THE RESULTS OF A COORDINATED RESEARCH PROJECT ON THE SURFACE CONTAMINATION OF PACKAGES

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ABSTRACT

A coordinated research project (CRP) on the surface contamination of packages was conducted by the IAEA in 2000-2003, in which detailed study was carried out to determine whether the current requirements for surface contamination of packages remain adequate. Deliberations to revise the regulatory limits on the surface contamination were followed. This paper presents the results of this study and final outcomes of the work done in the course of many meetings spread over 10 years.

1. INTRODUCTION

The production and use of radioactive material in activities relating to the generation of nuclear power involve the transport of radioactive material in the public domain. In accordance with its statutory authorization, the International Atomic Energy Agency (IAEA) publishes safety standards, including the Regulations for the Safe Transport of Radioactive Material (the Regulations, TS-R-1 [1]), which undergo continuous review and are revised as necessary. The IAEA transport regulations, which are prepared in consultation with experts from IAEA Member States and other organizations, are adopted essentially in their entirety by various international transport organizations and by many countries.

The Regulations, inter alia, stipulate the limits for the level of radioactive contamination on the external surfaces of packages that are used for the transport of radioactive material. The majority of packages, conveyances and associated equipment typically are not contaminated. However, packages, conveyances and equipment in use or associated with parts of the nuclear fuel cycle are prone to become contaminated during transport. The approach to non-fixed contamination control in the Regulations consists in applying derived limits for non-fixed surface contamination together with the requirement to keep contamination ‘as low as practicable’:

In para 507 of the Regulations, the provision stipulate the principle and the limit for contamination:

507. The non-fixed contamination on the external surfaces of any package shall be kept as low as practicable and, under routine conditions of transport, shall not exceed the following limits:
(a) 4 Bq/cm^2 for beta and gamma emitters and low toxicity alpha emitters; and
(b) 0.4 Bq/cm^2 for all other alpha emitters.
These limits are applicable when averaged over any area of 300 cm^2 of any part of the surface.

The original basis, known as the Fairbairn model, for the derived limits for non-fixed contamination was not transport specific but consistent with the models used for derived limits for contamination in laboratories, hospitals and industry. The approach has proved to be sound and was used in the first edition of the Regulations and has remained virtually unchanged up to the present day.

In 1998 the contaminated packages and wagons used routinely to transport irradiated fuel between Germany, Switzerland and France, and some shipments to the UK were found. Although this incident had not resulted in any significant radiological risks, the problems resulted in the suspension of the use of these packages for a few years.

In the follow-up period, extensive surveys and investigations were carried out to establish the causes, nature and magnitude of the contamination, to seek remedies and regulatory changes.

2. LIMITS AND OPTIMIZATION

Nuclear fuel transport packages may become contaminated with radioactive materials on both internal and external surfaces, from contact with water in the fuel storage pools at nuclear installations when loading or unloading fuel. This effect has been well known and reported for several decades.

Transport package surface contamination arises mainly from dissolved or particulate radioactive materials in the storage and cooling pool water. These materials are activated corrosion products from the primary reactor cooling loop and fission or activation products from the fuel elements. In light water reactors, the main contaminants are the activated corrosion products ^{60}Co, ^{110}Ag, or ^{54}Mn and the fission product ^{137}Cs.

The extent and magnitude of surface contamination is also dependent on the concentrations and chemical forms of the radioactive constituents in the pool water, and the contact time in the pool.

Surface contamination may require extensive decontamination in order to comply with the Regulations, both on dispatch and throughout the journey. Some of this difficulty is due to a phenomenon referred to as ‘weeping’ or ‘sweating’ where a surface that has been decontaminated becomes recontaminated. This phenomenon is due to fixed contamination becoming removable due to changes in humidity, temperature, handling, etc.

In the case of fuel packages which may have been immersed in contaminated cooling pool water whilst being loaded with irradiated fuel, contamination may become ingrained in pores, fine cracks and crevices, particularly in the vicinity of lid seals. Subsequent weathering, exposure to rain or even exposure to moist air conditions may cause some fixed contamination to be released or to become non-fixed. That could cause non-compliance with the contamination limits during or at the end of the journey.

However, this situation generally presents no significant hazard because of the pessimistic and conservative assumptions used in calculating the derived limits for non-fixed contaminations.

In some cases, consignors make extensive efforts to decontaminate the package to a fraction, such as 1/10, of the allowable limit in order to accommodate 'weeping' and prevent a non-compliance situation. This led to increased exposures of the workers carrying out these duties. The impact on the collective radiation dose of decontamination activities was evaluated. At higher direct exposure rates (0.1 and 1 mSv/h), any additional decontamination efforts resulted in higher total doses as the increase in worker dose was much greater than the resulting reduction in public dose.

These events have raised the question of how to ensure the optimization of radiation protection while maintaining the levels of non-fixed contamination below regulatory limits.

The IAEA Basic Safety Standards call for the optimization of protection and safety "in relation to exposures from any particular source within a practice..." and states that "...protection and safety shall be optimized in order that the magnitude of individual doses, the number of people exposed and the likelihood of incurring exposures all be kept as low as reasonable achievable...within the restriction that the doses to individuals delivered by the source be subject to dose constraints". In the case of non-fixed surface contamination on spent fuel packages there are exposures to both workers and the public that must be taken into account.

3. EFFORTS MADE TO RESOLVE THE ISSUE

3.1 Coordinated Reserch Project

The Transport Safety Standards Committee (TRANSSC) of the IAEA recommended at its May 2000 meeting that the IAEA undertake a Coordinated Research Project (CRP) on the Radiological Aspects of Package and Conveyance Non-Fixed Contamination. The CRP Report was issued in June 2005 as IAEA-TECDOC-1449 [2]. This TECDOC represents the final results of the research undertaken during this CRP.

The CRP reviewed the scientific basis for the current regulatory limits for surface contamination. The CRP was also to develop guidance material for evaluating the radiological significance of surface contamination to workers and the public in the light of state of the art research and technical developments and current transport practices.

The specific objectives of the work undertaken within this multi-national CRP were, in accordance with the terms of reference:

- To ensure that appropriate models exist for all package types including consideration of the aspects pertinent to assessing and revising a surface contamination model for transport;
- To collect — where possible — contamination, operational and dosimetric data to ensure modelling consistency;
- To develop models for assessing the radiation doses incurred in transport operations, and
- To consider preventive methods for package and conveyance contamination.

The CRP reviewed and analyzed the critical issues and characterized the dose pathways for non-fixed surface contaminants on packages and conveyances under routine conditions of transport and quantified the resulting internal and external radiation dose to transport workers and the public This

assessment was to take into account the different categories of radioactive material packages used in practice and related transport conditions typically encountered in the transport of radioactive material.

The Basic Model was developed to establish a direct relation between the non-fixed contamination level per unit area on the package and the annual dose limit for workers or public for each radionuclide. However, no such relation has been worked out because the CRP did not address the issue of making decisions on dose limits. The Basic Model results are linear and can be simply scaled for any given dose criterion. That is, given a dose criterion, surface contamination levels can be derived with the Basic Model.

Using the Basic Model, the CRP calculated doses to people from a reference contamination level of 1Bq/cm^2 assuming a uniform contamination of the package surface. The Basic Model provides individual doses for at least 49 selected radionuclides. Results for each of the 356 radionuclides that are listed in Table 2 of the Regulations were also calculated. The results for 49 radionuclides for four different package types have been presented in the TECDOC.

It should be noted that certain uncertainties inherent to this model, some of which are not conservative. These include the resuspension rate, which varies from 1x 10^{-3} per h to 1x 10^{-7} per h. The external doses due to beta and neutron emitters, and the influence on the skin dose or on ingestion of 'hot spots' are not taken into account.

However the Basic Model contains a number of assumptions that have been identified to be conservative, i.e. which lead to upper estimates of dose for a given surface contamination.

Observation and conclusion are summarized as follows:

1) The studies carried out under the CRP indicate that the present limits on non-fixed contamination on the surfaces of packages and conveyances are conservative.

2) Fairbairn model was limited and outdated and that a new comprehensive model (the Basic Model) should be produced to provide the basis for eventually deriving new contamination limits for transport situations.

3) The approach taken was to characterize and model the dose pathways for non-fixed contamination on packages and conveyances under normal transport conditions and to calculate potential radiation doses to workers and the public.

4) The transport and exposure conditions reflected in the Basic Model do not reflect a particular transport situation or geographical transport region, but are believed to provide a practical and reliable means for the assessment of occupational and public exposures that are unlikely to be exceeded under actual transport conditions.

5) With respect to the entire set of 49 nuclides, the values of the annual dose per unit surface contamination span nearly seven orders of magnitude for workers and nearly eight orders of magnitude for members of the public.

6) The highest of all values of annual dose per unit surface contamination (both for workers and the public) is calculated for ^{227}Ac. ^{227}Ac decays by both alpha and beta emission, emitting a range of alpha and beta particles together with electrons and gamma ray photons. This radionuclide produced doses of at least an order of magnitude higher than all other radionuclides assessed in this

study, and is several orders of magnitude higher than the majority of radionuclides. However, ^{227}Ac is not normally associated with practical contamination problems in transport.

7) The lowest range of values for the annual dose per unit surface contamination is calculated for very weak beta/gamma emitters like ^{59}Ni which have an extremely low radiological relevance.

8) Special consideration should be given to tritium, since this radionuclide raises important difficulties for prevention of contamination as tritium is present in water or steam form.

9) The categorization of radionuclides into 'beta and gamma emitters', 'low toxicity alpha emitters', and 'all other alpha emitters' (vide paragraph 214 of the Regulations) with only two values (4 and 0.4 Bq/cm^2) does not represent the span of seven orders of magnitude in the results of the Basic Model.

10) The distinction between "low toxicity alpha emitters" from the other alpha emitters is not valid in the context of the results of the Basic Model because some of the low toxicity alpha emitters listed in TS-R-1 are responsible for some of the highest doses, these being ^{232}U, ^{232}Th and Th-nat (natural).

11) Worker doses were found to be about three orders of magnitude higher than doses for critical members of the public for all radionuclides. Therefore, in deriving revised limits for the non-fixed surface contamination, workers would be more limiting than members of the public.

12) The dominating package type for the majority of radionuclides was 'small, manually handled packages', although there is little difference between the maximum doses obtained for all package types, assuming the same level of surface contamination. For each of the strong gamma emitters, the dominant package type was small, remotely handled packages.

13) Care should be taken if the results for small, manually handled packages are used for future regulations, and non-uniformity of contamination should also be considered because in reality, small, manually handled packages are very rarely contaminated, and all other package types that are contaminated tend to have non-uniform contamination on the surface.

14) It would be, however, an easy task to provide radionuclide specific rounded values for each radionuclide in a separate column of Table 2 in Section IV of the Regulations.

15) The analysis shows that
- assumptions on annual working time,
- the resuspension rate and
- assumptions on transfer of activity

are those parameters that have the highest influence on the overall result.

3.2 Outcomes of CSM in June 2006

The Consultant Service Meeting (CSM) was held in June 2006 in Vienna to review the CRP results to develop a rationalized approach to package removable surface contamination limits. The followings are the outcomes:

1) The majority view of the participants was to move to a dose-based system for defining contamination limits using a radionuclide-specific approach and a reference dose of 0.3 mSv/y for transport workers.
Some participants favored keeping the existing contamination limits and radiation emission grouping,

2) Difficulties in measuring radionuclide specific values were discussed, both at the consignor's facility where the radionuclide composition can be well defined and during transport when the radionuclide composition may not be as well characterized.

It was concluded that a radionuclide-specific approach would not be likely to introduce compliance difficulties, though some participants do not believe this is the case.

3.3 Outcomes of CSM in 2007

In February 2007 a further consultancy meeting in London was held to advance the work that has been done by the CRP and the results of the CSM held in June 2006. The outcomes were:

1) In order to retain consistency and simplicity the current values *(0.4 and 4 Bq/cm^2)* for contamination were retained. In addition, an option was added for a radionuclide specific approach.

2) The current contamination values are not derived from a transport specific dose model. The meeting agreed that the regulatory provisions should include values based on sound science developed in the CRP model.

3) Use of 1mSv/y for worker dose constraint.

4) The group agreed that there should be an upper and lower bound for contamination limits. The current values of 0.4 Bq/cm^2 for alpha contamination should be retained as the minimum value. An upper bound of 100 Bq/cm^2 was chosen to avoid gross contamination.

5) It was agreed that a radionuclide specific approach would be adopted for those isotopes above the minimum and below the maximum values. No grouping would be applied to these isotopes.

6) The meeting recommended that as the CRP dose model was based on uniform contamination over the whole package surface contamination measurements should also be averaged over the whole surface, rather than be set as a maximum allowed value.

3.4 Outcomes of TM in 2008

The Technical Meeting (TM) was held in November 2008 in Japan to further address the issue of surface contamination. Based on the results of CRP and efforts of subsequent meetings, although there are differences in opinion between member states, the meeting achieved significant progress and proposed consensus changes to the Regulations subject to further detailed review and the normal revision process for IAEA documents.

1) Radionuclide specific limits should be Subject to:
   - Multi-lateral approval,
   - Applied to Irradiated Nuclear Fuel (INF) packages with an activity greater than 10^5 A_2,
   - List in column 6 of Table 2 of the latest Regulations, based on 0.3 mSv dose criterion for CRP results based on INF packages with a lower boundary of 0.4 Bq/cm^2.

2) There was consensus that the numbers for radionuclide specific limits should be contained within the regulations and guidance will be given on the application and restrictions of applying this approach.

3) There was a consensus to use the current maximizing method where the maximum value found was compared with the contamination limit defined in the regulations.

4) It was agreed to have a capping method to provide a ceiling value, but the mechanism would need to be proposed.

5) On measurements techniques it was agreed to produce information on the use of radioisotope fingerprinting to make easier implementation

6) Proposed texts were developed for paras 401, 402, 403, 404, 508, 509, 513, 520, 820, 822, 832.

3.5 Decision of TRANSSC

In October 2009, TRANSSC, at its 19th meeting, addressed the issue of contamination and reviewed the suggestions made by the CRP and previous meetings. TRANSSC concluded that the surface contamination issue is not accepted for regulatory change at this time, mainly because simple regulations and regulatory limits were preferred and practicability of radionuclide specific control were of concern. The current regulations were considered to provide adequate safety.

4. CONCLUSIONS

Since the report that contamination in excess of the regulatory limits was observed in some flasks and wagons transporting radioactive material in 1998, it was recognized that the issues related to contamination control of package and conveyance needed to be re-evaluated. The existing conservative limits may lead to operational problems, non-compliance issues, and public perceptions of significant health risks when the actual risks are very low.

Optimization of protection, in compliance with the regulatory limits, warrants taking into account all the dose components, received in the various operations related to transport of a package and resulting from the package contamination.

Great efforts were made by coordinated research, meetings and discussions among the radioactive material transport community. Although these efforts do not result in regulatory changes at this time, they did demonstrate the resolution path for the problem.

Some important outputs are:

1) New dose-based approach (Basic Model) was developed specifically for the transport of radioactive material;

2) The conservatism of current limits was confirmed by the new model;

3) Optimization was implemented when assessing the surface contamination limits;

4) The reasons of recontamination during transport were recognized and measures coping with the problem were developed;

5) The radionuclide specific limits were suggested which are supposed to be the optimized approach for the surface contamination control.

Still some outstanding questions are not answered yet which are the obstacles to the application of the radionuclide specific limits and need further work:

1) Certain uncertainties inherent to new model may not be conservative like the resuspension rate. The external doses due to beta and neutron emitters, and the influence on the skin dose or on ingestion of 'hot spots' are not taken into account.

2) The measurements techniques for specific radionuclides approach need further work to make the method easer to implement.

Dose optimization, contamination prevention, and decontamination are topics that are inter-related. It was noted that in some countries the operational approaches now include additional contamination prevention measures that may result in less effort for decontamination and monitoring. Other efforts like on package design, decontamination and measurement technology may also contribute to the dose saving.

The work on optimization of surface contamination control during transport in the IAEA now is brought to a temporary close. The better approach is identified and further work is needed for application.

ACKNOWLEDGMENTS

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REFERENCES

[1] INTERNATIONAL ATOMIC ENERGY AGENCY, Regulations fro the Safe Transport of Radioactive Material 2009 Edition, IAEA Safety Standards Series No. TS-R-1, IAEA, Vienna (2009)

[2] INTERNATIONAL ATOMIC ENERGY AGENCY, Radiological aspects of non-fixed contamination of packages and conveyances, IAEA-TECDOC-1449, IAEA, Vienna (2005)