

BUILDING STRATEGIES FOR RESTORATION OF CONTAMINATED AREAS AFTER A NUCLEAR ACCIDENT : IMPLEMENTATION OF RELATED STUDIES AND FIRST ORIENTATIONS

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INTRODUCTION

The widespread contamination resulting from the Chernobyl accident has lead IPSN to implement a program, aiming at building strategies for environmental restoration after an accident potentially occurring in a reactor or a fuel cycle facility. Establishing optimized strategies requires to consider the incidence of all remediation operations, from the collection of the contaminated material to the final storage of the wastes issued from decontamination. Most of the decontamination strategies actually consist in shifting the polluting substances from the environment to a controlled and safer system, i.e. a storage or a repository, which result in the overall decrease of hazards undergone by the various groups of the population exposed. The usefulness of the treatment of contaminated material against the direct storage of the latter will therefore strongly depend on the ability of applied technologies to transform the material in an innocuous product or reduce significantly the volumes of wastes to store, so that doses and costs are globally spared. The incidence of storage will therefore be of great importance in determining optimized strategies. Another important aspect of remediation relates to the feasibility of the industrial implementation of technologies. Some considerations on the above issues are given in the present paper. As a starting point, only rural environments contaminated after a major Nuclear Power Plant accident were considered.

HYPOTHESES ON ACCIDENTAL SITUATION

Making prognosis on the radiological situations resulting from a potential accident is a difficult task considering the numerous parameters involved, such as the nature of radioactive releases, the meteorological conditions and the environmental components that may be touched. A good example of the variability of contamination distribution was given in the Chernobyl area where rainfalls during the period of the accident occasioned a deposition of radionuclides in spots and where the levels of activity were found to vary strongly with the distance according to the physico-chemical nature of the nuclides released, i.e. released in gaseous forms or included in fuel particles (1). The hypotheses presented below must therefore be considered only as rough estimations of what could be an « homogenous » radiological situation in rural environments after a severe accident in a French Nuclear Plant.

First estimations were made through a simulation exercise considering reasonable assumptions on a major accident that could occur on a French 1300 Mwe PWR plant (a S3/10 scenario was chosen for this exercise). In such case, deposition of released activity would lead to levels of the range of 10^4 Bq.m⁻² for ⁹⁰Sr, 10^5 Bq.m⁻² for ¹³⁴Cs and ¹³⁷Cs and 10 Bq.m⁻² for ²³⁹Pu at 2 km from the plant. If the site remediation objective was to bring the levels of public exposure down to recommended limits of 1mSv/year, this would lead to remove soils within a zone of 2 km radius around the plant during the first year after the accident, according to effective dose calculations carried out by mean of the ABRICOT code (developed for the modelling of nuclide transfers into the biosphere (2)). Considering a penetration depth in soils of about 5 cm for the major radionuclides, as could be observed in most sites of the Chernobyl area (3), a volume of about $7 \cdot 10^5$ m³ would have to be removed. This is considerably less than for the Chernobyl case for which the fulfilment of a less stringent remediation criteria (2,2 mSv/year after 4 years) would lead to remove about 10^9 m³ of soil (4). As a base for studies on the feasibility of site remediation, a contaminated area of 10 km radius around the plant was considered to be conservative (scenario S3 for which public exposures are estimated in most cases to be negligible outside the 10 km zone). Such scenario implies the treatment of about 10^7 m³ of soil and $3 \cdot 10^3$ to $3 \cdot 10^4$ t of dry vegetation depending on seasonal conditions. If the treatment was to be completed within 2 years, which we propose to be a working assumption, the removal and the treatment of 2 ha/h (1500 t/h) of soil as well as the treatment of 1 to 100 t/h of fresh plants would be necessary to ensure 24 hours a day. Such figures illustrate that besides the decontamination yields necessary to obtain, the industrial constraints of remediation are of key importance in determining strategies for site restoration.

FEASIBILITY OF INDUSTRIAL TREATMENT

A review of existing standard techniques commonly used in clean up industry was made, in addition to already published information (5). The techniques of heap leaching were at first examined. Preliminary investigations show that heap leaching does probably not allow efficient treatment of large volumes of soils contaminated after an accident of Chernobyl type. Besides the very large number of heaps and volumes of reagent needed for treatment, it is very doubtful that a substantial amount of Cs (which is predominant in

contaminations resulting from NPP accidents) may be removed within the time recommended for treatment. Simulations have been performed in order to evaluate the efficiency of leaching for ^{137}Cs . The results are given in figure 1.

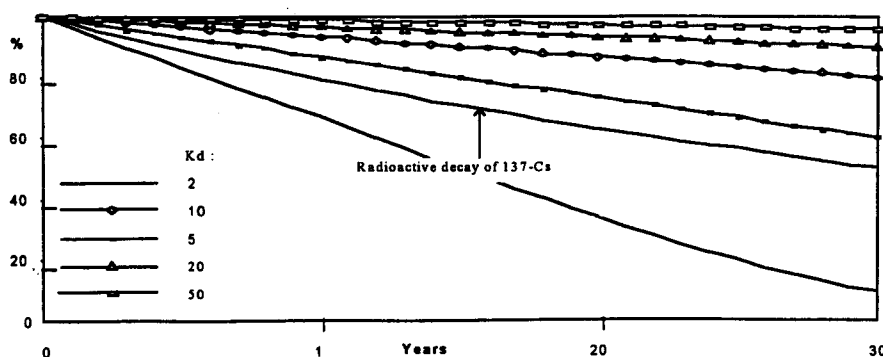


Figure 1. Simulation of the fraction of ^{137}Cs removed (in %) by leaching at a rate of percolation of $0.1 \text{ l.h}^{-1}.\text{m}^{-2}$ save for radioactive decay

It appears that for a percolation rate of $0.1 \text{ l.h}^{-1}.\text{m}^{-2}$, which seems a maximum for heaps of about 10 m height, no significant fraction can be removed within two years even for a distribution coefficient (K_d) of 2 ml/g. For smaller heaps of 3m height where larger percolation rates may be obtained (1 to $5 \text{ l.h}^{-1}.\text{m}^{-2}$), better results can be achieved but still, a very low K_d value is needed. The laboratory tests made on samples from the Chernobyl area show a very strong sorption of Cs on soils which does not allow significant removal by leaching with standard acid reagents. Even if a very efficient reagent was available, the industrial constraints would still remain since enormous amounts of reagent would be needed for efficient leaching (up to $16500 \text{ m}^3/\text{h}$). It appears therefore that leaching techniques cannot be applied to the treatment of contaminated soils in large volumes. It may nevertheless be remembered for smaller amounts of material contaminated with soluble species.

Other standard techniques used in clean up industry follow principles of separation and sorting of material according to their physico-chemical nature so that adapted treatments can be applied to each component of the contaminated media. In rural environments, one would therefore recommend the separate collection and treatment of vegetal cover and soils.

The collection of vegetal cover at the rates indicated in the previous section appears feasible. Several types of engines adapted to the collection of many kinds of cultures and fields at rates ranging from 20 to 40 ha/day are already available at moderate costs. The economical incidence of radioprotection depends on the choice of designing special engines or « nuclearize » existing ones. This choice and the level of protection required for workers are highly related to the time passed before collection since it has been shown that exposures from atmospheric pathways decrease by several orders of magnitude after a few months. The protection required after such period would therefore concern only external exposure from soils. The methods of treatment of the collected vegetation are based on the potential of the latter for volume reduction. A reduction of a factor 10 can easily be expected when using standard techniques of drying and incineration of collected fresh material. Operations can be conducted in units of moderate sizes, easy to set up in a short time scale, or even in mobile units able to treat 60 to $300 \text{ m}^3/\text{h}$ of material. The techniques of treatment of secondary products, such as steam and smoke are well proven and can probably easily match safety requirements through the use of high efficiency filters. Drying and incineration techniques are however demanding in energy. An appropriate supply of fuel or electricity on site must therefore be anticipated. The investments and operational costs (without manpower) were roughly estimated to be of the range of 600 FF (French francs) per ton of treated material. Above operations would lead to the production of a maximum of 30000 m^3 of ashes, which is close to the annual volumes of wastes received by the « Aube » repository (low level waste repository operating in France at present). According to predicted levels of activity deposition presented in previous section, the storage of such material (as well as the direct storage of contaminated soils) should be compatible with the overall amount of activity allowed to be received in LLW repositories (« radiological capacity »). These wastes may however have to match other requirements (such as massic activity limits, immobilization and chemical passivity of product) in order to be accepted for disposal in such facilities.

As for vegetal products, the collection of contaminated soils is feasible at moderate costs and submitted to the constraints that were mentioned above. The principles of treatment of collected soils is based on the separation of mineral and organic particles in a first stage, followed by the removal of polluting substances driven in solution by use of a reagent. The resulting effluents are then decontaminated by standard techniques (concentration by evaporation, co-precipitation, elution in ion-exchange resins, etc.). The separation techniques

are mostly based on gravitational sorting. « Dry » and « wet » processes of sorting can be applied. The dry techniques can consist of a sedimentation of particles in a column blown by an upward air stream, or by sieving on rotating meshes. Nevertheless these techniques have been seldom used for industrial purposes and their ability to treat large volumes of material still needs to be assessed. They are furthermore probably extremely demanding in power supply. The wet techniques are much more frequently used in clean up industry and consist in the decantation of particles in a water stream or the flotation of the small fraction driven to surface by injection of compressed air in water. These operations are often simultaneously completed by the addition of reagents for removal of polluting species. As mentioned before, processes of decontamination using the chemical properties of the polluting species are not promising for contaminated soils of Chernobyl type, since the major radionuclides they contain have shown a very strong affinity to the substrate. On the other hand, it has been shown that the major part of the deposited radioactivity was always contained by the small grain size fraction of soil. The separation processes based on size fractioning could therefore act as decontamination techniques for soils contaminated by a PWR accident. However, the efficiency of former techniques for decontamination purposes still needs to be assessed and must allow the separation of sufficiently fine fractions to provide significant reduction of the volumes of material to be stored. Furthermore, the wet separation techniques need heavy infrastructure and are extremely demanding in water supply (up to 8 m³/s) so that their implementation requires delay and appropriate siting. An optimistic estimation of investments and operational costs for the treatment of the overall 10 km zone (based on the assumption that decantation and flotation can treat a load of 100 g/l of suspended material which is probably a maximum) would be of the order of 3 10⁸ FF, but the manpower needed for building the facilities and the transport of material to the treatment plant may considerably increase this cost.

CONCLUSIONS

The studies carried out on the ability of standard industrial techniques to treat very large volumes of contaminated material from rural environments have shown that it is certainly possible to treat a contaminated vegetal cover within a short time without undergoing major industrial and economical constraints. The drying and incineration of collected plants may allow a reduction of the volumes of wastes to store by at least a factor of 10. Some investigations on the nature of the wastes produced are nevertheless necessary in order to assess if additional conditioning is needed to match safety requirements for disposal. The optimization of the treatment of vegetal cover raise the question of the time necessary to leave before beginning operations. An early collection may potentially allow the removal of most of the deposited activity but present higher risks of exposure for workers. The question of finding a balance is, in other respects, also raised for the implementation of some early counter-measures of agricultural type (ploughing, planting) which enable an immediate reduction of exposures by dilution of deposited activity but involve additional difficulties in achieving total decontamination (increase of the volumes to treat). The treatment of soils in large volumes is much less promising, because of heavy constraints of industrial implementation (large infrastructures and very important needs for water and power supply). Furthermore, all the reviewed techniques have to be calibrated with respect to their ability to achieve significant decontamination for soils contaminated after a NPP accident. The technologies based on size fractionating seem the most interesting to test since they may allow good decontamination and a reduction of the volumes of wastes to store. But their efficiency must be assessed at industrial scale and the constraints of implementation still remain for the treatment of large volumes. From above considerations, we would recommend to address the following issues : treat vegetal covers and soils separately ; look for a reduction of the volumes of soil to treat by methods of radiological sorting ; calibrate and improve technologies based on size fractionating ; examine the ability of wastes produced to meet safety requirements for disposal. Finally, optimization should be made by considering, in particular : the balance between the doses spared for people living on decontaminated sites against the doses received by workers as well as by people exposed to the secondary products of decontamination ; the balance between the benefit of early counter measures against total and long term decontamination ; the balance between the radiological and economical costs of the reduction of the volumes of wastes to store against direct disposal of all collected materials.

These conclusions concern of course only rural environments contaminated after an accident of Chernobyl type (NPP accident). We believe that a real optimization must consider together all the categories of environments touched (rural, forests, aquatic, urban). Other types of accidents must also be addressed.

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