THE PACKAGE FOR THE RECOVERY OF A DAMAGED SOURCE

A. Orsini (1), A.M. Spano (2), G. Gualdrini (3), and S. Rizzo (4)

(1) ENEA-AMB-TEIN, 00060 Roma, Italy
(2) CISAM, 56010 S. Piero a Grado (PI), Italy
(3) ENEA-AMB-IRP, 40126 Bologna, Italy
(4) NUCLECO S.p.A., 00060 Roma, Italy

SUMMARY

The paper describes the repacking activities performed on a radioisotopes generator prototype, charged on 1976 with a Sr 90 source and found damaged during the preparation for transport to an interim storage on 1994. The radsource was surrounded by metallic depleted uranium blocks, which, due to the high temperature and to the loss of inert gas after the long storage, had begun to oxidise, changing its structure with a big volume increase. The damage produced obliged to remove the source from the generator, modify the container and arrange a new package for a safe transport of the source.

INTRODUCTION

Concerning the research programs for the utilization of the energy produced by radioisotopes and following the growing interest in the 70's on the exploitation of submarine oil resources, SNAM PROGETTI Research Laboratories - ENI Group, together with CNEN (now ENEA), developped the design of a radioisotopic thermoelectric 5 W generator for submarine appliances up to 200 m depth.The generator could also be used as long-lived electrical power supply for instrumentation and equipment in inaccessible sites, as radio and TV repeaters, radar and radio-beacons for navigation, amplifiers for submarine cables, sonar buoys, etc. .A radioisotopic generator must be particularly stable and safe, and for this reason a prototype was carried out, in 1973, choosing a 20.000 Ci Sr90 radioactive source, due to long half-decay time (28 years), high specific power (0,95 W/gr) and a very stable chemical physical state (Strontium Titanate), in addition to a good commercial availability and low cost. As biologic protection, a continuous metallic depleted Uranium shielding was choosen around the rad source, to have an optimization of the weight and volume of the assembly. The real hot source consisted of the Uranium shielding, containing the radsource in thermal touch with it, so the maintenance of the system could be easier, avoiding the need of radsource handling. The lack of thermal efficiency of the energy conversion was balanced by restricting the thermal exchange out of the surface strictly necessary for conversion and providing a thermal insulation all around. The external container should comply with IAEA regulations for transport of radioactive material as type B package, so mechanical test was performed at University Scalabatraio Center in Pisa. To withstand the free drop test conditions, an antishock frame to apply outside of the vessel was provided, while the system design was strong and rigid enough to withstand the other

test conditions. The task of assembling the whole system was committed on 1976 to Agipnucleare and CAMEN (now CISAM), a military research institute, already cooperating at the site of G. GALILEI experimental Reactor in S. Piero a Grado - Pisa, due to some interesting military appliances of the radioisotopic generator. At reactor site there were human and technological resources available, as shielded hot cells and remote handling equipment, allowing the safe execution of the assembling operations and of the research program following. After the complete packaging of the radioisotopic generator prototype in the hot cell, the research program was stopped in 1980, due to a decreased interest on the industrial appliances of radioisotope generators. Then the assembly was stored in the reactor test channel.
On 1994, cause to the developping of the decommissioning program of G. GALILEI Reactor, Eniricerche, receiving the problem by inheritance from Snamprogetti Research Laboratories, Agipnucleare being disappeared, charged Nucleco with the transport of the generator to Casaccia ENEA Center for interim term storage.

PROBLEM OUTLINING

After a preliminary survey on the generator, an high radiation dose rate on the upper part of the generator (maximum 6,40 mSv/h) was checked and it was decided to open the package. The reason of the lack of shielding should be verified and the electrical transducer and the thermoelectric module should be removed to make the package safely storable in an interim term storage. The sight disclosed at the opening was striking and surely unforeseen: all the metallic blocks (flanges and sheets with 2 cm thickness), securely bolted and restrained, were warped and the bolts (12 mm dia.) seemed torn up.
What's happened ?
Cause to the presence of black powder, it was inferred that the metallic depleted uranium surrounding the radsource, due to the high temperature and to the lost of inert gas after the long storage, has been beginning to oxidize changing its structure with a big volume increase causing the effect of a rising cake. After the opening and the removal of some metallic parts, the oxidization became faster and it was necessary to decide a repacking procedure. The only information available on the generator was as follows:

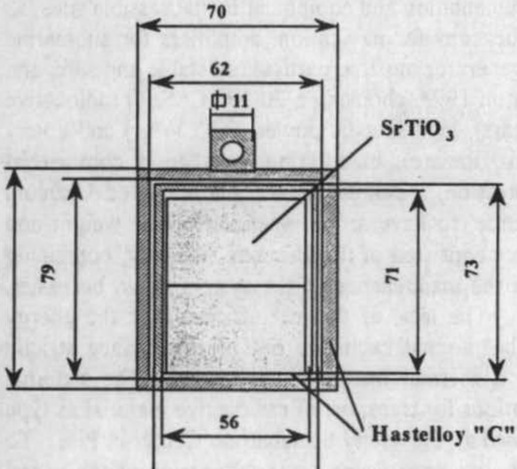

Fig. 1 - Sr 90 Source

Radioactive source (see fig. 1)

Radioisotope:	Sr 90
Chemical form:	$SrTiO_3$ sintherized
Containment:	Double Hastelloy cap
Activity:	19118 Ci, measured on 27/1/1977
Decay:	Beta emission (E=0,54 MeV), Y90 (E=2,2 eV)Zr90
Radiation:	X Bremsstrahlung - E= 0-2,2 MeV

Container (see fig. 2)
The Sr 90 source capsule was contained in a stainless steel basket (cont. 3), including some depleted Uranium blocks completely sorrounding the source and, in the upper conical part, the housing for the thermoelectric module and the metallic block ("cold finger"), necessary for the thermal exchange to outside. The cold finger was fixed to the top flange by means of four bolts, discovered torn up at the vessel opening.
Between the stainless steel outside vessel (cont. 1) and the basket it was inserted an heat insulator material (expanded rock wool 85 mm thick).

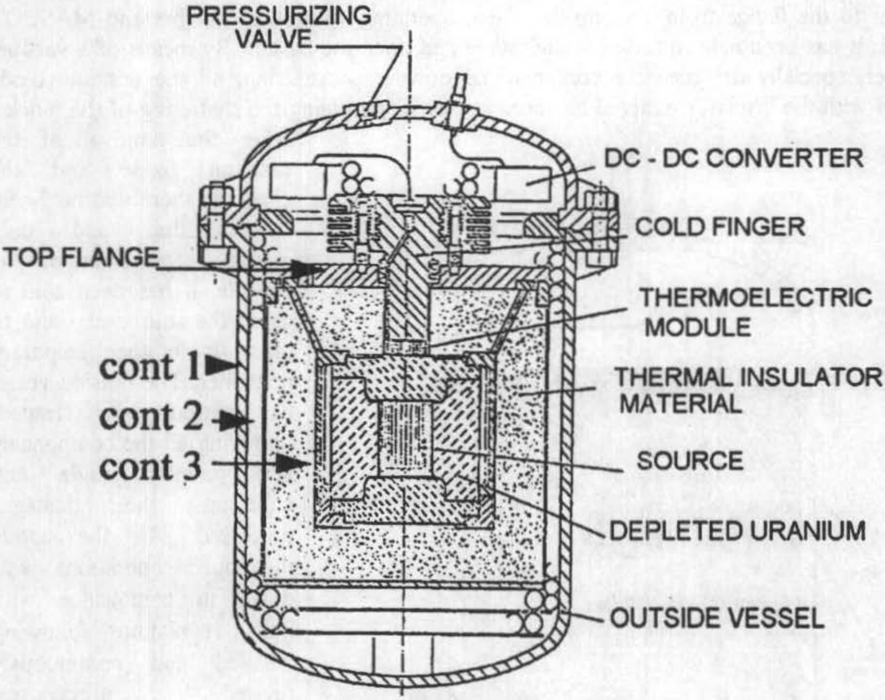

Fig. 2 - The original container

Repacking procedure
Because no Sr 90 contamination was detected, it was decided that the source capsule should be not damaged and it had be able to proceed as follows:

- remove the source capsule from the generator, by means of the original equipment, designed for source charging, and place it in a temporary container;
- modify the container, removing the uranium blocks and the insulator material, to insert lead blocks as new shielding;
- check the surface source capsule contamination and condition and transfer the source in the modified container;
- introduce the repacked generator in a "special arrangement" transport container to ENEA Casaccia Center interim term storage.

All the operations could be done remotely in the Reactor hot cell, by means of the MASCOT robot, equipped with cameras.

SOURCE REMOVAL

Checking very carefully the generator conditions without detailed drawings, the situation was more complicated than foreseen. In fact the top flange had been lifted up for about 30 mm, due to the uninterrupted uranium oxidization effect, in a such abnormal way to cause the getting stuck of the flange. Then it was decided to design and manufacture a special tool to extract the flange, taking advantage of the presence of two 12 mm diameter screwed holes on the flange itself. Remotely operating the special extractor, it has been possible to lift up the depleted uranium-source block only for about 100 mm, before the breaking of the tool due to extreme mechanical stress. Another careful and detailed visual inspection by means of the MASCOT cameras showed the presence of socket head screws fixing the basket to the flange from bottomside. Then, operating with small mirrors and MASCOT hands, it has been able to unscrew the flange and open the basket. By means of a vacuum cleaner, specially designed for contaminated powder transferring, all the uranium oxide mixed with the insulator material has been removed, providing the shattering of the blocks. After the removal of the uranium oxide and the checking there was no Sr 90 contamination and only plastic deformation of the capsule, it has been able to catch the source cap and to place it in the temporary container. The outside vessel has been completely cleaned, removing all the components and powder inside and checking the damages produced. All the above mentioned operations was done, in compliance with italian regulations, following detailed and continuously revised procedures, previously discussed and approved by the G. GALILEI Reactor Safety Committee and by ANPA (Safety and Radiation Protection Italian Authority).

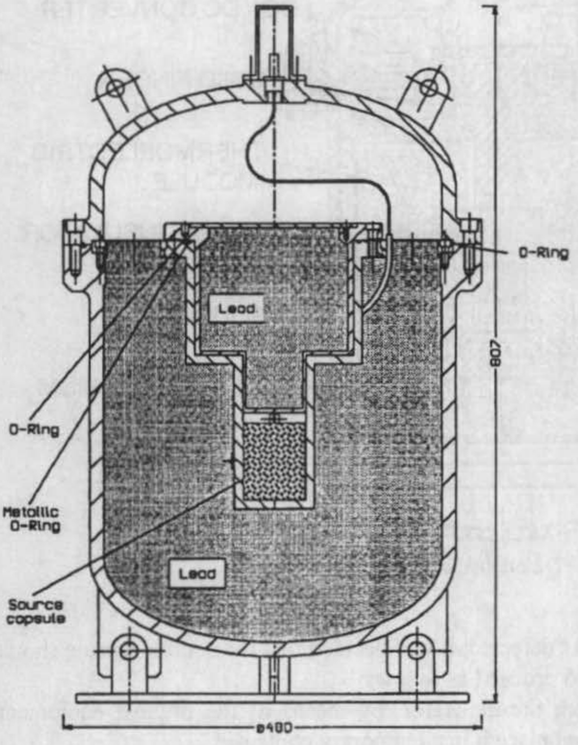

Fig. 3 - The modified container

CONTAINER MODIFICATION

The new container design criteria (see fig.3), based on the reusing of the outside vessel, have been fixed as follows:

- source radiation protection. Lead has been choosen as shielding material;
- checking of the thermal exchange to outside, considering the source thermal power (85,7 W);

- source containment system in normal and accident conditions

Shielding calculations.

The $SrTiO_3$ 466 Tbq source that is located in the internal container is characterised by the ^{90}Sr nuclide nuclear decay. The β decay (Emax=0.54 MeV) leads to ^{90}Y that again decays to stable ^{90}Zr through a β disintegration (Emax=2.2 MeV). Therefore the process that has to be analysed to design the shields is the photon bremsstrahlung production within the source. The bremsstrahlung spectrum is characterised by a continuous behaviour with energy ranging from 0 to 2.2 MeV /1/ and was described by means of seven discrete lines (see Table 1) .

Energy (MeV)	photon emission rate (photons/W s)
0.25	1.705 E11
0.5	6.612 E10
0.8	1.958E10
1.1	6.103E9
1.4	1.650E9
1.7	3.027E8
2.0	2.082E7

Tab. 1 - Sr 90 bremsstrahlung spectrum

Employed methodologies

As far as the employed transport codes to design the flask shield are concerned, both deterministic (one dimensional SN multigroup) and stocastic (three dimensional Monte Carlo) methods were used. The two approaches allow to obtain results in good agreement if the treated geometry can be assumed as monodimensional and the multigroup cross section library well represents the changement of the photon spectrum along the investigated shield thickness. A preliminary dose equivalent evaluation outside the radial shield was performed using the authomated sequence of the codes NITAWL/XSDRN-PM/XSDOSE (SAS1) provided by the modular code SCALE (version 4) /2,3,4,5,6/. The sequence solves the transport equation through the SN method. The sequence is described in Figure 4. The flask was modelled through coaxial infinite cylinders (see Figure 5) .

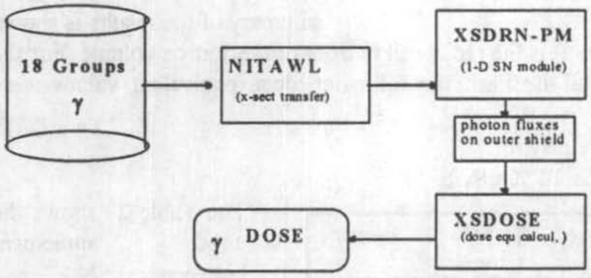

Fig. 4 - The SCALE authomated sequence SAS1

To provide a more accurate set of dose equivalent data outside the flask, including a complete mapping of the top and bottom shields, the MCNP Monte Carlo code /7/ (version 4.a) was employed. The code is provided with a very powerful and flexible geometry package as well as a point-wise cross section library where photon interactions are treated in great detail (e.g. form factors for coherent and incoherent scattering, fluorescent emission as well as bremsstrahlung photon production are included)/8/.

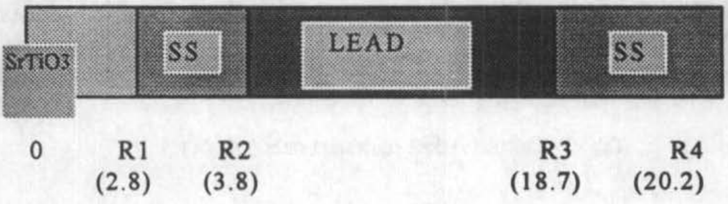

Fig. 5 - 1-D SAS1 flask cylindrical modelling (dimensions in cm)

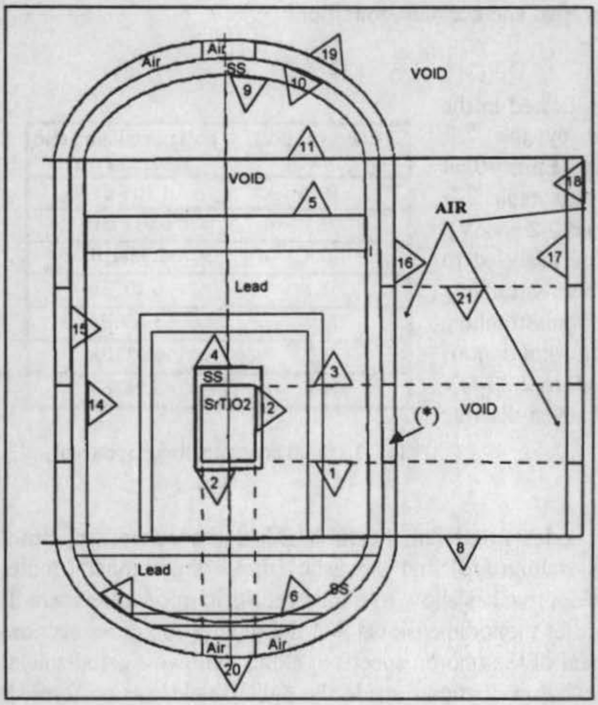

Fig. 6 - MCNP geometry model

The real geometry was modified only slightly (see Figure 6) . The source was supposed to be uniformly distributed within the internal cylindrical cavity. The investigated response was the air kerma in various radial volumes and also in volume shells placed at the top and bottom of the flask. This allowed a consistent comparison with experimental data obtained with instruments calibrated in terms of air kerma free in air. Tracklength estimators (volume tallies) as well as ring detectors (next event estimators) were undependently employed, obtaining results in good agreement. To optimise the top and bottom dose equivalent calculations spatial and directional biasing of the source was applied. In Table 2 a summary of the results is shown . The space portion (*) (see Figure 6) is the radial cell in front of the source volume. For the top and bottom outer surfaces of the flask the following dose equivalent values were obtained:

top dose equivalent rate: 9 ± 6 % μSv/h

bottom dose equivalent rate: 33 ± 14 % μSv/h

	Side wall	1 meter
a) SCALE-4 - cylinders	146	1,5
b) MCNP (volume tally) outside cell (*)Fig. 7 max dose equivalent rate	143 ± 1,9%	3,8 ± 4,0%
c) MCNP (ring detector) outside cell (*)Fig. 7 max dose equivalent rate	134 ± 3,0%	4 ± 4,0%
d) MCNP (volume estimator) avg dose equivalent rate	56 ± 2,2%	3,6 ± 3,0%

Tab . 2 - Calculated dose equivalent rates (μSv/h)

The Table 2 shows the good agreement between SN and Montecarlo values except for the value at 1 m. from the flask. This can be ascribed to the low ratio H/D (H=source height, D=source to detector distance) that causes the lack of the XSDOSE module for this condition.

Thermal exchange calculations

Different limit conditions have been evaluated. First of all the real situation has been evaluated, considering 1 mm air gap between different metallic materials. In this situation the maximum temperature in the lead blocks is 45°C. However a temperature sensor inside the container shielding has been foreseen to check the temperature during the beginning temporary phases and later on. An air layer inside the lead shielding with different thicknesses has been evaluated to check the limit conditions of air bubble inside the lead fusion so great to cause the lead melting. This has been calculated for a layer thickness of 31 mm. An increasing outside accidental temperature has been evaluated to check the limit conditions for lead melting inside the shielding. The result of this calculation is an outside temperature of 300 °C. This temperature will be taken into account for the final design of the transport overpack to get the "special arrangement" approval.

Containment system

Although the capsule resulted with no visible cracks, a checkable air seal system has been foreseen between the stainless steel plug and the source pit, consisting of an elastomeric and metallic O-rings. A second air seal system, not checkable, has been foreseen on the outside vessel. All the bolts have been verified for a vertical acceleration of 100 g.

The container modification has been performed complying to a Quality Control Plan, based on the following "hold points":

- Outside vessel air leakage control. This has been done before the lead casting, by keeping the inside vessel at 3 ata for 10 minutes checking any pressure decrease.
- Source pit - plug coupling air leakage control. This has been done in the same way, by keeping at a pressure of 3 ata the volume between the two O-rings.
- Lead casting homogeneity control. This has been done by checking the weight of container before and after lead casting.
- After the charging of the Sr 90 capsule inside the modified container, measuring of the outside vessel surface temperature (43°C with external 23°).
- After the charging of the Sr 90 capsule inside the modified container, measuring of the dose rate on 24 points on the external surface (see fig. 7) and at 1 meter (see fig. 8).
- After the charging of the Sr 90 capsule inside the modified container source pit - plug coupling air leakage control.

CONCLUSIONS

The damage of radioisotope generator was completely unforeseen since it was not planned to have the Sr90 and the uranium shielding at high temperature in ambient atmosphere where the chemical reactivity of uranium is too high: on the other hand the efficiency of thermoelectric generator increases with the temperature but the reactivity of metallic uranium much more. Uranium shielding must be carefully analysed in particular when the package is undergone to the fire test. The removal of Sr90 source was a hard job worsened by the lack of any drawings: after 20 years there was only one operator remembering by heart some details of the generator. All repacking operations, dealt by different partners with the supervision of the Competent Authority, have taken more than two years. Now the repacked source is stored always in the former nuclear reactor site, waiting for a future transport in "special arrangement" condition.

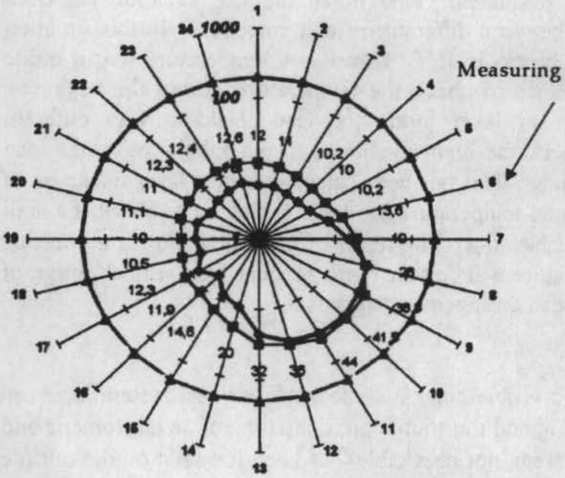

Fig. 7 - Surface dose rate (μSv/h)

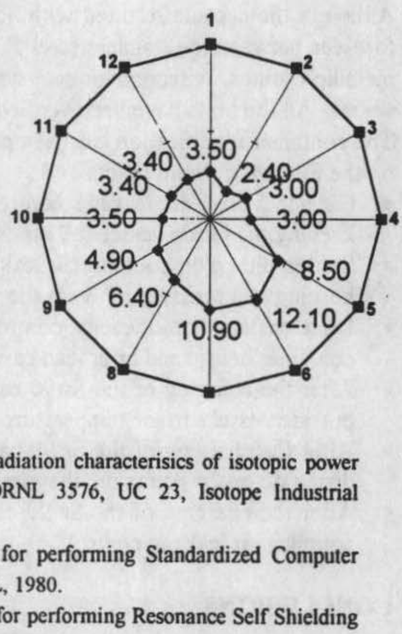

Fig. 8 - 1 meter dose rate (μSv/h)

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