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NEUTRON AUTORADIOGRAPHY: THE TECHNIQUE

Neutron autoradiography on paintings has been performed since the 1960s. The first description of neutron autoradiography on paintings was published by Sayre and Lechtman (1966; 1968). The technique was further developed at the Metropolitan Museum, leading to the book of M. W. Ainsworth (1982). The interesting findings published in this book lead to the first installation at the thermal column of the Berlin research reactor. This was the start of a very fruitful collaboration.

Since more than 20 years, the Gemäldegalerie in Berlin investigates in close collaboration with the Helmholtz-Zentrum Berlin für Materialien und Energie, the former Hahn-Meitner-Institut, paintings of old masters with neutron autoradiographs (Fischer et al. 1988a; 1988b; Laurenze-Landsberg et al. 2006; Kleinert et al. 2011). Up to now, more than 70 paintings have been investigated. Thus, the Gemäldegalerie is the only museum worldwide employing this analytical technique systematically. Neutron Autoradiographs (NAR) provide images from structures underneath the surface and, in addition, it is possible to identify the pigments by γ-spectroscopy.

In order to adapt the experiment to the specific needs of the paintings, a special irradiation set-up has been installed at the Berlin research reactor, the instrument B8.

The experiment

The instrument B8

Fig. 1 Irradiation chamber of instrument B8. The yellow ellipse marks the end of the neutron guide. The painting is mounted under a small angle with respect to the neutron guide and is moved up and down in order to provide a homogenous irradiation over the whole surface.

The instrument B8 of the Berlin Neutron Scattering Center (BENSC) is dedicated for the irradiation and activation of cultural heritage objects with cold neutrons from the research reactor BER II. The neutron flux at the instrument is $\Phi n = 1 \times 10^{9}\,cm^{-2}\,s^{-1}$. The irradiation time is only three hours, so only about 4 of 10^{12} atoms become radioactive (Fischer et al. 1999). For this reason, the method is considered to be a non-destructive investigation.

The painting is fixed at a small angle (<3 °) with respect to the neutron guide axis with an open end of $3.5 \times 12.5\,cm^{2}$ (**fig. 1**). Thus, a 12.5 cm high and about 120 cm wide stripe of the painting is irradiated. The painting is mounted on a support, which is moved up and down with a few centimetres per minute in order to induce a uniform activation of the selected area. The stroke of the support permits to irradiate entire pictures with dimensions up to $120 \times 120\,cm^{2}$ in one session; larger paintings are investigated in several sessions. The size of the support holds paintings up to $1.7 \times 2\,m^{2}$. The instrument is installed in a dedicated container providing not only

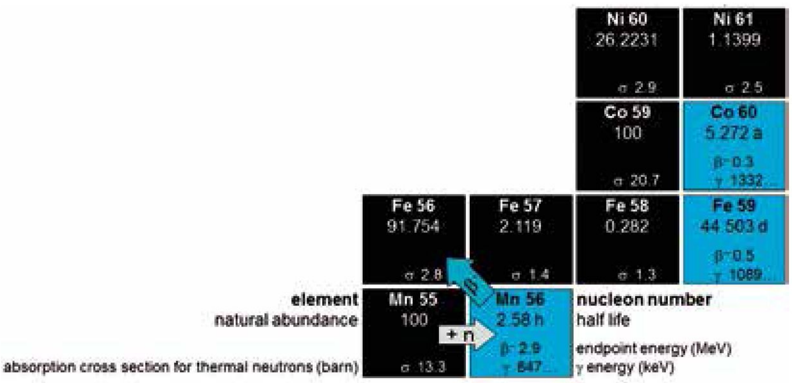

Fig. 2 Extract of the nuclide chart.

isotope	half-life	pigment	visible on NAR no.
^{56}Mn	2.6 h	umber	1
^{64}Cu	13 h	azurite, malachite	2
^{76}As	1.1 d	smalt, realgar, auripigment	3
^{122}Sb	2.7 d	naples yellow	3
^{124}Sb	60 d		4
^{32}P	14 d	bone black	4
^{203}Hg	47 d	vermilion	4
^{60}Co	5.3 a	smalt	

Tab. 1 Pigments typically used in paintings of old masters, the isotopes, their corresponding half-lives, and the number of the NAR, on which they are visible. Calcium, iron, and lead are not visible on NARs, as they do not have suitable radioactive β-emitters.

the required security for the painting, but where also the air humidity can be adjusted to the conservation requirements of the painting. After the irradiation, the painting is analysed on a special table in a shielded, climate-controlled, secure room: the induced radioactivity is measured with X-ray films or imaging plates and analysed with γ-spectroscopy.

Production of radioactive isotopes

During the neutron irradiation different light and heavy isotopes are created. **Figure 2** shows a part of the nuclide chart. In order to provide a useful autoradiograph, one needs a suitable probability for the capture of a neutron, given by the absorption cross section. E. g. ^{208}Pb has a neutron capture cross section for thermal neutrons of $\sigma = 0.00023$ barn. Long-lived isotopes like ^{60}Co (with neutron capture cross section of ca. 37 barn) are a problem, as a long-lived radioactivity is induced. For this reason, an *a priori* knowledge about possible elements in the painting is mandatory. Finally, the energy of the produced β-radiation must be sufficiently high to be detected. For this reason, Fe is an »invisible« element in the neutron autoradiographs. The isotopes relevant for the analysis of old masters are presented in **table 1**.

Data analysis

The area distribution of the induced radioactivity of the isotopes is measured using highly sensitive X-ray films (Kodak XAR5, 35 × 43 cm^2) and imaging plates (Fuji BAS 2000, 20 × 40 cm^2, **fig. 3**). The exposure time depends on the half-life of the specific isotope and for the films ranges from one hour in the beginning for the short-lived isotopes up to several weeks (**fig. 4**). The advantage of the films is that they provide a 1:1 image of the isotope and, thereby, also the pigment distribution with an excellent spatial resolution which is defined by the grain size of the film. The imaging plates are ten times more sensitive than the X-ray films, leading to shorter exposure

Fig. 3 Applying of the X-ray films (**a**) or image plates (**b**).

times. They can be placed on the painting and exposed between two exposures of X-ray films. The information recorded on the imaging plates is read by a laser scanner, which defines the spatial resolution. The data of the image plates are available for further digital processing.

In addition, between exposing the X-ray films and imaging plates a germanium detector with a multi-channel analyser is used to analyse the γ-radiation from the whole painting as well as from specific locations on the painting (qualitatively using the Neutron Activation Analysis technique). The size of the analysed spot is about 5 cm in diameter. This method allows to draw conclusions about the complete range of pigments that have been employed in the painting and of pigment mixtures used for colouring (**fig. 5**).

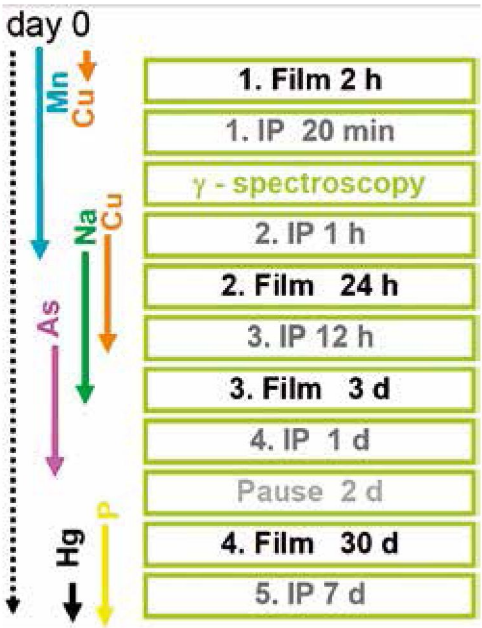

Fig. 4 Time schedule after the irradiation.

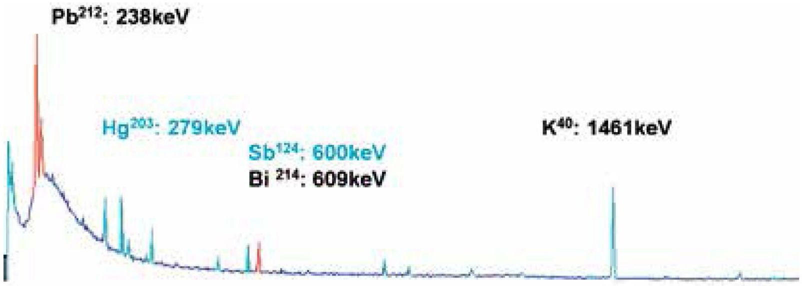

Fig. 5 γ-spectrum of a painting, showing lead, mercury and antimony. The potassium line is due to room background.

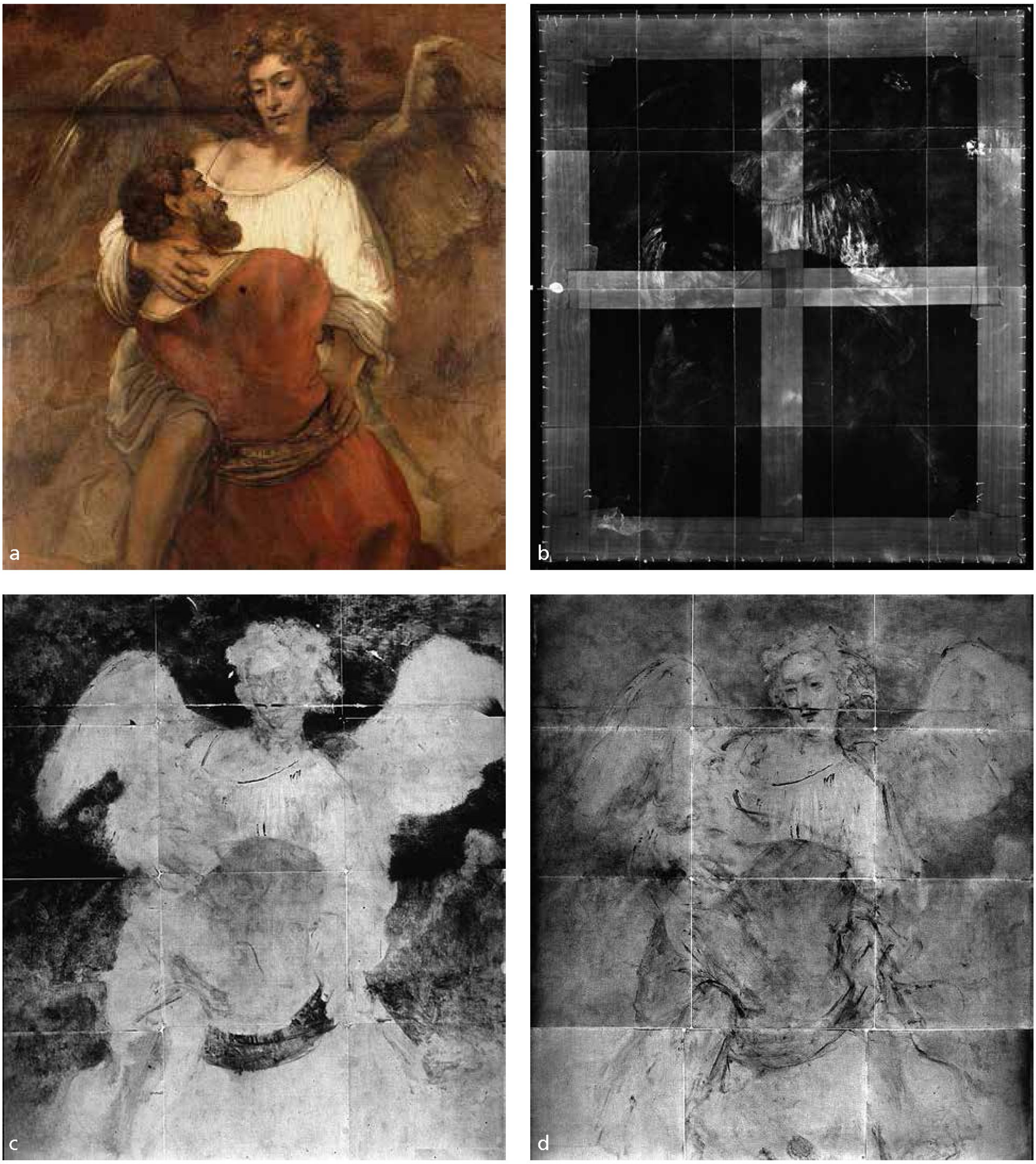

Fig. 6 Rembrandt, »Jakob wrestling with the angel«, ca. 1659/1660, 137 × 116 cm^2, Gemäldegalerie, Staatliche Museen zu Berlin: **a** photo. – **b** X-ray image. – **c** autoradiograph; As (smalt) – background and belt. – **d** last autoradiograph; P (boneblack) – sketch, background; Hg (vermillion) – lips.

Example: »Jacob wrestling with the angel«

As an example the neutron autoradiographs of »Jacob wrestling with the angel« is shown. This painting by Rembrandt (**fig. 6a**) dates around 1660 (Laurenze-Landsberg, this volume). The X-ray image (**fig. 6b**) mainly shows the distribution of lead, used in lead white in the highlights, e. g. on the angels face as well as in the white shirt of the angel. Whereas in the X-ray image white areas mean a high abundance of the element – as the element has blocked the X-rays from reaching the film – in neutron autoradiographies dark areas represent areas with high elemental abundance: Here the emitted radiation of the element in question has blackened the film. The neutron autoradiographies provide further, complementary information: In the third autoradiography, the distribution of Arsenic becomes visible (**fig. 6c**). Arsenic is a component of smalt, which was used here to change the handling properties of the paint. Additional information is supplied by the last autoradiography (**fig. 6d**), showing the distribution of the long lived isotopes of Phosphorus and Mercury. The art historical significance of these images will be discussed in the following section by Claudia Laurenze-Landsberg.

Safety issues

The induced radioactivity decays with time and the most activated nuclides are very short-lived. The exposure time is calculated with respect to the expected pigments. A short test irradiation provides further information about the elements present and the main irradiation is only performed after careful investigation of the results of the test irradiation. Only four in 10^{12} atoms become radioactive. The experimental schedule is about six to eight weeks. The last film layer alone remains for about 30 days on the painting, because almost the entire radioactivity has already disappeared. After that, the residual radioactivity of the painting is measured and documented. Earlier experiments performed on mock-ups have shown that no damage to the painting is induced. Finally, the painting is returned to the museum going through a standard control procedure regulated by the authorities, which certifies that there is no risk for the public or the conservators.

Conclusion

In the larger museums, infrared reflectography and X-ray transmission are available and used as standard methods to investigate paintings. For NAR, it is necessary to move the painting to a neutron source. However, this increased effort yields in a non-destructive way two-dimensional distribution of elements and thus the pigments. The actual condition of the painting becomes visible; changes in composition and information about the artist's brushstroke are obtained. In addition, information about elements not visible in photon techniques, such as Phosphorus can be obtained.

References

Ainsworth 1982: M. W. Ainsworth, Art and Autoradiography. Insights into the genesis of paintings by Rembrandt, van Dyck, and Vermeer (New York 1982).

Fischer et al. 1988a: C.-O. Fischer / J. Kelch / W. Leuther / K. Slusallek, Die Neutronen-Aktivierungsanalyse. Verfahrenstechnik und Anwendung am Beispiel eines Gemäldes von Esaias van de Velde. Restauro 94/4, 1988, 259-268.

1988b: C.-O. Fischer / J. Kelch / W. Leuther / K. Slusallek, Weiterentwicklung der Technik der Neutronen-Autoradiographie durch Verwendung eines Neutronenleiters. Jahrestagung des Arbeitskreises Archäometrie, 1988/03/02-1988/03/05 (Bonn 1988) n/a.

Fischer et al. 1999: C.-O. Fischer / M. Gallagher / C. Laurenze / C. Schmidt / K. Slusallek, Digital imaging of autoradiographs from paintings by Georges de la Tour (1593-1652). Nuclear Instruments and Methods in Physics Research A 424, 1999, 258-262.

Kleinert et al. 2011: K. Kleinert / A. Denker / C. Laurenze-Landsberg / M. Reimelt / B. Schröder-Smeibidl, The genesis of Jan Steens painting »As the old ones sing, so the young ones pipe« from the Gemäldegalerie Berlin. Nuclear Instruments and Methods in Physics Research A 651, 2011, 273-276.

Laurenze-Landsberg et al. 2006: C. Laurenze-Landsberg / C. Schmidt / B. Schröder-Smeibidl / L. A. Mertens, BENSC neutrons for cultural heritage research: Neutron autoradiography of paintings. Notiziario: Neutroni e Luce di Sincrotrone 11, 2006, 24-27.

Sayre / Lechtman 1966: E. V. Sayre / H. N. Lechtman, Revelation of Internal Structure of Paintings Through Neutron Activation Autoradiography, 1966 Proceedings First International Conference on Forensic Activation Analysis (San Diego, CA) 119-132.

1968: E. V. Sayre / H. N. Lechtman, Neutron Activation Autoradiography of Oil Paintings. Studies in Conservation 13, 1968, 161-185.

Summary / Zusammenfassung

Neutron Autoradiography: the Technique
In collaboration with the research reactor in Berlin, the Helmholtz-Zentrum für Materialien und Energie, the Gemäldegalerie Berlin is the only institute worldwide, which systematically employs the method of Neutron-Activation-Autoradiography to analyze paintings. Today we have investigated about 70 paintings. This is an effective, non-destructive, however, exceptional method for the investigation of paintings and paint techniques. It allows the visualization of structures and layers beneath the top surface and, in addition, enables the identification of elements contained in the pigments. The instrument B8 at the research reactor BER II is dedicated to this research. It allows to irradiate and activate artistic, technical, or geological items (foils, stones, etc.) and other materials with cold neutrons having a flux of $\Phi n = 1 \times 109\,cm^{-2}\,s^{-1}$ and to investigate it afterwards with imaging plate technique and/or to analyse it with gamma spectroscopy. The irradiation process, the investigation of the induced radioactivity and the possible identification of the pigments are presented.

Neutronen-Autoradiographie: die Technik
Die Gemäldegalerie Berlin ist das einzige Institut weltweit, das in Zusammenarbeit mit dem Forschungsreaktor in Berlin, dem Helmholtz-Zentrum für Materialien und Energie, systematisch die Methode der Neutronen-Aktivierungs-Autoradiographie verwendet, um Gemälde zu analysieren. Bis heute haben wir ungefähr 70 Gemälde untersucht. Die Methode ist effektiv, zerstörungsfrei, aber außergewöhnlich zur Untersuchung von Gemälden und Maltechniken. Sie erlaubt die Visualisierung von Strukturen und Schichten unter der Oberfläche und gestattet zusätzlich die Identifizierung von Elementen, die in den Pigmenten enthalten sind. Das Instrument B8 am Forschungsreaktor BER II ist für diese Forschung eingerichtet. Es ermöglicht, künstlerische, technische oder geologische Proben (Folien, Steine etc.) oder andere Materialien mit kalten Neutronen zu bestrahlen und mit einem Fluss von $\Phi n = 1 \times 109\,cm^{-2}\,s^{-1}$ zu aktivieren und sie anschließend mit der Bildspeicherplattentechnik (imaging plates) und/oder mit Gamma-Spektroskopie zu untersuchen. Der Bestrahlungsprozess, die Untersuchung der induzierten Radioaktivität und die mögliche Identifizierung der Pigmente werden beschrieben.

Keywords

neutron autoradiography / γ-spectroscopy / imaging / paintings