CRITICALITY CONTROL DURING THE DISMANTLING OF A URANIUM CONVERSION PLANT

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ABSTRACT

Within the Commissariat à l'Energie Atomique, in the Cadarache Research Center in southern France, the production at the Enriched Uranium Treatment Workshops started in 1965 and ended in 1995. The dismantling is in progress and will last until 2006. The decommissioning is planned in 2007.
Since the authorized enrichment in ^{235}U was 10% in some parts of the plant, and unlimited in others, the equipment and procedures were designed for criticality control during the operating period.
Despite the best previous removing of the uranium in the inner parts of the equipment, evaluation of the mass of remaining fissile material by in site gamma spectrometry measurement shows that the safety of the "clean up" operations requires specific criticality control procedures, this mass being higher than the safe mass.
The chosen method is therefore based on the mapping of fissile material in the contaminated parts of the equipment and on the respect of particular rules set for meeting the criticality control standards through mass control. The process equipment is partitioned in separated campaign, and for each campaign the equipment dismantling is conducted with a precise traceability of the pieces, from the equipment to the drum of waste, and the best final evaluation of the mass of fissile material in the drum.
The first results show that the mass of uranium found in the dismantled equipment is less than the previous evaluation, and they enable us to confirm that the criticality was safely controlled during the operations.
The mass of fissile material remaining in the equipment can be then carefully calculated, when it is lower than the minimal critical mass, and on the basis of a safety analyzis, we will be free of any constraints regarding criticality control, this allowing to make procedures easier, and to speed up the operations.

ENRICHED URANIUM TREATMENT WORKSHOPS

General description – goals

The Enriched Uranium Treatment Workshops (in French ATUE, standing for Ateliers de Traitement de l'Uranium Enrichi) is one of the 19 nuclear facilities within the research center of CEA-Cadarache, located in south east of France, near the junction of the rivers Durance and Verdon, 50 km from Marseille.
The ATUE was built in the early sixties and was operated from April 1964 to 1995 ; at this time, CEA refocused on R&D activities.
Upstream in the fuel cycle, the ATUE was mainly devoted to develop and operate chemical processes for uranium purification and conversion into a sinterable oxide powder. Sintering and fuel elements were to be made in other facilities.

The processes

Two main conversion processes were used. The wet process was used for the recovering and the recycling of uranium from scraps by the steps of nitric acid dissolution, solvent purification by exchange columns, ammonium precipitation and hydrogen reduction. The dry conversion process converted UF_6 coming from isotopic enrichment into UO_2.
The uranium oxide powder produced was used in other facilities for nuclear fuel fabrication, destined mainly for research reactors and fertile cover of fast breeder nuclear reactors.
The processes are designed with cylindrical or conical vessels, with volume range from some liters to 2 m^3, circuits of metallic pipes of small diameter (average 1 inch) and numerous different types of pumps and valves, without any radiation shielding.
Material is mainly stainless steel or special alloys as Inconel or Monel.

Dry process technology – workshop B

The main equipment (vertical vessel and rotary kiln) of the UF_6 dry conversion process is electrically heated and thermally insulated.
Gaseous UF_6 and water steam are injected for an immediate hydrolysis ; the UO_2F_2 powder falls on a screw leading it inside the rotary kiln, in wich counter-current flows of water steam and hydrogen are injected, leading to UO_2.
The UO_2 powder is removed on line and transferred towards a mixing unit in order to make homogeneous batches.
The criticality safety of the dry process is controlled by the temperature monitoring, set at the proper level to avoid any condensation to warrant the standard of H/U ratio, and the mass of the batch of UO_2 powder as well as its humidity.

Wet process technology – workshop C and D

The wet process of uranium conversion has several steps allowing to adapt the treatment at the nature of the entering product and at the impurity level aimed.
The nitric dissolution is a batch process, the unclean uranium oxide powder is dissolved in concentrated nitric acid. The uranyle nitrate is then purified in pulsed extraction columns by a solvent (mixture of tributylphosphate and kerosene), and re-extraction is done by nitric acid.
The precipitation is a continue process, the uranyle nitrate solution is poured in a vessel and mixed under agitation with the ammonia solution. Under gravity, the ammonium diuranate fall on a rotating drum-filter on which the « yellow cake » is collected and leaded into a rotary kiln in which it is dried and the oxidized into the U_3O_8 powder form.
U_3O_8 is then processed in a rotary kiln under an hydrogen flow for reduction into UO_2 sinterable powder form.
The criticality safety of the wet process uses the different approaches : mass limitation and geometrical design in the dissolution unit, geometrical design for the columns, the precipitation and the reduction unit, and mass as well as humidity for the homogenization.

General program of dismantling

After the end of the production in 1995, the Final Shut Down operations were mainly aimed to the removal of the nuclear material and its transfer to other facilities, the emptying of the

chemical reactants, the mapping of the contamination, and the securing of the electrical circuits.
To the Final Shut Down phase was added a « Clean Up » consisting of the removal of the peripheral equipment (such as reactant and gas circuits, insulation materials and electrical circuits) and the removal of the part of the process equipment requiring no special heavy dismantling technologies (the pipes under a diameter of 80 mm, the valves and other small elements). An important goal is to decrease the quantity of fissile material still present in the equipment.
This Clean Up, which began in 2001, was performed under clearance of the French Nuclear Safety Authority.
The decree allowing the dismantling of the whole plant is expected in 2003, and the final decommissioning in four phases will be finished in 2007.
The first phase is the dismantling of the remaining process equipment, their framework, and the effluents pipes and tanks.
Ventilation and electrical circuits will be dismantled during the second phase, replaced meanwhile by site devices.
Final clean up of the empty buildings, by concrete scrapping, and examination of the radiological mapping will be done within the two last phases, followed by a final report.

CRITICALITY CONTROL

During the production period the uranium conversion processes were either geometrical safe, or were operated under strong administrative control to prevent any criticality accident. The ATUE facility was and is still equipped with a Criticality Accident Detection Device, which covered mainly the wet process area and the radioactive effluents circuits.
Dismantling operations will modify the global geometry, so it is necessary to control the criticality risk by a specific ^{235}U mass management.
The evaluation of ^{235}U quantities involved in the dismantling operations has been carried out for the whole facility inventory. It is mainly based on in site gamma spectrometry measurements, modeling, and radiation flow rate measurements associated to transfer functions.

In site gamma spectrometry

Gamma spectrometry enables to identify radio isotopes present in the facility by their gamma ray emission characteristics and to quantify their activity.
The activity and the fissile material mass are checked out by means of measurements and modeling.
The activity of a simple shaped piece (cylindrical column, flat filter,…) with an homogeneous contamination, is determinated with the following procedure:

- gauge the spectrometry measurement system with a perfectly known point source at a distance "d",
- measure the radioactive piece to be analyzed at the same distance "d",
- compute the "equivalent activity" which is the activity value of a point source that would give the same measurement value,
- compute the "measurement sensitivity" for a point source at a distance "d" and for the piece geometry at the same distance,
- transform the equivalent activity in real activity with: Areal = Aeq * Sref /Sgeom
    - *Areal : contaminated piece activity*

- *Aeq : Point source equivalent activity*
- *Sref : measurement system sensitivity for point source*
- *Sgeom : measurement system sensitivity for the contaminated piece geometry*

Radiation flow rate measured on the equipment

The radiation flow rate measurement with radioprotection tool equipped with gamma probe corresponds to a global gamma rays measurement. A method based on the extrapolation of gamma spectrometry results presented above makes this measurement representative for ^{235}U, whose characteristic spectrum is the 186 keV ray.

Computed from the gamma spectrometry results, a transfer function, linking the gamma ray measurements to the geometry, is worked out for each simple shape. The MIP10 measurement value is combined with the proper transfer function and the geometrical and physical characteristics in order to get its activity.

Each part of the process equipment is split in simple geometrical shapes and a computation code whose entry data are the flow rate values calculates the fissile material mass for the whole process.

Fissile material mapping – some figures

With a total of 45 in site spectrometry measurements we explored all the different enrichments used in the ATUE, as well as a representative number of elements leading to the definition of transfer functions characterizing 5 simple shapes and 7 material types.
The complete fissile material mapping of the whole process required about 3 months of measurement, modeling and computing for a team of 3 specialized people.

The mapping enabled us to have the evaluation of the total mass of ^{235}U in each workshop:

Workshop B 350 g, Workshop C 480 g, Workshop D 980 g

SAFETY DURING THE OPERATIONS

Minimal and safe critical mass

In the ATUE facility, the ^{235}U enrichment can vary up to 93,5 % but the spectrum is homogenous for each workshop.
To take into account the possibility of "double loading" the ^{235}U mass involved has to be lower than 61 % of safe critical mass of the considered workshop, as shown in table 1.

Table 1. Safe critical mass in the workshops

Workshop	Reference mixture	Enrichment	Safe critical mass	Safe critical mass with double loading
B	sintered UO_2-H2O	10 %	840 g	500 g
C	UO_2F_2 –H_2O	93,5 %	570 g	350 g
D	UO_2F_2 -H_2O	10%	900 g	550 g

Consequently, the values given by the mapping in workshops C and D, higher than the safe critical mass, confirmed the necessity to take into account the criticality control during the operations. Staying in a consistent frame, the workshop B campaigns were conducted the same way, even if the evaluation (350 g) is lower than the safe mass (500 g).

Dismantling campaigns

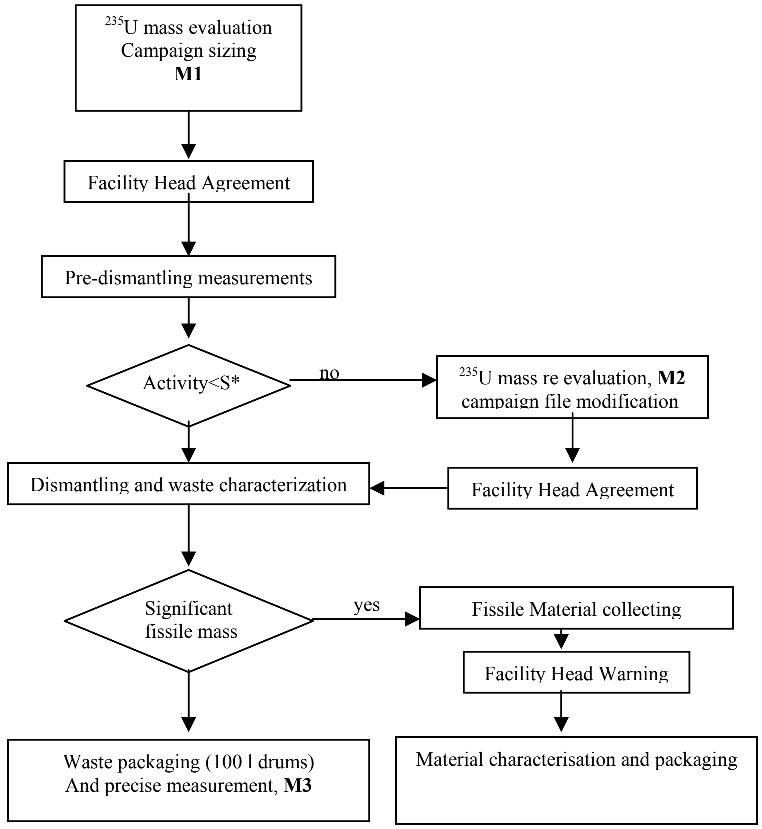
In order to limit the ^{235}U mass involved in the dismantling operations under the safe mass given above, we organized the operations in a program of separate campaigns.
The process equipment was partitioned with the help of the blueprints and the fissile material mapping. Each dismantling campaign, designed for an homogeneous part of the process, was limited in ^{235}U mass below the safe mass and to one workshop. Only one campaign could be carried on at a time in one workshop.

Detailed dismantling operations

In the first phase of the dismantling - the removal of the processs peripherals - 13 campaigns have been designed .
Before each campaign, the Head of the ATUE facility gives his agreement, according to the study file presenting the list of equipment pieces to be deposed, with their activity, and the total ^{235}U mass involved.

During the operations, each dismantled piece is controlled both radiologically (radiation flow) to check the coherence with ^{235}U mass evaluation and when possible visually to check the absence of any significant amount of fissile material ; this latter would be recovered and packed in a safe geometry container.

Campaign Flowchart



(*) radiation flow leading to the activity used for campaign sizing

The track of each piece to be dismantled, from its first location to the final drum is written down in a check list for track record. This check list includes marking off, emptiness checking, cleaning, measuring, waste drum identification.

The two first steps of ^{235}U mass evaluation M1 and M2 are made in the frame of the criticality control ; M1 is the primary global evaluation through on site measurement, and M2 is a reevaluation, using the transfer function, made in case of hot spot discovering during fine measurements by in site gamma spectrometry.
The value M3 is worked out after a gamma spectrometry measurement of each waste drum. M3 contents the mass of the fissile material recovered during the dismantling.

The figures 1 and 2 show the part of the process before and after dismantling the peripheral equipment within the campaign F1.

Fig.1. process before campaign F1

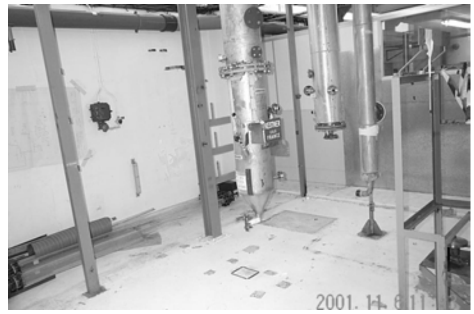


Fig.2. process after campaign F1

Lessons learned

Dismantling operations are now in progress. The results of the first ten campaigns out of 13, are summarized in the Table 2.

Table 2. Results of the first ten campaigns

Campaign number	**Workshop**	**^{235}U safe critical mass (g)**	**^{235}U mass global evaluation M1 (g)**	**^{235}U mass reevaluation M2 (g)**	**Final measurement M3 (g)**
F1	D	550	32,9	32,9	0,3
F2	D	550	36,1	51,0	13,8
F3	D	550	22,6	25,0	6,4
F4	D	550	214,3	237,9	16,6
F5	D	550	191,1	220,9	75,9
F6	D	550	53,3	55,6	9,9
Total workshop D			*550,3*	*623,3*	*122,9*
F7	B	500	34,4	48,9	10,3
F8	B	500	54,1	110,5	18,2
F9	B	500	124,4	342,2	24,1
F10	B	500	59,9	140,4	42,8
Total workshop B			*272,8*	*642,0*	*95,4*
Total			823,1	1265,3	218,3

In each campaign, the list of equipment pieces to be dismantled was chosen with regard to their location in the building, and not with the concern of reaching an amount of mass as close as possible to the safe critical mass ; this explains the relatively small values of M1.

M2 values are very close to the M1 values in the workshop D campaigns, the workshop B M2 values are about twice of M1. In addition, the ^{235}U mass recovered during operation in workshop B (5300 g of material including 26 g ^{235}U from 4,2 tons of waste) is relatively big compared to mass recovered in the workshop D (580 g of material including 12,7 g ^{235}U for 11,2 tons of waste).

That gives a good evaluation of the quality of the production line cleaning: the wet process was cleaned with acid solution, hence the average contamination was lower, and homogeneous ; on the contrary, the dry cleaning of UF6 conversion process was not so efficient.

In all cases, the final value M3, which is more precise, is lower than the evaluation given by radiation flow and transfer function, the average ratio being 0,22 for the wet process equipment, and 0,35 for the dry process equipment. This shows that despite the important measurement error there was a comfortable margin to the critical safe mass.

The sub-total of M1 give the ^{235}U mass involved: 550 g in workshop D and 272,8 g in workshop B ; that means the remaining masses are respectively 430 g and 77 g, by difference with the global evaluated masses given earlier. Consequently, after using the ratio for the two processes, we can assume that only about 95 g ^{235}U in workshop D and 27 g ^{235}U in workshop B remain.
This values are small enough compared to the safe critical masses to enable us to make the procedures of criticality control easier. Therefore we prepared a safety analysis based on the results of the clean up campaigns and the nature of the operations in order to stop and dismantle the Criticality Accident Detection Device.
However, we will keep by precaution the same organization with campaigns we described before during the dismantling of the remaining process equipment, radioactive effluents circuits and ventilation pipes.

CONCLUSION

The dismantling operations since 2001 in progress in the CEA-ATUE facility were designed to manage criticality risk through procedures based on traceability and mass control.
Prior to the dismantling operations, the mapping of the ^{235}U contamination of the equipment was drawn up, based on the computation of the radiation flow rate with specific transfer functions derived from on site gamma spectrometry.
The first results of the final waste characterization show that the selected method is reliable. Joined to the trackrecord of the dismantled pieces, the ^{235}U estimation is accurate enough to leave a comfortable safety margin, since there is 3 to 5 times less ^{235}U in the drums of waste than estimated in the equipment.
At this point of the dismantling, the first experience shows that the fissile material remaining in the process is small enough to carry on the operations with lower constraints. This will be particularly acted by the dismantling of the Criticality Accident Detection Device of the facility.