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RADIOGRAPHIC EXAMINATION OF IRRADIATED IN-CORE NEUTRON DETECTORS

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For the non-destructive investigation of irradiated in-core neutron detectors neutron-radiography has been applied. Both by direct and indirect methods a good resolution for critical detector parts was achieved. With these two methods irradiated self-powered neutron detectors with Co-, Er-, Hf- and Pt-emitters and a miniature fission chamber were investigated thus revealing the limits of conventional radiography compared to neutron-radiography when highly active objects are investigated.

1. Introduction

In-core neutron detectors are exposed to the worst environment experienced by any commercial product. Besides the exposure to high temperature and pressure the detectors must also withstand intense gamma- and neutron irradiation. Therefore very high quality standards are applied to any component of these neutron flux sensors. The former USAEC has released a series of standards covering different parts of in-core neutron detectors[1–5]).

At present there are two types of in-core detectors which are used in boiling and pressurized water reactors, the miniature fission chamber and the self-powered neutron detector. Both detector types combine several advantages with some disadvantages. The fission chamber produces a prompt and strong detector signal resulting from gas multiplication effects in the filling gas (argon) under the applied voltage. The gas and the chamber's voltage are also the main disadvantages, because any loss of argon and the production of leakage currents must be prevented in order to get a true detector signal.

The self-powered neutron detectors show either a prompt (Co, Er, Hf, Pt) or a delayed (Rh, V) detector signal which is rather weak compared to the fission chamber. This is their main disadvantage. These detectors are however all solid detectors and therefore more rugged and cheaper in production.

Figs. 1 and 2 show schematic diagrams of both detector types. Their design, operation characteristics and in-core behaviour have been described in earlier papers[6–15]).

About the operation experience and long-term behaviour of in-core detectors only few data are published[16–18]) and detector producing companies are reluctant to release any realistic figures about failure rates. Failures have occurred both with fission chambers and self-powered neutron detectors which are partially caused by systematic errors in the detector design. Failed in-core detectors are subjected to extensive investigations usually by destructive methods in order to analyze the critical detector parts.

The critical detector parts with fission chambers are the electrode with the uranium coating, the metal-to-ceramic seal and the mineral insulated cable.

The fission chamber electrode is usually coated with

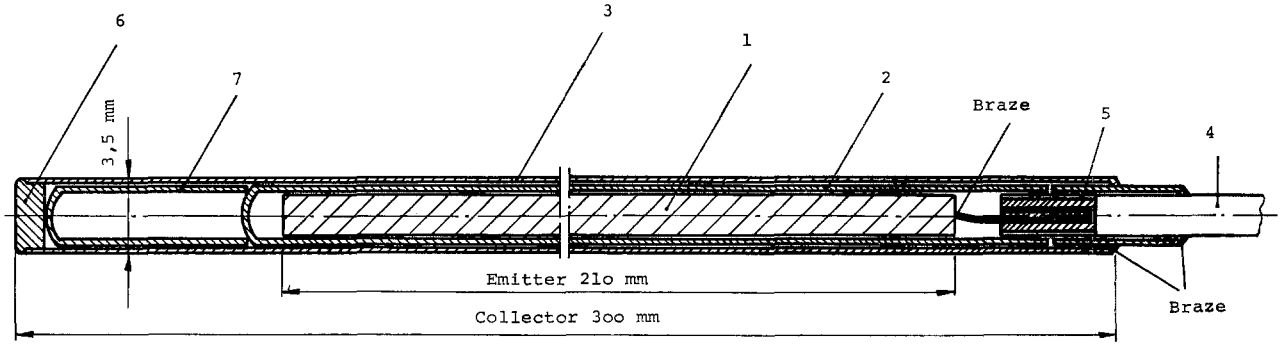

Fig. 1. Schematic diagram of a self-powered neutron detector. (1) Emitter – vanadium, rhodium, cobalt, platin, etc.; (2) insulator – aluminiumoxide; (3) collector – Inconel; (4) cable – mineral insulation, two cores, sheath of Inconel; (5) adapter – Inconel; (6) end plug – Inconel; (7) aluminiumoxide tube, thermal shield during brazing.

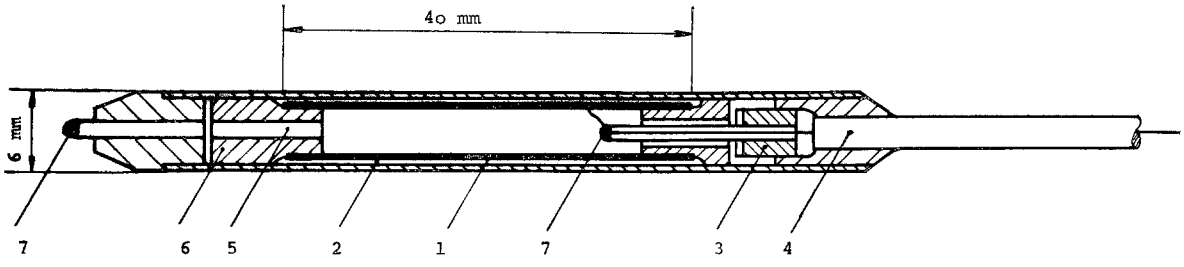

Fig. 2. Schematic diagram of a miniature in-core fission chamber. (1) Electrode – titanium coated with 93% enriched ^{235}U; (2) electrode gap – 0.2 mm, filled with argon; (3) metal–ceramic seal; (4) mineral insulated cable, one core, sheath of Inconel; (5) channel to fill the chamber with argon; (6) ceramic electrode alignment; (7) braze.

93% enriched ^{235}U by an electrolytical or evaporation process. The radiation stability of the uranium layer has been investigated in ref. 19. It was shown that the neutron sensitive layer performs distinctive transformations depending on the neutron dose. A destruction of the uranium layer will result in a voltage breakdown thus putting the fission chamber out of service.

In ref. 20 it was shown that a high temperature burden of the chamber will cause a uranium diffusion into the titanium electrode resulting in a sensitivity decrease of the fission chamber. The metal-to-ceramic seal must prevent a gas loss from the chamber's volume into the cable. Any gas loss would lead to a sensitivity decrease of the fission chamber[17, 21–23]).

Being all-solid detectors there is no problem with gas loss at self-powered neutron detectors. One critical part at these detector types is the braze of the emitter cable to the emitter. As mentioned before, the emitter material can differ according to the detector applications. The braze must be compatible with emitter and the cable material and should withstand temperatures exceeding the operation temperature by about 150°C. It should have a low neutron absorption cross section in order to prevent rapid transmutation during in-core exposure of the braze atoms into atoms with other mechanical, electrical or nuclear properties. A typical braze not recommended for self-powered neutron detectors with long-term exposure is Au which transmutes rapidly in Hg having a considerably higher vapor pressure at the operating temperature. This could easily result in a short circuit between emitter and collector.

The aim of the present work was the development and tests of a non-destructive examination method for highly active in-core neutron detectors. The described radiographic methods allow the investigation of certain detector parts as well as of the whole detector in order to detect any deterioration inside the detector. Neutron radiography of in-core detectors has the advantage to show especially the detector parts with high neutron cross section thus giving an image of what a reactor neutron actually sees during the detector operation. Further the transmutation of materials can be followed by this method.

2. Neutron radiographic methods

For neutron-radiographic inspections different methods exist[24]). The most extensively used neutron source for neutron radiography is the nuclear reactor. The method is based on the different attenuation coefficients of different elements or even isotopes of the same element for thermal neutrons. The main advantage of neutron radiography compared to X-ray radiography or gamma radiography is the fact that the attenuation coefficient for thermal neutrons often changes very much for neighbouring elements and is large for hydrogen and fissile material. A revised diagram comparing the linear absorption coefficients for neutrons ($v = 2200$ m/s) with these for X-rays (126 kV and 500 kV) of the elements and some important isotopes and compounds was prepared recently by Ross[39]). So neutron radiography is a method supplementing conventional radiography in several cases. The major application of neutron radiography is found in nuclear industry, especially for inspection and non-destructive evaluation of nuclear fuels[25–28]). Here for example measurements of enrichment, of burn-up, of dimensional changes, of hydrations in the cladding can be performed. The purpose of the present paper is to show the applicability of neutron radiography for the investigation of neutron detectors of power reactors.

To obtain a neutron-radiograph the transmission image of the object being available as differences in neutron intensities behind the object must be detected. For this purpose it is necessary to convert the neutron beam into ionizing radiation. For the final detection of the image photographic X-ray films are generally used.

Depending on the particular application two different methods using films can be applied (fig. 3). In the direct method the neutron converter and the film are in direct contact and exposed together to the neutron beam. The advantages of this method are shorter exposure times[24]) and – using Gd-converters – very good resolution. The optimum thickness of the most commonly used Gd-converter foil which emits conversion electrons after neutron capture is around 25 μm[29]). This thickness gives a typical screen unsharpness of 100 μm[30]), being defined as the distance between the 10% and the 90% density points on the micro-densitometer trace across the radiograph of a knife-edge. This definition – though very useful when a linear density curve in this region is assumed – gives a slightly too bad value. Berger[31]) gives for the resolution of a 12 μm thick Gd-foil behind the film a resolution of 9.6 μm being defined as the minimal discernable distance between the holes of a Gd test object.

The disadvantage of direct methods is the fact that they cannot be used for inspections of highly active objects and that the gamma-ray content of the beam used for the investigation must not exceed certain characteristic tolerable values[30]). Besides the use of metallic converters the possibility of the application of neutron scintillators exists. These scintillators give in general higher speeds but worse resolution.

For the investigation of radioactive specimens usually transfer techniques are used. The most general applied converter foil in this case is dysprosium metal. In this technique the foil is exposed to the neutron beam thus being activated and bearing the image of the object in the form of radioactivity differences. When this foil is put on a photographic film the radioactive image is transferred to the film by autoradiography. Dysprosium gives the shortest exposure times in transfer techniques and a resolution of 50 μm according to Berger[31]) or 200 μm using the definition of Hawkesworth[30]). The optimum foil thickness for the transfer techniques is about 200–250 μm[29]).

Recently the use of track-etch techniques was introduced to neutron-radiography[32, 33]). The image in this case is presented on a plastic foil which must be etched by a caustic solution after exposure. The main advantage of this method is also its insensitivity to gamma-rays and to light. For neutron-radiography in this case a converter must be used which transfers the neutrons into heavily ionizing radiation.

Recently cellulose nitrate films being coated with a boron compound became available[34]). The track-etch technique will be increasingly used in the next years because of its advantages compared to the transfer technique[35]), which are the shorter time until the image is available and no saturation effects, thus being applicable to very low intensity neutron-radiography.

For the imaging of small in-core detectors an enlargement of the neutron radiographs is necessary to obtain a good visibility of details. Therefore the resolution of the image has to be considered. The main influences on the resolution are the inherent resolution of the converter and of the film as mentioned above, and the geometric unsharpness as a result of the finite size of the neutron source. This geometric unsharpness is given by

$$U_g = \frac{Sl}{L},$$

where

S = size of source,
l = distance object – film,
L = distance source – object.

Based on arguments using communication theory

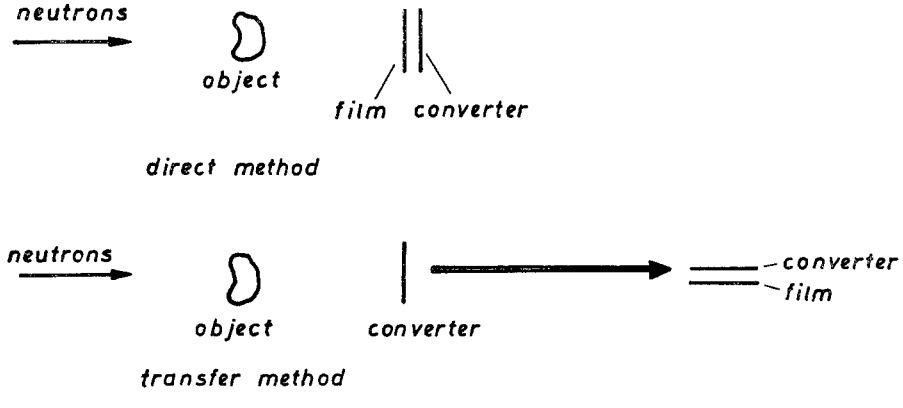

Fig. 3. Experimental arrangement for direct and indirect radiographic examinations.

Spiegler and Norman[36]) concluded that the total unsharpness is given as

$$U_t = (U_g^2 + U_{fc}^2)^{\frac{1}{2}},$$

with U_{fc} the film–converter unsharpness.

3. Experimental arrangement

The neutron radiographs were performed at the neutron-radiography facility at the TRIGA-Mark II reactor Vienna. This facility uses a divergent collima-

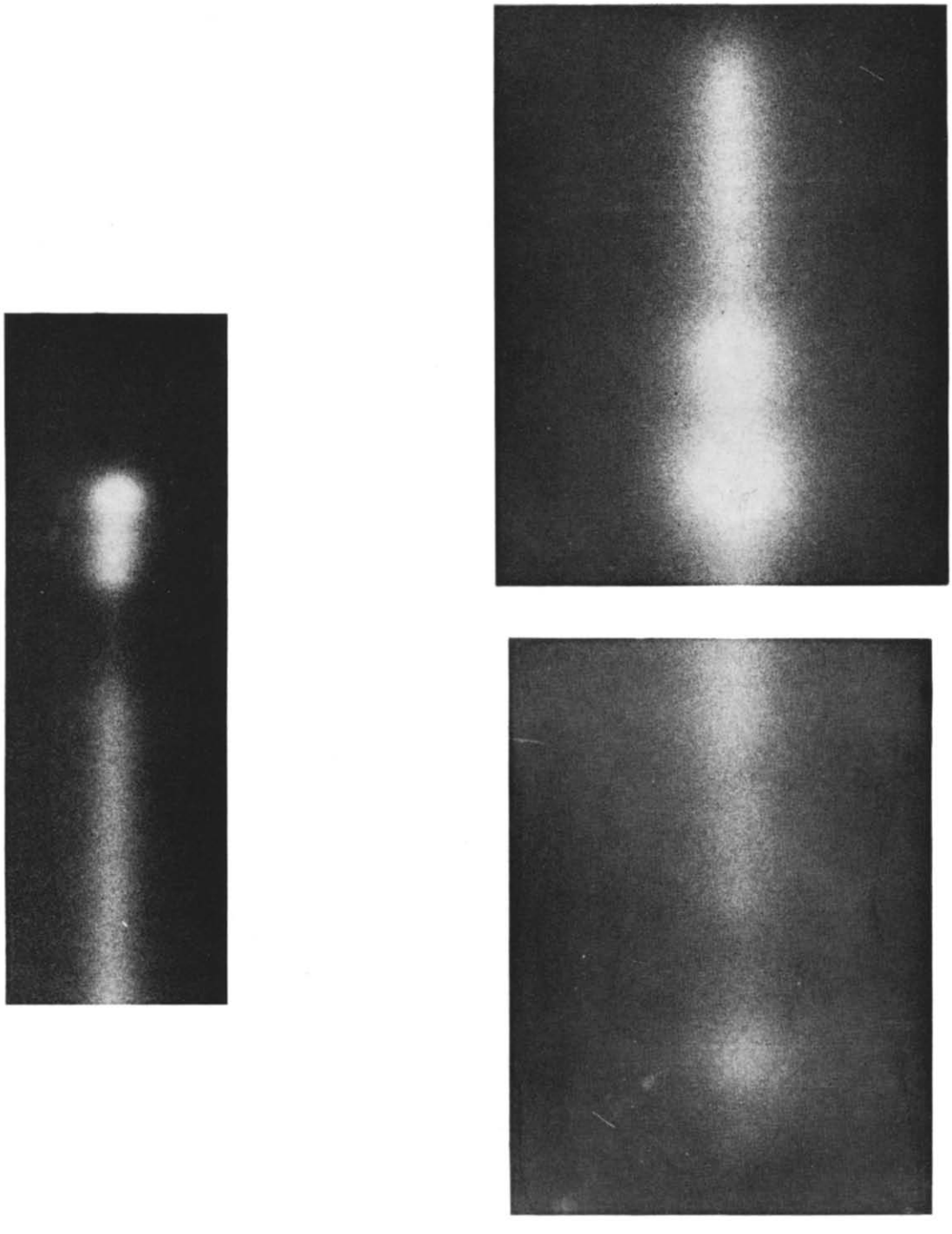

Fig. 4. Autoradiograph of a self-powered neutron detector with a Hf emitter (left) and an in-core fission chamber (right).

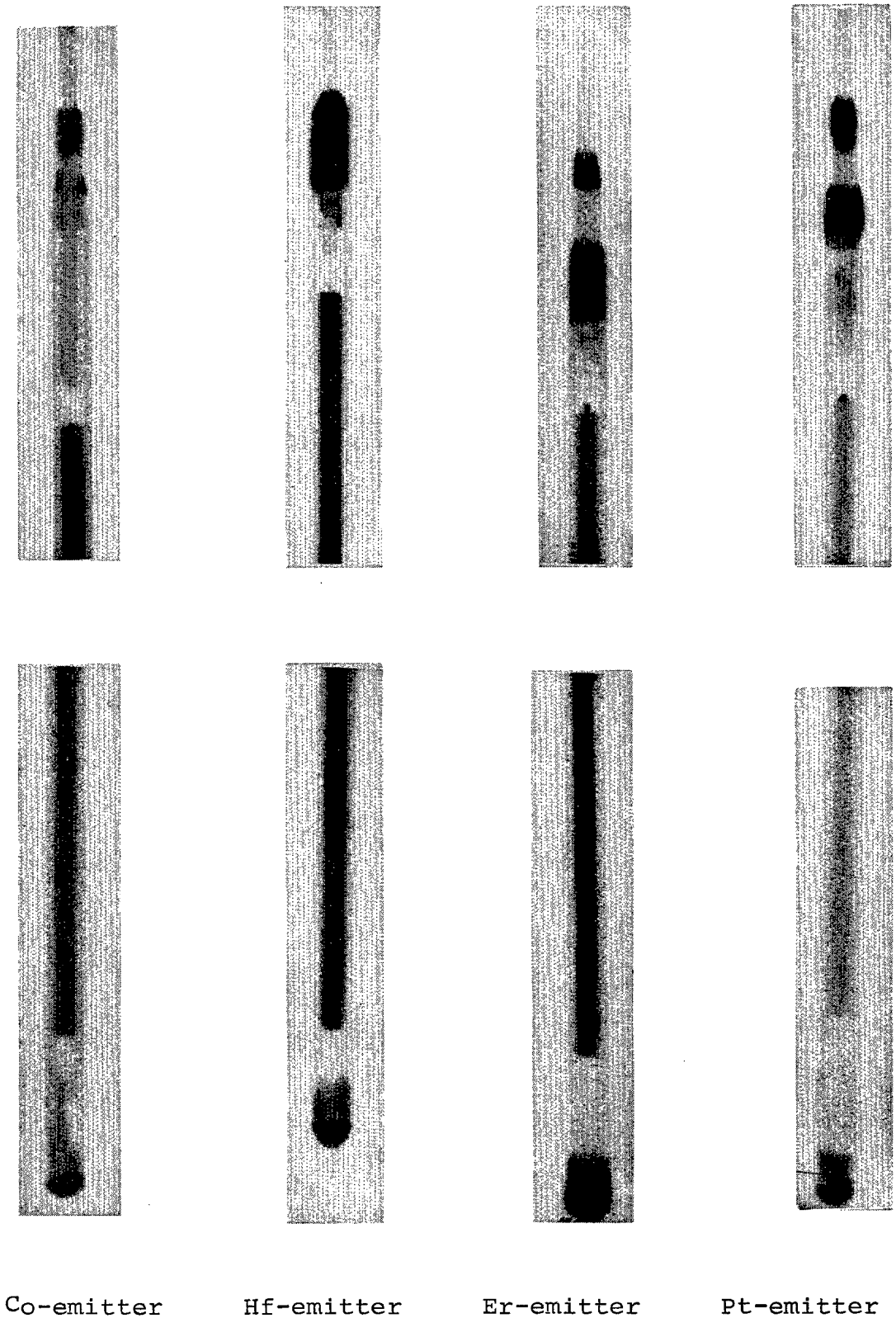

Fig. 5. Indirect neutron radiographs of self-powered neutron detectors with different emitter materials.

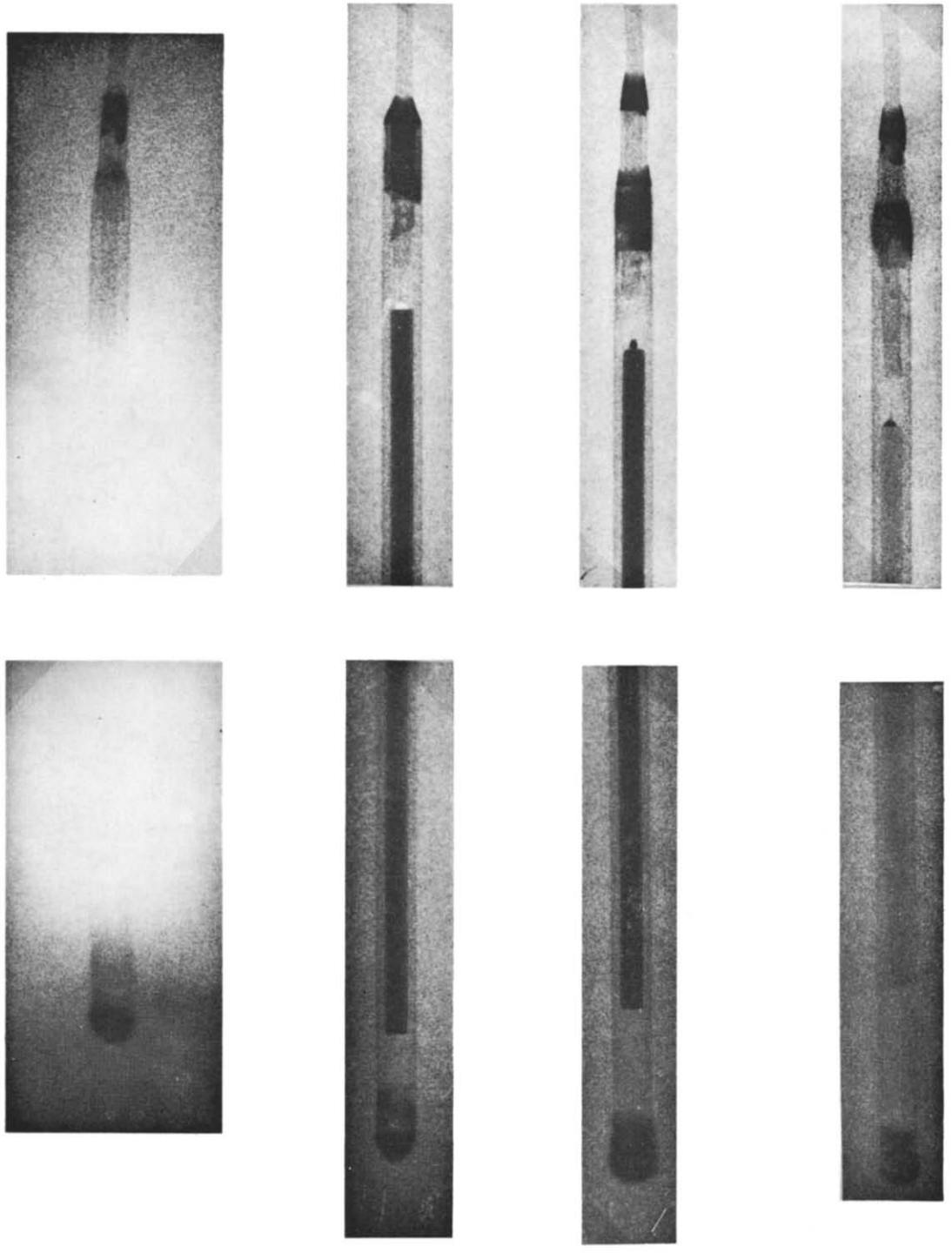

Fig. 6. Direct neutron radiographs of self-powered neutron detectors with different emitter materials.

tor[37]). The inner diameter and therefore the size of the neutron source was 15 mm, the distance from the source to the image plane was 3 m and the distance from the object to the image plane was 2 cm. The geometrical unsharpness was therefore approximately 0.1 mm. When the above mentioned resolution data of Berger[31]) are accepted, the local image unsharpness using Gd for the direct method is therefore approximately 0.1 mm. If Dy is used for the transfer method the unsharpness increases to about 0.11 mm. Taking half of the value of Hawkesworts[30]) is a reasonable assumption. In this case the total unsharpness is 0.11 mm for Gd and 0.14 mm for Dy. This seems to be for our case the more realistic assumption, since the sharpness of the images using Gd and Dy is quite different. These considerations are quite important, because they influence the enlargement of the radiographs which can be achieved sensibly. In the present work it is not sensible to produce enlargements greater than a factor of two of the originals.

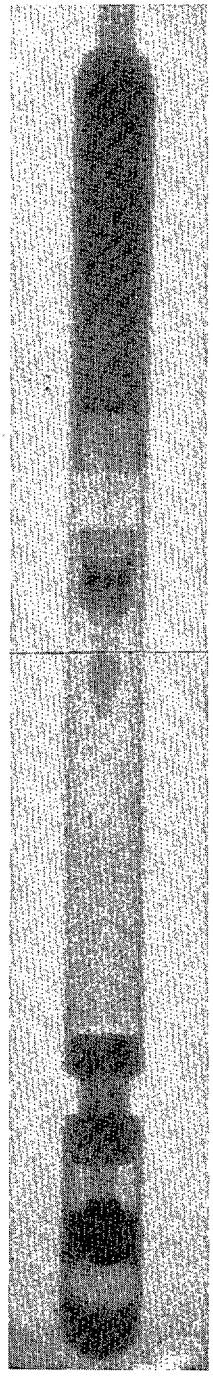

Fig. 7. Indirect neutron-radiography of an in-core fission chamber.

For the radiographs we used Osray TAT4 DW film, a Gd-converter of 0.025 mm thickness for the direct method and a Dy-converter of 0.100 mm thickness for the transfer method. The activation time of the Dy-converter was chosen to be 30 min in a thermal neutron flux of 6×10^5 n/cm^2 s. This allows to take more than one autoradiographic copy of the same activated foil for different purposes. The neutron radiographs were enlarged on hard photographic paper which was developed using a document developer for enhancing the contrast. For control purposes we made some autoradiographs of the self-powered neutron detectors also with the same X-ray film and an exposure time of 10 min. The exposure time for the direct method was 5 min.

4. Results

For the investigations four self-powered neutron detectors and one fission chamber were available. All detectors had been exposed earlier to a neutron fluence of about 1×10^{19} n/cm^2 at steady state operation and to about 20 reactor power pulses with a peak neutron flux of 1×10^{16} n/cm^2 s [13,14]). These irradiations resulted in a detector activation and in a gamma dose rate from 0.5 R/h (Pt-detector) to 3 R/h (Co-detector). Fig. 4 shows typical autoradiographs of the Hf-detector and of the fission chamber. The main activity of the Hf-detector is concentrated in the emitter and in the braze at the top of the detector, while at the fission chamber the position of the highly active uranium-coated electrode can be clearly distinguished.

Autoradiographs from an irradiated Co-detector are useless, because of the high detector activity, while the irradiated Pt-detector shows only a very weak picture due to its small activation.

Fig. 5 shows the result of the above mentioned

transfer method with converter foils. All photos reveal very clearly the emitter as well as the cable and the braze between emitter and cable. Dark spots at both ends of the detectors are due to a neutron-absorbing braze (Ag–Cd). As all four detectors were radiographed together at the same time with the same neutron beam slight differences in the resolution are due to beam inhomogeneities (i.e. lower part of the Pt-detector).

Fig. 6 shows a set of photos produced by direct neutron-radiography. It is evident that the resolution is slightly better for Hf-, Er- and Pt-detectors, because of their rather low activity. The Co-detector shows a very interesting superposition of neutron- and gamma-autoradiography, thus revealing the advantage of indirect radiographic methods for highly active subjects.

Fig. 7 shows an indirect neutron radiograph of the miniature in-core fission chamber. At the upper part of the photo the cable, the metal–ceramic seal and the electrode holder can be distinguished. The lower part of the picture shows especially the arrangement to fill the fission chamber with argon. The uranium-coated electrode extends from the metal–ceramic seal down to the braze with an actual length of about 40 mm. Just along the outer jacket a very fine dark line can be seen in fig. 7 which represents the 93% enriched uranium layer.

The small graduation in contrast perpendicular to the fission chamber axis in the lower photo is caused by a ceramic alignment which keeps the electrode centred in the fission chamber. A small channel can be observed along the axis which allows the gas to move freely between the active volume in the gas gap and the inactive volume inside the hollow electrode. The design and the testing of the same type of fission chamber as used for our investigations has been described in ref. 38.

In summary it can be concluded that neutron-radiography is a very valuable tool for the non-destructive investigation of highly radioactive components of the reactor instrumentation and safety system. Both methods, the direct and the indirect or transfer method can be applied according to the specific problem. The achieved resolution by neutron-radiography allows to investigate the physical integrity of such critical parts of in-core detectors like the emitter or the electrode, the cable, the cable-to-detector connection or the metal–ceramic seal. As a non-destructive method the investigations can also be performed during ex-core operation or they can be repeated after certain intervals to analyze burn-up dependent effects.

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