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Abstract

After some general remarks concerning the continuous monitoring of process solution of a reprocessing facility the instruments for measuring on-line the α -activity of such solutions are described. One of these is the OLAM (On Line Alpha Monitor) based on a cerium-activated glass scintillator. After its description the tests of the monitor concerning the minimum detectable amount, the contamination of the detector during use in a test loop in the laboratory and its decontamination are described and the results discussed (Variation of plutonium between normal background in a glove box till 80 mg/l corresponding to a variation of the signal of 0,9 cps to 1620 cps). It could be shown that organic solution with dispersed aqueous phase could be not so easily decontaminated. The background after this experiments was about 35 cps. Using middle active waste solution with a plutonium content between 8,9 mg/l and 74 mg/l the countrate was at each addition of plutonium too high and dropped after an interval of some hours to the 2,5 to 3-fold of the expected. The decontamination was only possible to a countrate corresponding to 10 mg/l. Alpha-spectra of the monitor and autoradiographs are given. The contamination could not be explained.

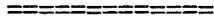
Zusammenfassung

Test des α -Monitors OLAM.

Nach einigen allgemeinen Bemerkungen die kontinuierliche Analyse von Prozesströmen in Wiederaufarbeitungsanlagen betreffend werden die Instrumente zur kontinuierlichen Messung der α -Aktivität beschrieben. Eines davon ist OLAM (On Line Alpha Monitor), der auf einem mit Cer aktivierten Glasszintillator basiert. Nach dessen Beschreibung werden die Tests diskutiert, die vor allem die untere Erfassungsgrenze, den Aufbau der Kontamination während des Betriebes und die Dekontaminierbarkeit untersuchten. Die Plutoniumkonzentration wurde zwischen dem normalen Untergrund in einer Handschuhbox und 80 mg/l variiert entsprechend einer Zählrate zwischen 0,9 cps und 1620 cps. Dabei zeigte

sich, daß die Dekontamination am schlechtesten in organischen Lösungen mit wässrigem Entrainment gelang. Die Zählrate nach diesen Dekontaminationen betrug 35 cps. Schließlich verwendeten wir eine mittelaktive Wastelösung, deren Plutoniumgehalt zwischen 8,9 mg/l und 74 mg/l variierte. Die Zählrate war zunächst immer zu hoch und fiel während einiger Stunden auf das 2,5- bis 3-fache des erwarteten Wertes. Die Dekontamination war nur bis zu einer Zählrate, die etwa 10 mg/l entsprach, möglich. Die Ursache der Kontamination konnte nicht eindeutig erklärt werden.

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1. INTRODUCTION

For the control of the process in a reprocessing facility the concentration of plutonium is, besides other constituents of the process streams, of great importance. This analysis can be done in an analytical laboratory. However the analytical results are only available with a more or less large delay and the information on this subject is a static one with lacks of information in between, depending of the sampling intervalls.

Another way to do this analysis is to use a suitable in-line instrument. By measuring a physical parameter of the solution the concentration can be calculated. The nature of the used physical effect is dependend

- on the constituent to be analyzed,
- on the concentration range of this constituent to be analyzed,
- on the other constituents in the process solution and
- on the concentration of these other constituents and their effect on the measuring device.

One of the methods to measure the concentration of plutonium is to measure the α -radiation of the process solution. When the main contribution to this effect is from plutonium, the concentration of plutonium can be calculated from the count rate, if the isotopic composition of the processed plutonium is known. This is mostly the case, because after the dilution of the fuel-element the isotopic composition of uranium and plutonium is measured by mass-spectrometry.

The principle construction of an α -monitor is based on the properties of the α -radiation and the reduction of the background of the β -, γ -radiation of the fission products in the solution. Due to the strong interaction of α -particles with matter their range in air is about 3,5 cm and in solutions about 50 μm . Therefore it is necessary for having a high counting efficiency to place the detector in contact to the measured process solution or in a small distance.

This is the basic criterium for the construction of the following types of instruments developed in the past:

- the weir cell [1 - 4],
- the rotating drum cell [5 - 13],
- the flow cell with a polymer window [14 - 18] and
- the cell with direct contact of the detector to the solution [1, 20 - 24].

2. DESCRIPTION OF THE OLAM

The OLAM (On Line Alpha-Monitor) is manufactured by the IRT Corporation, San Diego, U.S.A. The representation in Germany was till this year EG & G Instruments, München and has now changed to Münchner Apparatebau Kimmel GmbH & CO KG, Ottobrunn.

The principle of this detector by using alpha scintillating glass has first been published by U.L.Upton [25]. C.E.Huck and J.D.Lodge [26] described in 1964 test results of this monitor. The used scintillating glass Vycor 7913 is manufactured by Corning Glass Works; it is 254 μm thick. It is reported, that there was no alterations in the counting yield observable after contacting the glass to nitric acid of 0,25M, 1M and 4M over a test period of 40 days at 323 K. Also a fluoride content up to 0,01M sodium fluoride complexed with 0,16M aluminium nitrate in the solution had no effect on the counting efficiency of the detector contacting it for a period of 3 months at 303 K.

The cerium activated high-silica glass has been refined to attain maximum α -sensitivity, while minimizing β - and γ -sensitivity. The glass is grinded after the cerium layer has been diffused [29]. The thickness of the sensitive cerium layer is about 40 μm . The difference in shape of scintillation peaks induced by α - and β -radiation is described by T.Gozani [28]. He describes also other optimization problems [27]. The specifications of the instrument are listed in table 1 as they are given by IRT. In fig.1 the cross section of the instrument is shown. Opposite the detector a shaped deflector constricts and directs the flow of liquid across the face of the sensor in a thin sheet with a maximum thickness of approximately 0.12 - 0.50 mm. With respect to the operating conditions of such an instrument in a reprocessing plant two difficulties may occur during normal operation:

- (a) The small thickness of this liquid layer leads to the risk of blockage by small amounts of solid material. Such material is

Dimensions:	19cm × 26cm
Maximum sensitivity for low β-solutions:	10 ⁻⁸ Ci/l of α-activity in a 15 minute measurement
Sensitive area diameter:	2,54 cm
Maximum fission product activity:	30 Ci/l
Flow rate:	1 l/min
Sensitive area diameter:	0,2 - 6 cm (optional)
Weight:	70 lbs

Table 1 Specification of OLAM (Paper from IRT Corporation).

present in many process streams and is due to incomplete dissolved fuel elements and to incomplete separation of this solids. On the other hand during normal operation of such a plant solids are builded up.

- (b) The direct contact of the solution to the detector surface will give a high sensitivity. On the other hand this may result in deposition of solids or other liquid phases (water in organics, organics in water) on the detector surface. If this other phase has no or only a small concentration of α-emitters compared to the bulk liquid these deposits will decrease the count rate. The conclusion would be that the concentration of α-radiators in the solution decreases. If this deposits consists of plutonium or other α-emitting material the count rate would be increased as it is the case if the concentration of plutonium is raised in the bulk phase. By these effects the use of the instrument would be limited.

C.E.Huck and J.D.Lodge [26] report the installation of this instrument on the purex L-8 sampler in the Hanford Purex Plant to monitor the combined XAW-XSW waste stream losses of Pu-239. In a test loop in the laboratory the count rates were linear with the plutonium concentration varying from 100 cpm at 10⁻⁴ g/l to 830000 cpm at

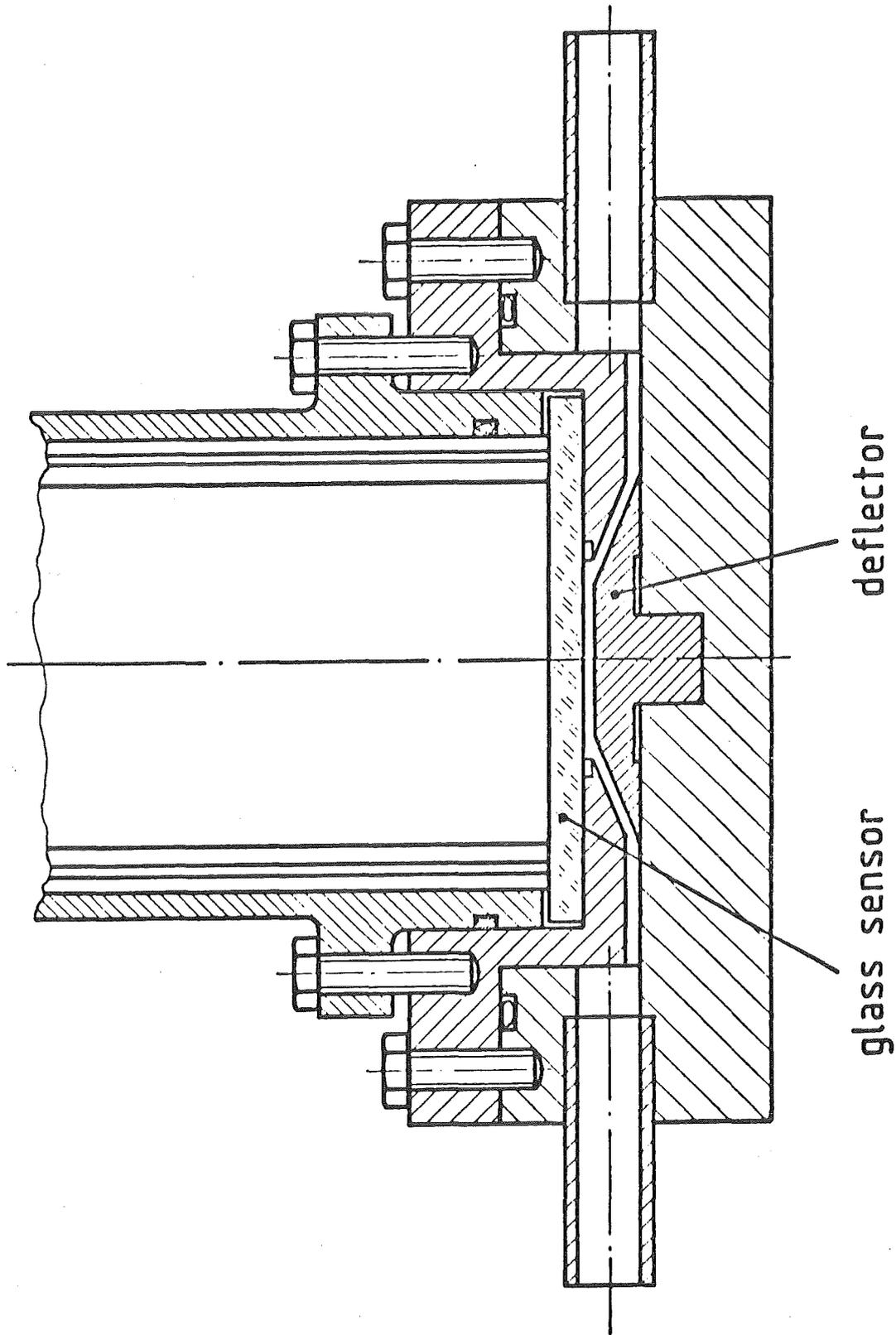


Fig.1: Cross section of OLAM [26].

1 g/l. Glass and cell decontamination performances have been good.

K.J.Hofstetter et al. describe in 1978 tests before the application of this monitor to process streams in a nuclear fuel reprocessing plant [30]. The lines at the Barnwell Nuclear Fuel Plant where the installation was planned are listed in table 2.

Monitor Point	Conc. g Pu/l	β -, γ -Act. dps/ml	Stream Composition
1CU Stream	$2 \cdot 10^{-4}$	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U
1BP Stream	4,82	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U
2AW Stream	$4 \cdot 10^{-3}$	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U
2BW Stream	$1 \cdot 10^{-4}$	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U
1SW Stream	$2 \cdot 10^{-4}$	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U
1SF Stream	$2 \cdot 10^{-4}$	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U
3AW Stream	$2 \cdot 10^{-4}$	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U
3BW Stream	$2 \cdot 10^{-4}$	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U
POR Stream	$2 \cdot 10^{-4}$	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U
3PD KO Pot	$2 \cdot 10^{-4}$	$2,6 \cdot 10^5$	0,17M HNO ₃ , 0,28M U

Table 2 Streams at BNFP to be monitored with OLAMS.

The OLAM had been subjected to a testing program in a laboratory system at BNFP to establish sensitivity, accuracy, reliability, long term stability and useful concentration range (10^{-4} to 3 g/l). Also decontamination procedures have been studied. The full-width at half-maximum resolution of the alpha peak was determined to be 30%. The circulated solution was 2,6M HNO₃, the flow rate was 125 ml/min, driven by gravity. The liquid system was closed due to avoid evaporation of the liquid, resulting in a concentration effect. The study with this system lasted 3-4 weeks.

After each plutonium solution a flush solution containing 125 ml of 2,6M HNO_3 was added to the system after emptying it and has been circulated for at least 24 hours. This process of flushing the instrument was continued till the concentration of plutonium in the flushing solution was less than 1.10^{-5} g/l. In general five flushes were required. The spectra of a characteristic flush serie are shown in fig.2. The accumulation time was 22 hours. Hofstetter et al. explain the peak in the fourth spectrum by the effect that the plutonium is "plating out" on the detector surface. One may wonder that this effect, which can constrict the applicability of the instrument, has not been investigated more intensive and thoroughly.

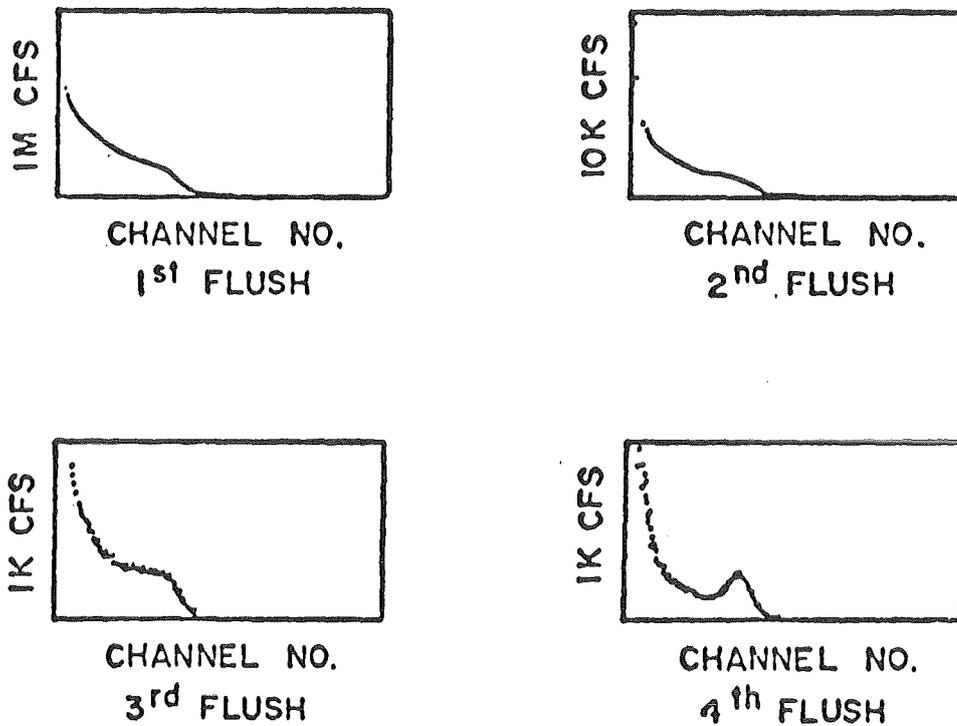


Fig.2 Effect of consecutive flushing on spectra [30].

The use of Sr-90 simulating the different β -, γ -activity in plant solutions showed effect on the countrate. The used specific β -activity was 10^3 to 10^6 dpm/ml.

The short term stability was better than 1% relative standard deviation. For plutonium concentrations between 0,001 and 0,1 g/l the one sigma relative standard deviation was 11,8%. The amount of plutonium that plated out on the detector was about 5-6 times the background rate.

3. DESCRIPTION OF THE TEST OPERATIONS

Starting from the above characterized situation of knowledge out of the literature we had some doubt that there will not be interference of deposits on the detector surface to the data specified by IRT Corporation. Especially the decontamination procedure must be tested carefully. For this test we used an installation schematically shown in fig.3. The solution to be circulated is transported by a pump to a higher level reservoir. In the system there is also a possibility to draw samples for analysis.

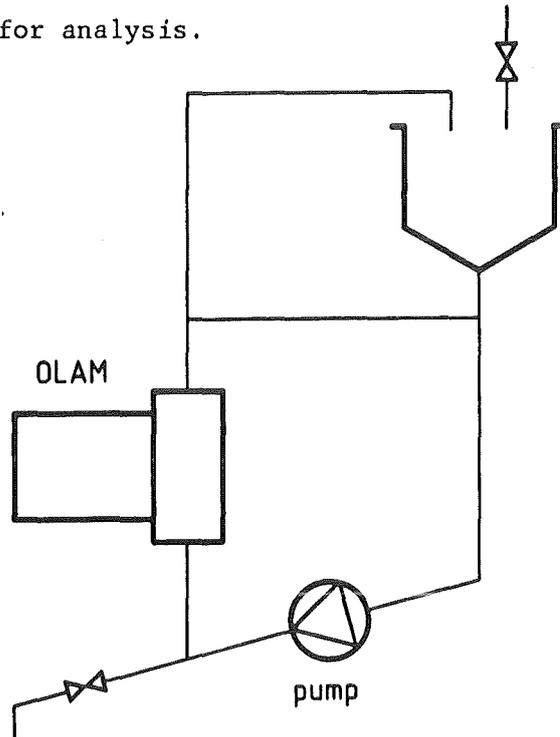


Fig.3 Scheme of the installation for testing the OLAM.

The initial count rate was 0,9 cps corresponding to a minimum detectable amount of plutonium of 0,13 g Pu/l using plutonium of light water reactor with a medium burn-up. In a first step we measured the sensitivity of the OLAM using pure aqueous plutonium-solutions with concentrations between 1 - 10 mg/l. Fig. 4 shows a typical spectrum recorded on a multichannel-analyzer using 80 channels. The peak area starting at channel 20 is used as a measure of the α -activity of the solution. This limit avoids possible interference due to β - and γ -activity. The calibration of the OLAM is influenced by the choice of

this limit. We found in good agreement with other authors the sensitivity of 20 cps for a plutonium concentration of 1 mg/l.

After this step we contaminated the detector by using a solution of 103mg Pu/l in 1,5M HNO_3 . This gave a countrate of 2060 cps. After 6 rinses with 1,5M HNO_3 we found a final countrate of 1,3 cps. After additional rinses this countrate could not be changed. The spectrum found after this rinsing steps is shown in fig. 5. The shape of the curve had changed and had a maximum at the channels 35 - 40. This is an indication that there was a deposit of plutonium direct on the surface of the detector.

To simulate the composition of process solutions we treated then the detector with an aqueous plutonium-solution with 1,2% dispersed organics (30% TBP in kerosene). The concentration was 0,1M HNO_3 and 101 mgPu/l. We found a countrate of 2060 cps, which is in good agreement with the countrate measured before. After 6 rinses with the aqueous solvent, free of organics the countrate dropped to 40 cps.

Then we used a similar solution but with higher acid content (2,7M HNO_3) and 83mg Pu/l. The countrate was 1620 cps, which is also in agreement with the results measured before. After 6 rinses with 1,5 M HNO_3 the final countrate was 35 cps.

We investigated also the situation if the OLAM is installed in organic process streams. We used 30% TBP in n-alkane, wherein 2% 1,5M HNO_3 has been dispersed. The plutonium concentration was 81 mgPu/l. The countrate was 1620 cps and in good agreement to the other results. After 4 rinses with the solvent the countrate could not be lowered than 350 cps. Probably there have been droplets of nitric acid deposited on the detector surface carrying this amount of plutonium. After 4 additional rinses with 1,5 M HNO_3 the countrate dropped to 35 cps.

Finally we tested the OLAM with a process solution of the PUREX-process. We used MAW-solution as it is produced by our experimental installation MILLI. This solution carried 8,9 mg Pu/l.

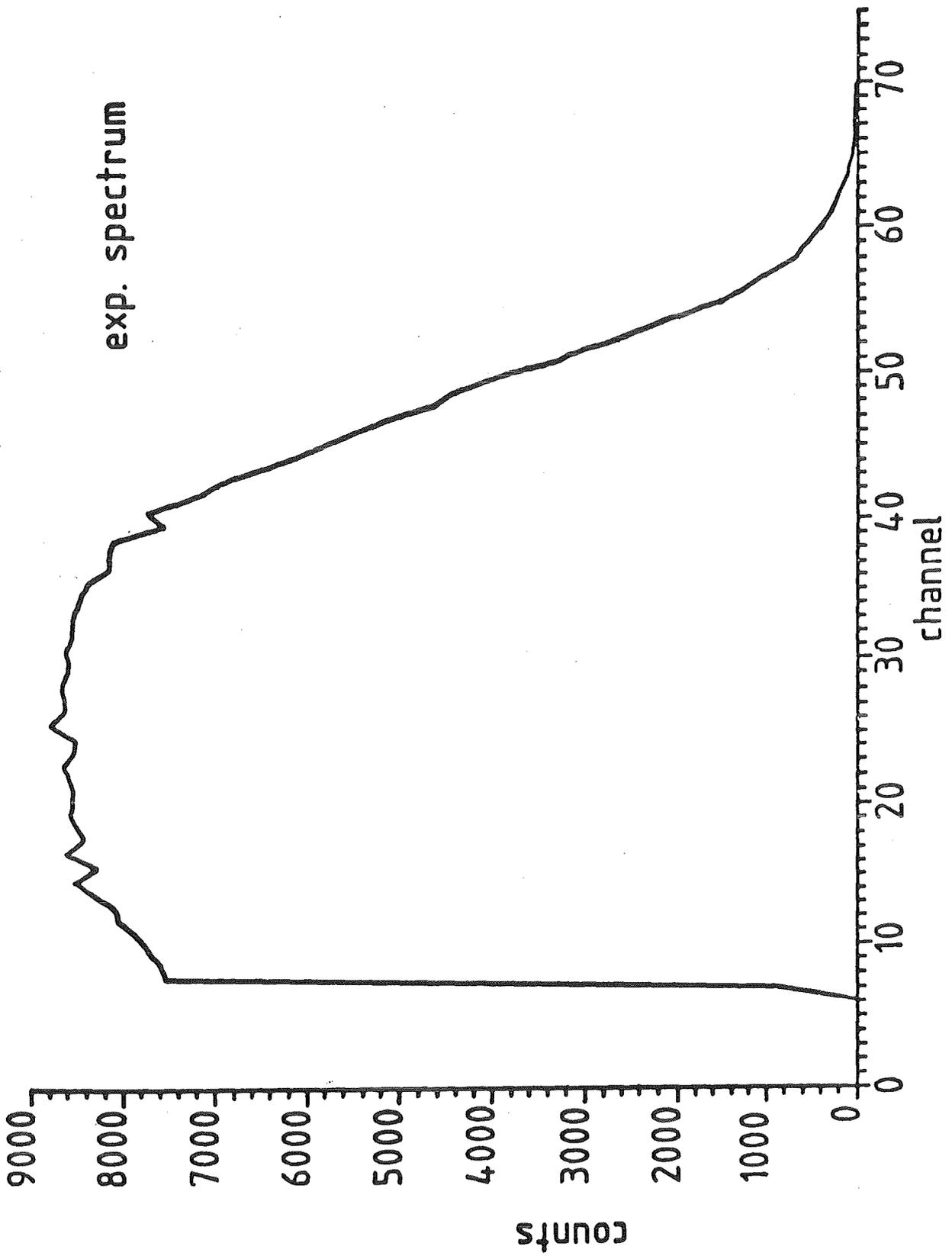


Fig.4: Normal α -spectrum obtained by the OLAM.

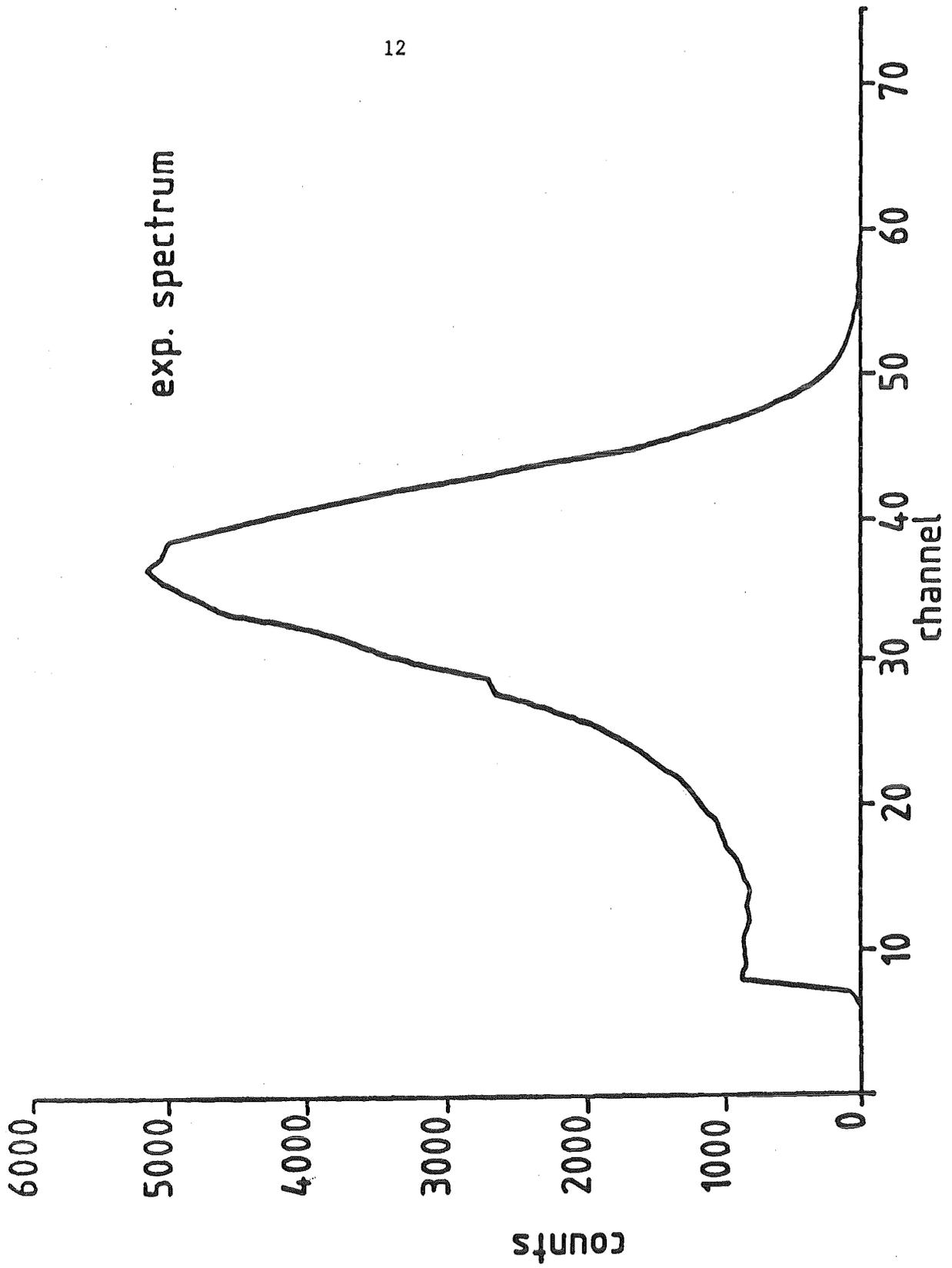


Fig.5: Alpha-spectrum of the OLAM after decontamination procedure indicating contamination on the surface of the detector.

The activities of the fission products have been:

Ce-144: 0,12mCi/l

Cs-134: 0,36mCi/l

Cs-137: 0,98mCi/l

Ru-106: 0,44mCi/l

Eu-154: 0,04mCi/l

The countrate was 800 cps, approximately constant over a period of 4 days. This value is roughly by a factor of 4 higher than the expected countrate (180 cps), based on the same isotopic composition of the plutonium. But the used MAW was produced by reprocessing a large variety of fuel elements and therefore the isotopic composition could differ from the used plutonium solution. The isotopic composition has not been analyzed. Then we increased the plutonium concentration to 34 mg Pu/l by adding 1 ml of high-concentrated Pu-solution direct into the test-loop. The initial countrate was 6300 cps, dropping within 18 hours to 1600 cps, which is too high by a factor of 2 compared with the other results (expected count rate 680 cps). After increasing the plutonium concentration to 59 mg Pu/l the initial countrate was 3400 cps, decreasing within 3 hours to 2100 cps, which is also too high by a factor of 2 (expected count rate 1180 cps). This result could be interpreted as a large memory-effect of the instrument. When passing the instrument first, the solution had not been mixed. The monitor observed a high plutonium concentration, which contaminated the surface of the detector. After contacting the detector with the mixed solution for hours the contamination had been reduced. When this procedure has been repeated to increase the plutonium concentration, the same effect has been observed. After increasing the plutonium concentration to 74 mg/l the countrate increased to 4000 cps, decreasing within 24 hours to 2400 cps, which is also too high (expected count rate 1480 cps).

At this point we tried to decontaminate the detector. After six rinses with 1,5M HNO₃ we reached a stationary countrate of about 200 cps. We could not improve the decontamination by using other solvents as 8M HNO₃, RBS, hydrogen superoxide. The large remaining

count rate correspond to about 10 mg/l plutonium.

We opened then the measuring cell to inspect the detectors surface. Whether visually nor by microscope we could find any deposit. The count rate of the detector could be verified by a surface barrier detector. The spectrum of the contamination is similar to that shown in fig.2.

We investigated then the distribution of deposited plutonium on the surface of the detector. For that purpose we used an autoradiographic method (Film: Kodak no screen film x-ray). The results are shown in fig.6. They show that the activity is concentrated in very small spots. The reason for that could be that the surface is changed by something in this region. One of the possibilities could be, that the surface has been treated by mechanically polishing. When this surface comes into contact with higher concentrated nitric acid, there are disturbances in the structure of the glass on the surface, which are enlarged by the acid. In this microcavities the plutonium is deposited. But why this is the case with MAW-solution and not with pure plutonium solution could not be clarified. Also the publications report no tests of the OLAM in reprocessing solutions as they came out of an operating plant.

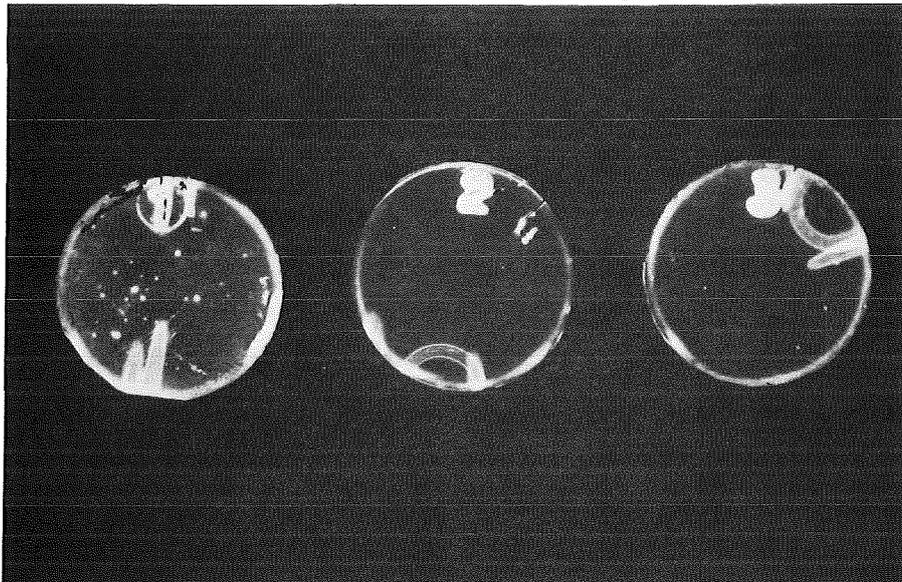


Fig.6 Autoradiographs of the surface of the detector in OLAM.

(Film: Kodak no screen film x-ray, exposure time: 100 hours).

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