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A multitask neutron beam line for spallation neutron sources

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Abstract – Here we present a new concept for a time-of-flight neutron scattering instrument allowing for simultaneous application of three different techniques: time-of-flight neutron diffraction, neutron resonance capture analysis and Bragg edge transmission analysis. The instrument can provide average resolution neutron radiography too. The potential of the proposed concept was explored by implementing the necessary equipment on INES (Italian Neutron Experimental Station) at the ISIS spallation neutron source (UK). The results obtained show the effectiveness of the proposed instrument to acquire relevant quantitative information in a non-invasive way on a historical metallurgical sample, namely a Japanese hand guard (tsuba). The aforementioned neutron techniques simultaneously exploited the extended neutron energy range available from 10 meV to 1 keV. This allowed a fully satisfactory characterization of the sample in terms of metal components and their combination in different phases, and forging and assembling methods.

Neutron techniques for the characterization of materials are becoming appealing for a wide category of scientists in different fields ranging from metallurgy to archaeology.

In this paper the experimental test of a new neutron beam line concept is presented and the results obtained on a 19th century Japanese hand guard (tsuba) are reported. The proposed new beam line is capable of performing simultaneous investigations with complementary neutron techniques. This approach minimizes neutron exposure time, allowing multiple investigations under the same experimental conditions and at the same time. It also helps minimizing the cost of transportation of the samples to different facilities in order to fully characterize the artifacts through non-invasive methods. As mentioned, simultaneous Time-of-Flight Neutron Diffraction (ToF-ND), Neutron Resonance Capture Analysis (NRCA), Bragg Edge Transmission (BET) and Neutron Radiography (NR) measurements are presented on a metallic composite ancient artifact, namely a Japanese sword hand guard (tsuba) [1,2]. The tsuba is part of the collection of Japanese swords of the Stibbert Museum (Firenze, Italy) and it was manufactured by the Japanese master Hidehisa. The archaeometric goals of these measurements are the identification of the metal components and their combination in different phases, and the determination of the forging and assembling methods.

It is well known that these complementary measurements provide quantitative details on the phase (ToF-ND), elemental composition (NRCA), mosaicity and presence of strains (ToF-ND and BET) and inner structure assembly (NR). All this information together is of importance for any work aiming at characterizing historical and archaeological samples, such as metal artifacts.

ToF-ND is a powerful technique for investigating the crystal structure of materials [3,4]. It provides quantitative information on the phase content through Rietveld refinement [5,6], domain size, strain level and texture, through line-shape analysis [7,8].

Epithermal neutrons impinging onto materials have a probability to be captured by the nuclei, enabling NRCA measurements [9,10]. The absorbed neutron can induce the transition of the nucleus to an excited state and the

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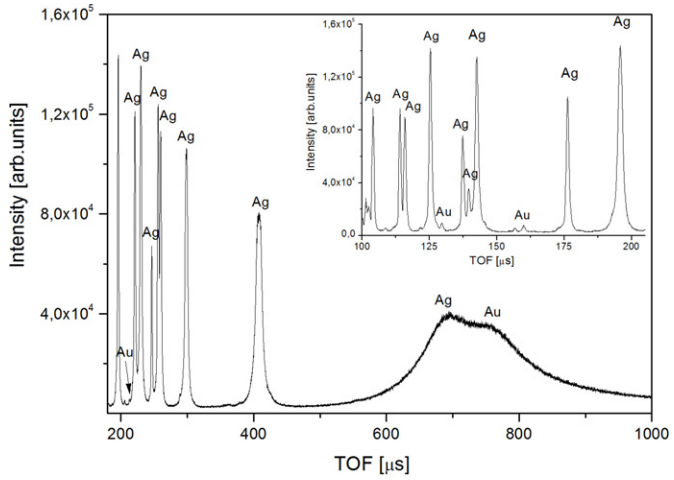

Fig. 1: Neutron Resonance Capture Analysis spectrum recorded by the YAP from the tsuba in the ToF region up to 1 ms. The peaks are labeled indicating the corresponding recognized element.

subsequent de-excitation to the ground state produces a prompt cascade of gamma photons. It is possible to derive the energy of the absorbed neutron from the time-of-flight relation using a time-resolved gamma-sensitive detector, so that an elemental analysis is possible by exploiting calibration curves of the cross-sections of the isotopes involved.

Bragg edges appearing in the spectrum of neutron transmitted through a polycrystalline material represent the negative counterpart of the coherent-scattering response at $2\vartheta = 180°$. Structural information such as texture, strain or grain size may be derived from the intensity, position and shape of these features called Bragg edges [11]. In this work we will focus on edge position and qualitative derived information on grain size and texture properties only.

Neutron radiography is a method of testing the internal structure of an object. A neutron beam impinging onto any heterogeneous object is transmitted depending on thickness, density, chemical composition and total cross-section of the material along the line of sight [12].

The simultaneous ToF-ND/NRCA/BET, followed by NR measurements on the metallic tsuba were performed on the INES beam line [13,14] at the ISIS pulsed neutron source [15]. This beam line is characterized by a white pulsed neutron beam, moderated by a water moderator at 295 K, and a sample-to-moderator distance $L_0 \approx 22.8$ m. The area of the beam at the sample position is $3.8 \times 3.8\,\mathrm{cm}^2$. ToF-ND was performed using an array of 144 ^{3}He gas tubes at 20 bar pressure covering an angular range from 12° to 171°. NRCA measurements were performed using an Yttrium-Aluminum-Perovskite (YAP) crystal coupled to a photomultiplier tube and placed on top of the sample tank. This scintillator material has shown to be effective in neutron spectroscopic applications at the ISIS source [16,17]. BET spectra were recorded by a $^{\mathrm{nat}}$Li-glass transmission detector placed at 1.2 m from the sample position at $2\vartheta = 0°$. NR was done, a few seconds before the other measurements, with an imaging device that has been specifically built for sample alignment and can be automatically removed from the beam after use [18].

Table 1: Main peaks of the NRCA spectrum (see fig. 1). The value of the resonance energies E_r was calculated from the ToF position of the peaks using $E = m_n \frac{L_0^2}{2t^2}$, m_n being the neutron mass, L_0 the neutron flight path and t the neutron ToF.

ToF (μs)	E_r (eV)	Element
745	4.9	Au
724	5.2	Ag
408	16.4	Ag
299	30.7	Ag
260	40.3	Ag
256	41.7	Ag
246	45.0	Ag
230	51.4	Ag
221	55.9	Ag
212	60.3	Au

Table 2: Summary of the results of the quantitative analysis of the ToF-ND and NRCA data from the tsuba. For the ToF-ND we report the recognized phases and their relative percentages in weight (wt%) and the cell parameter. For the NRCA we list the identified elements and their relative percentages in atomic % [A%].

Neutron Diffraction			Neutron Resonance Capture Analysis	
Phase	wt%	Cell parameters (Å)	Element	A%
Silver	97.45(3)	$a=b=c=4.085(1)$	Silver	99.922
α-copper	2.42(3)	$a=b=c=3.635(5)$	Gold	0.078
Chlorargite	0.13(3)	$a=b=c=5.541(4)$		

Figure 1 shows the NRCA spectrum recorded for an integrated proton current of 800.0 μAh, while table 1 lists the resonance energies of the labeled peaks using public databases [19]. A semi-quantitative analysis was performed to estimate the weight ratio, ρ, between Au and Ag identified in the sample, by considering the relative intensities of the peak maxima, I_P, in the spectra in fig. 1(a). Following ref. [9], ρ was calculated as $\rho = \frac{I_{P,Au}\sigma_{Ag}(t_2)^2}{I_{P,Ag}\sigma_{Au}(t_1)^2}$, neglecting small corrections, σ_{Ag} and σ_{Au} being the radiative capture cross-sections of natural Ag and Au respectively, and t_1 and t_2 the time positions of the resonance peaks of Ag and Au (see inset in fig. 1). This procedure yields $\rho = 0.078 \pm 0.002$, corresponding to the weight percentages quoted in table 2.

Figure 2 shows the ToF-ND diffractogram (fig. 2(a)) and BET (fig. 2(b)) spectrum, respectively. In the ToF-ND spectrum two face-centred cubic phases (see table 2): β-silver, α-copper [20] can be identified. A third minor phase

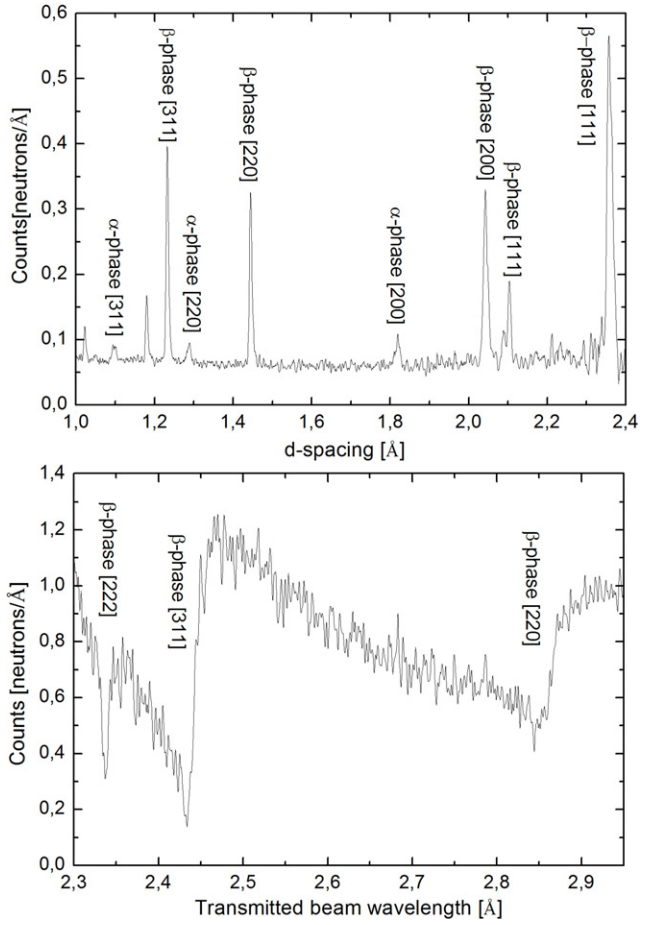

Fig. 2: (a) Time-of-Flight Neutron Diffraction and (b) Bragg Edge Transmission spectra from the tsuba. Some of the main peaks in (a) and the Bragg edges in (b) are labeled with the corresponding phase and the Miller indexes.

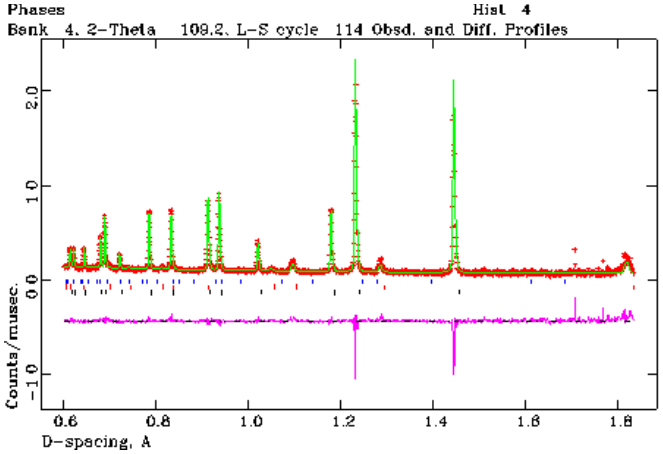

Fig. 3: (Color online) Diffraction pattern as a function of d-spacing (red crosses) of the Japanese tsuba. Rietveld refinement best fit of data is also shown (green line), together with peak position of the different components (bars), *i.e.* silver, copper and chlorargite, and fit residuals (lowest continuous line). The beta phase exhibits texture in [220] and [311] reflections, so that the fit for these single peaks shows an intensities discrepancy. An accurate quantitative analysis taken using different diffraction banks as is the case for ToF-ND cannot include the fit of the texture [22].

was found as chlorargyrite (silver chloride —0.13% in weight), likely formed on the surface through the reaction of silver with the patination solution (chlorine-containing weak acid) commonly used on Japanese metal work to protect them against corrosion [21]. Rietveld refinement of the Japanese tsuba is reported in fig. 3, for one of the nine diffraction banks. The sample is mainly composed of β-silver (97.45%) with a small amount of α-copper (2.42%), most likely added to increase the hardness of the artefact (see table 2). The relative intensities of the two metal phases are slightly different from the ones relative to an isotropic powder. This is due to a weak presence of texture (orientation of the crystallites) induced by mechanical working. The main phase composition of the sample was identified from ToF-ND, while NRCA allowed for recognizing elements in traces (Au).

The Bragg Edge Transmission spectrum in fig. 2(b) is characterized by fairly sharp Bragg edges at neutron wavelengths of 2.34 Å, 2.44 Å and 2.87 Å, corresponding to the [222], [311] and [220] Bragg reflections of the β-silver phase. The FWHM of the edge (measured by means of a Gaussian fit of the five point average smoothed first derivative of the spectrum) is 0.006 Å for the [222] reflection, 0.009 Å for the [311] reflection, and 0.010 Å for the [220] reflection, respectively. These three edges were analyzed since they are the most significant ones for a FCC structure, with the higher-index reflections being just a repetition. These BET structures indicate the presence of average-to-big grain size and of low texture since the shape is quite regular and the rising part is sharp [11]. In the specific case of the tsuba, the full capability of the BET to recognize internal strain was not exploited, as this requires the analysis of BET spectra of two samples with the same composition, one subjected to an applied stress and the other one not [23]. Alternatively, one can obtain BET spectra from different regions of a sample using a pencil beam [24,25] but this was not available on INES. An NR image of the tsuba, obtained with an exposure of 30 seconds, is shown in fig. 4 along with a picture of the sample. The NR image shows a thick part around the blade hole and a thinner outer part. It also shows that the work on both parts was done homogeneously and that no cavities nor casting defects are present. The spatial resolution of the picture is 0.4 mm so that defects smaller than this size are not visible in this measurement.

In summary, a novel concept of a neutron instrument for simultaneous measurements of ToF-ND, NRCA, BET and NR has been tested on the INES beam line at the ISIS spallation pulsed neutron source. The simultaneous use of complementary techniques is very important for non-invasive quantitative characterization of materials of historical and archaeological interest.

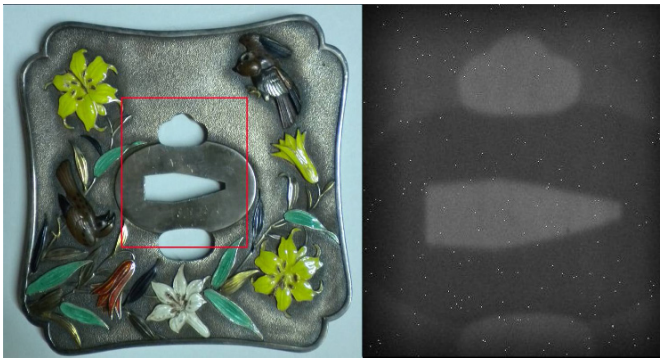

Fig. 4: (Color online) Left: picture of the tsuba (6 cm × 6 cm size and 4 mm nominal thickness). Right: neutron radiography of the irradiated area marked by the red square in the picture on the left.

The different characterization capabilities of the aforementioned techniques make the conceived multitask instrument an effective combination of complementary techniques for the investigation of materials in terms of phase identification, internal strain, texture, and assembly techniques. This instrument can provide solutions for a variety of research fields, ranging from metallurgy to archaeology and cultural heritage.

* * *

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