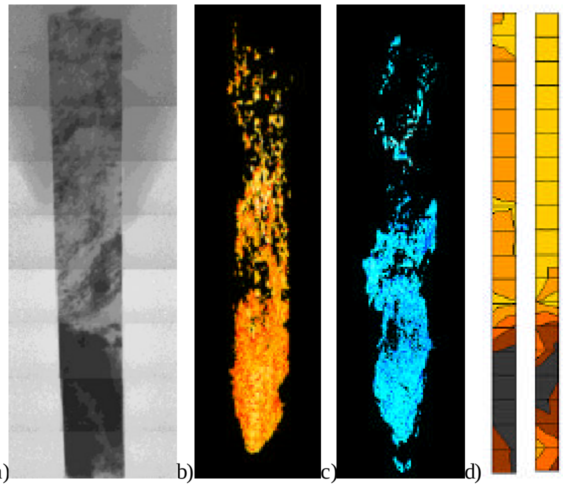
a) b) c) d)

FIGURE 2. Stone coming from a natural nuclear reactor of Oklo in Gabon.(a) Radiograph at 87 keV, (b) 3D reconstruction of the uranium distribution in the sample, (c) 3D reconstruction of the lead distribution. The bars (d) show the individual distributions of ^{235}U and ^{238}U, respectively left and right measured with PGAA. The resolution of the PGAA-scan was 1 mm. The color of the images corresponds to the density. Darker means higher density. The absolute values are not indicated.

The second sample is on which element sensitive tomography has been performed is a cinnabar stone from a mercury mine in Toscana (Italy). In figure 3 we show the result of a tomography with a monochromatic X-ray beam with energy just above the K-edge of mercury. At this energy the absorption contrast is very high. This allows us to identify clearly the mercury (the layer on top of the sample). With a dual energy scan, above and below the K-edge of mercury, we could programmatically separate the mercury from the stone and determine the exact mercury density and quantity present in the stone.

Commercial X-ray devices also use a dual energy technique to identify objects within samples by changing the high-voltage on the X-ray tube. Additionally, dedicated image processing is used to separate different objects superimposed on one another in the projected image. The measured densities are compared with library values of the densities of known materials. This method is nowadays very common in airports for the detection of weapons and explosives.

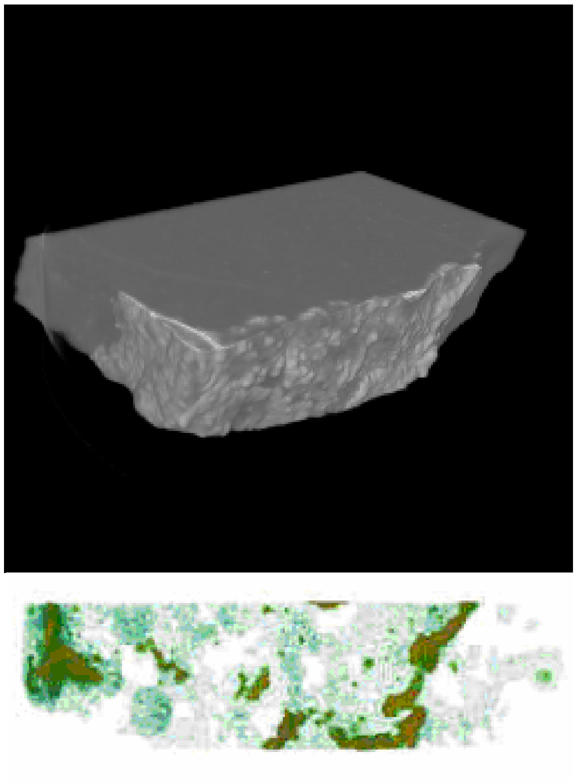

FIGURE 3. Reconstruction of the mercury distribution in cinnabar stone from Toscana.

More results of monochromatic X-ray tomography can be found in [1], [2], [3], [4].

Neutron imaging

First cold neutron tomography experiments were performed at the cold neutron source of SINQ at PSI (Villigen CH). The neutrons are produced by spallation reaction of protons in a lead target. Neutron guides extract cold neutrons to the experiments from a cold moderator of about 20 liters of liquid deuterium, at the very low temperature of 25 Kelvin. The beam intensity at the sample is 10E8 neutrons/cm^2.s.
The spatial resolution, an important parameter for radiography and tomography, of the neutron images has been determined by the sharp edge method. From

the line profile over the edge of a strongly absorbing 25 µm thick Gd foil, one can calculate the line spread function. The line profile is differentiated and a gauss curve can be fitted to the data. The FWHM is expressed in number of pixels. This value is multiplied by the pixel size of the image, including CCD-binning. Figure 4 shows the spatial resolution for a 460 µm converter screen. The image resolution on the screen is 320 µm in the case of the 460 µm converter. The reason for the increasing spatial resolution is the 0.4° beam divergence. One can easily calculate the divergence of the beam from the fit to the data.

$$\text{Div} = \text{Arctan}(\frac{7.3192}{1000}) = 0.42^{\circ} \qquad (1)$$

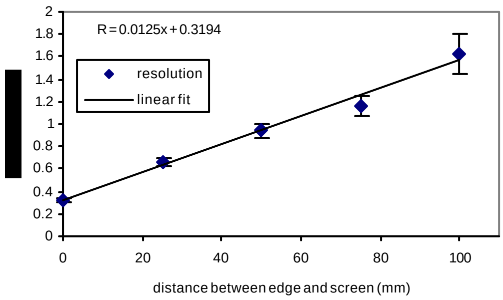

FIGURE 4. Neutron radiograph resolution as function of the distance from the conversion screen.

The cold neutrons are transported from the spallation target to the experiment room by means of supermirror coated neutron guides. The cold neutrons are reflected by the walls and have consequently a certain angle range. The divergence of the beam is also a function of the energy (higher energies have smaller scattering angles, also meaning less divergence):

$$\text{Div}(^{\circ}) = 0.093 \times \lambda(\text{Å}) \qquad (2)$$

The maximum of the spectrum is 4Å. The beam spectrum begins at 1.8 Å has a tail up to 10 Å. The samples are therefore to be placed as close as possible to the screen. For tomography this distance is determined by the sample size.

By using a neutron-focusing lens, it is possible to change the beam geometry from parallel beam to cone-beam. The lens is composed of a large number of poly-capillary fibers, parallel at the lens entrance and bent in such a way that all fibers converge towards a focal point. The diameter of the focused beam at the waist is smaller than 0.5 mm and the flux gain is greater than 30. At the waist a 5mm thick Li pinhole collimator stops background neutrons. The collimator opening defines the source and the diameter of the opening the source size. Normally, this will determine the image resolution. The advantage of the lens set-up is the possibility to magnify the sample projection.

$$\text{Magnification } M = \frac{d}{D} \qquad (3)$$

D is source to screen distance, d the source to sample distance. The resolution of the screen is fixed, but when M is bigger than 2, the resolution becomes better than 250µm. A second property of the lens is the possibility of increasing the beam size to investigate larger samples. A disadvantage of the lens is the lower neutron flux. The acquisition time, with the Sensicam 12bit cooled CCD camera is more than 2 minutes compared to 200ms.

Cold neutron tomography applied to coral-sample:

Figure 5 show the reconstruction of a coral. The size of the coral is about 2 x 2 x 2 cm. Both samples are low Z material and are therefore ideal for neutron tomography. The cross-section as a function of Z, for neutrons, cannot be expressed with a simple equation like for X-rays. Since they have completely different attenuation coefficients, neutron tomography is complementary to X-ray tomography.

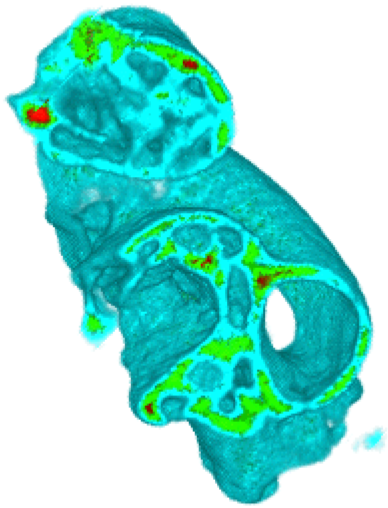

FIGURE 5. Neutron micro-tomography of a coral

The same coral was scanned with X-ray at the Micro focus X-ray source of the department for geology and soil science of the Ghent University [7]. The results of the X-ray scans showed clearly much more details due to the better scanning resolution. Effects of beam hardening were present due to the poly-chromaticity of the X-ray source.

In the near future thermal neutrons will be used for the optimization of surface treatment techniques for building materials. Natural sand stone, as well as concrete samples will be subjected to different NDT methods before and after surface treatment. Neutrons will be used to study the on-line water take-up before and after treatment.

Conclusion

Both, monochromatic X-rays and neutrons, have interesting properties for tomography in the field of geology. We have presented some results of element sensitive X-ray tomography at the ESRF. The neutron tomography image spatial resolution is around 300 µm. This large value is due to the conversion screen thickness and the rather large neutron beam divergence. In spite of the bad image resolution for micro tomography interesting results have been obtained for stone samples from the natural fission reactor Oklo in Gabon.

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