

Newcastle University e-prints

Date deposited: 25th June 2010

Version of file: Author, final

Peer Review Status: Peer-reviewed

Citation for published item:

Gray I, Sievwight B, Reeks M. [Modelling of release of particulate material from transport containers](#). *Packaging, Transport, Storage & Security of Radioactive Material* 2010,**21** 1 13-18.

Further information on publisher website:

<http://www.maney.co.uk/> (*Website*)

Publishers copyright statement:

This paper was originally published by Maney Publishing, 2010 and can be viewed (with permissions) from the URL below:

<http://dx.doi.org/10.1179/174650909X12511278683302>

Always use published version when citing.

Use Policy:

The full-text may be used and/or reproduced and given to third parties in any format or medium, without prior permission or charge, for personal research or study, educational, or not for profit purposes provided that:

- A full bibliographic reference is made to the original source
- A link is made to the metadata record in Newcastle E-prints
- The full text is not changed in any way.

The full-text must not be sold in any format or medium without the formal permission of the copyright holders.

<p>Robinson Library, University of Newcastle upon Tyne, Newcastle upon Tyne. NE1 7RU. Tel. 0191 222 6000</p>

Modelling of release of particulate material from transport containers

I. Gray^{*1}, B. Sievwright² and M. Reeks³

The Nuclear Decommissioning Authority (NDA) is developing a family of Standard Waste Transport Containers (SWTCs) for the transport of unshielded intermediate level radioactive waste packages. The SWTCs are shielded transport containers designed to carry different types of waste packages. The combination of the SWTC and the waste package is required to meet the regulatory requirements for Type B packages. One such requirement relates to the containment of the radioactive contents, with the IAEA Transport Regulations specifying release limits for normal and accident conditions of transport. In the impact tests representing accident conditions of transport, the waste package will experience significant damage and radioactive material will be released into the SWTC cavity. It is therefore necessary to determine how much of this material will be released from the cavity to the external environment past the SWTC seals. Typical assessments use the approach of assuming that the material will be evenly distributed within the cavity volume and then determining the rate at which gas will be released from the cavity, with the volume of radioactive material released with the gas based on the concentration of the material within the cavity gas. This is a pessimistic approach as various deposition processes would reduce the concentration of gas-borne particulate material and hence reduce their release rate from the SWTC. This paper assesses these physical processes that control the release rate and develops a conservative methodology for calculating the particulate releases from the SWTC lid and valve seals under normal and accident conditions of transport, in particular:

- a) the flows within the SWTC cavity, especially those near the cavity walls;
- b) the aerodynamic forces necessary to detach small particles from the cavity surface and suspend them into the cavity volume;
- c) the adhesive forces holding contaminant particles on the surface of a waste package;
- d) the breakup of waste material upon impact that will determine the volume fraction and size distribution of fine particulate released into the cavity.

Three mechanisms are specifically modelled, namely Brownian agglomeration, Brownian diffusion and gravitational settling, since they are the dominant processes that lead to deposition within the cavity and the easiest to calculate with much less uncertainty than the other deposition processes. Calculations of releases under normal conditions of transport concentrate on estimating the detachment of any waste package surface contamination by inertial and aerodynamic forces and show that very little of any contamination removed from the waste package surface would be released from the SWTC. Under accident conditions of transport, results are presented for the fraction released from the SWTC to the environment as a function of the volume fraction of the waste package contents released as fine particulate matter into the SWTC cavity. These show that for typical release fractions of 10^{-6} to 10^{-8} for the release of radioactive material from waste packages into the SWTC cavity, the release fraction of the waste package inventory from the

¹AREVA Risk Management Consulting Limited, UK

²School of Mechanical and Systems Engineering, Newcastle University, UK

³Nuclear Decommissioning Authority, Radioactive Waste Management Directorate, UK

*Correspondence author, email iain@ilsgray.co.uk

SWTC of typically 10^{-9} to 10^{-10} . Hence, the effective decontamination factor provided by the SWTC is 10^2 to 10^3 . Whilst this analysis has been carried out specifically for the SWTC carrying waste packages, it is applicable to other arrangements and its use would reduce the high degree of pessimism used in typical containment assessments, whilst still giving conservative results.

Keywords: Containment, Transport, Particulate

Introduction

The Nuclear Decommissioning Authority is developing a family of standard waste transport containers (SWTCs) for the transport of unshielded intermediate level radioactive waste packages, namely four 500 L Drums within a stillage; one 3 m³ Drum or one 3 m³ Box. The SWTCs provide shielding for the waste packages during transport and also form the containment boundary.

For the containment of the radioactive contents, there are release limits specified in the IAEA Transport Regulations¹ for normal and accident conditions of transport. Under accident conditions, impact tests may lead to radioactive material being released into the SWTC cavity. It is therefore necessary to determine how much of this material will be released from the cavity to the external environment past the SWTC seals. Typical assessments use a pessimistic approach that takes no account of deposition processes that reduce the concentration of gas-borne particulate material in the cavity and hence reduce the release rate from the SWTC.

This paper considers the behaviour of gas-borne particulate material within the SWTC cavity and subsequent leakage via the SWTC lid and valve seals under both normal and accident conditions of transport.

Physical processes

The methodologies in normal and accident conditions of transport are similar but there are important differences that reflect the way fine particulate material is released into the SWTC cavity. Both methodologies illustrate the important role that particle size plays in determining both types of release. The various physical processes that control the release rate comprise:

- flows within the SWTC cavity, especially those near the cavity walls
- aerodynamic forces necessary to detach small particles from the cavity surface and suspend them into the cavity volume
- adhesive forces holding contaminant particles on the surface of a waste package
- break-up of waste material upon impact that will determine the volume fraction and size distribution of fine particulate released into the cavity.

Three mechanisms are specifically modelled, namely Brownian agglomeration, Brownian diffusion and gravitational settling, since these are the dominant processes that lead to deposition within the cavity.

There are a number of processes that are all in some way concerned with transport of individual particles within the flow in the SWTC cavity. These can be subdivided into two categories: deposition processes which lead directly to deposition on surfaces exposed to the flow and those processes which lead indirectly to

deposition by enhancing the underlying deposition processes.

Direct deposition processes refer to convection-diffusion processes that can be characterised by a deposition velocity and are broken down into those whose diffusion (close to the depositing surface) is molecular-thermodynamic in origin and those associated with the turbulent motion of the underlying carrier flow.

Molecular-thermodynamic processes refer to Brownian diffusion (arising from molecular impacts with gas-borne particles) and thermophoresis in which particles migrate with a given convective velocity from high to low regions of temperature, i.e. in the opposite direction to the temperature gradient. Both processes are molecular in origin, although Brownian diffusion for gas-borne particles is the most understood and the most accurate and reliable to calculate even when the sizes of the suspended particles are very small ($<10^{-9}$ m) (when the gas can no longer be regarded as a continuum).

The transport of particles undergoing Brownian motion is analogous to the transport of heat, and the heat mass transfer analogy can be used. Consequently, the deposition velocity [expressed in terms of the Sherwood number (Sh)] can be expressed as a function of the particle Reynolds number (Re) and Schmidt numbers (Sc). This function is equivalent in heat transfer to the Nusselt number being a function of the Reynolds and Prandtl numbers.

The other deposition mechanism is turbulent deposition arising from the underlying turbulent motion of the carrier flow. What is crucial in all turbulent flows is the flow behaviour near the depositing wall and in particular in the thin turbulent boundary layer where the turbulence intensity and timescale change steeply in a direction normal to the wall. This implies that the process is very much dependent upon particle size which determines firstly how far the centre of a depositing particle is from the wall when impact occurs. Thus the greater the size of the particle, the greater the deposition velocity: this is called 'interception'. Secondly particle size determines the particle inertia and its lack of response to changes in flow velocity as a depositing particle approaches the surface. More specifically, particle inertia is measured by the Stokes number of the particle, which is the ratio of the particle response time to the timescale for changes in flow.

It is important to appreciate the sensitivity of the particle inertia to the particle diameter compared to the particle density. Particle diameter plays a crucial role in all the physical processes discussed in this paper that are relevant to the release of particles. There is an extensive literature on turbulent deposition and transport and it is in general significantly higher than deposition due to Brownian motion. The reason is bound up in length scale associated with turbulent motion (eddy length scale) which is much greater than for Brownian motion.

In wall deposition, however, the very small gas velocities encountered by a particle as it approaches the wall means that small particles (i.e. low Stokes numbers) find it increasingly difficult to reach the wall with reducing size, so that at some stage the deposition due to Brownian motion will start to dominate over that due to turbulence near the depositing wall.

Gravitational settling is the most straightforward and easiest to apply of all the deposition mechanisms, with the settling velocity determined by a balance of the particle weight with its aerodynamic drag.

In the release rate calculations presented here, it is the growth of an individual aerosol particle that can have the controlling or rate limiting influence on releases since it enhances the settling out of aerosol particles under gravity and so reduces the release rate to the external environment. There are a number of such processes, such as growth by condensation or scavenging. The one that is used in this paper is particle agglomeration, i.e. where there is a growth in particle size as smaller particles come together to form agglomerated larger particles.

Agglomeration is intimately associated with colliding gas-borne particles and is to do with their relative motion. This can be modelled by considering the behaviour of two colliding spherical particles, using individual particle volume as a variable in preference to particle radius, since this is conserved in a collision so in the absence of deposition the total volume fraction of gas-borne particles remains constant during agglomeration, as only the actual volume distribution of individual agglomerate particles changes.

The relative motion giving rise to particle collision may be induced by a number of processes: Brownian motion, non-uniformity in the flow such as shear or turbulence or external forces such as gravitational, electrostatic or Van der Waals Forces. The analysis focuses on Brownian agglomeration, which is the most understood, most documented in the literature and the one with the least uncertainties, and by ignoring all other agglomeration processes makes the release rate results highly conservative. As far as non-spherical particles are concerned, the agglomeration is enhanced owing to the greater surface area compared with that of spheres of the same volume.

A well-established similarity/self preserving particle size distribution method is used explicitly to model the agglomeration process. Solutions found in this way imply that after a long time (depending on the number of particles per unit volume), a stable size distribution will be formed regardless of the initial size distribution. The resulting size distribution is called the self-preserving size distribution and is approximately lognormal.

Conservative calculations of agglomeration rates

In the calculation of the release of particles from an SWTC, use is made of self-preserving size distribution and the expression given by the volume average radius based on the continuum approximation for agglomerating spheres. For very small particles, of the order of 10 nm in size, agglomeration rates are based on the free molecular regime which takes into account the slip factors as a function of radius. In these cases no self

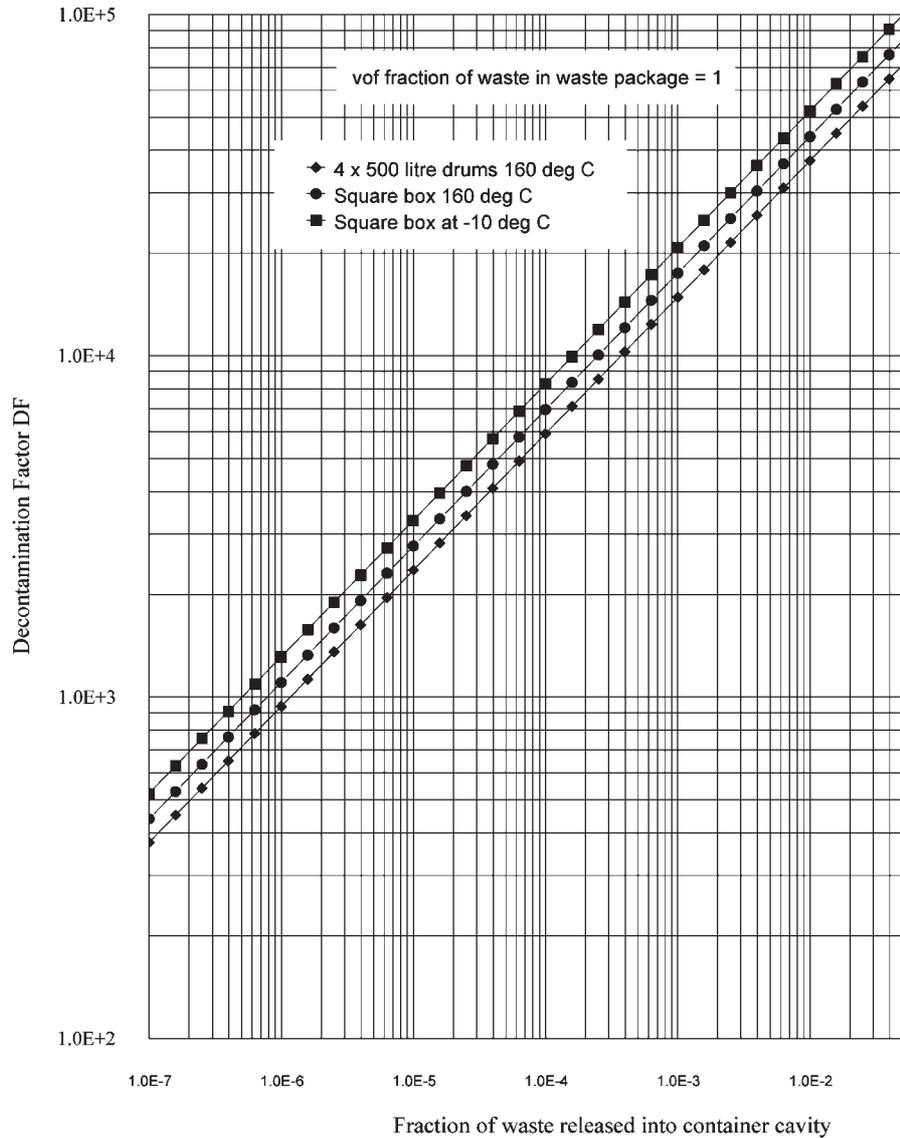
preserving distribution exists and the calculations would in principle be more complicated, though forms for the particle size distribution have been calculated assuming a log normal distribution with analytical forms for both the geometric standard deviation and geometric mean particle volume have been derived.² An important factor as far as releases from the SWTC are concerned is that, for a given initial aerosol size distribution and volume fraction, the rate of increase of both geometric mean and standard deviation of the agglomerate size distribution are all significantly greater than the corresponding values based on a continuum approximation (zero Knudsen number), so use of continuum approximations means the calculations are conservative. Furthermore in the evolution of the agglomerate particle size distribution towards eventual deposition, most of the agglomeration period will be spent in the continuum regime, where the number density of particles, which control the collisions rate, will be considerably less than in the free molecular regime.

Collision efficiencies for Brownian agglomeration

The formulae for the agglomeration kernel and for the self-preserving size distribution and ultimately for the deposition rate constant for gravitational settling implicitly assume that particles adhere to one another after a collision. This depends on a number of factors, in particular particle size, the adhesive force between particles during impact (which depends on the adhesive surface energy) and the amount of energy lost during deformation due to impact. Based on work on elastic/plastic spheres undergoing normal impact,³ it can be shown that a collision efficiency of ~ 1 based on a distribution of impact velocities due to Brownian motion, is an acceptable assumption for particles sizes $>10^{-9}$ m. These assumptions result in a collision efficiency increasing with increasing particle size, approaching 100% efficiency for particle sizes $>10^{-8}$ m. However a significant feature of the analysis is that, even allowing for a dramatic reduction in the adhesion during impact due to surface asperities (of size typical of the primary particles that make up the agglomerates), and where it is most likely to affect the formation of agglomerates $<10^{-8}$ m in size, this has little effect on the quantity of material released from the SWTC. This lack of sensitivity to the collision efficiency for these very small agglomerate sizes is a reflection of the fact that the agglomeration rate in this regime is so great and their lifetime so small that their contribution to the release from this very small size regime is negligible.

The basic assumptions for both accident and normal transport conditions are as follows:

- particles released into the gas volume are fully mixed (i.e. of uniform concentration)
- movement of the gas within the cavity is due to natural circulation (i.e. buoyancy driven)
- deposition of suspended particulate to surfaces exposed to the gas is due to gravitational settling and Brownian motion
- gravitational settling and Brownian diffusion in the capillary have been ignored, with transmission of particles through the capillary occurring up to a particle diameter equal to the capillary diameter, as this simplifies the calculations.



1 Decontamination factor for release of material from SWTC in relation to fraction of waste package contents released into SWTC cavity under accident conditions

Normal conditions of transport

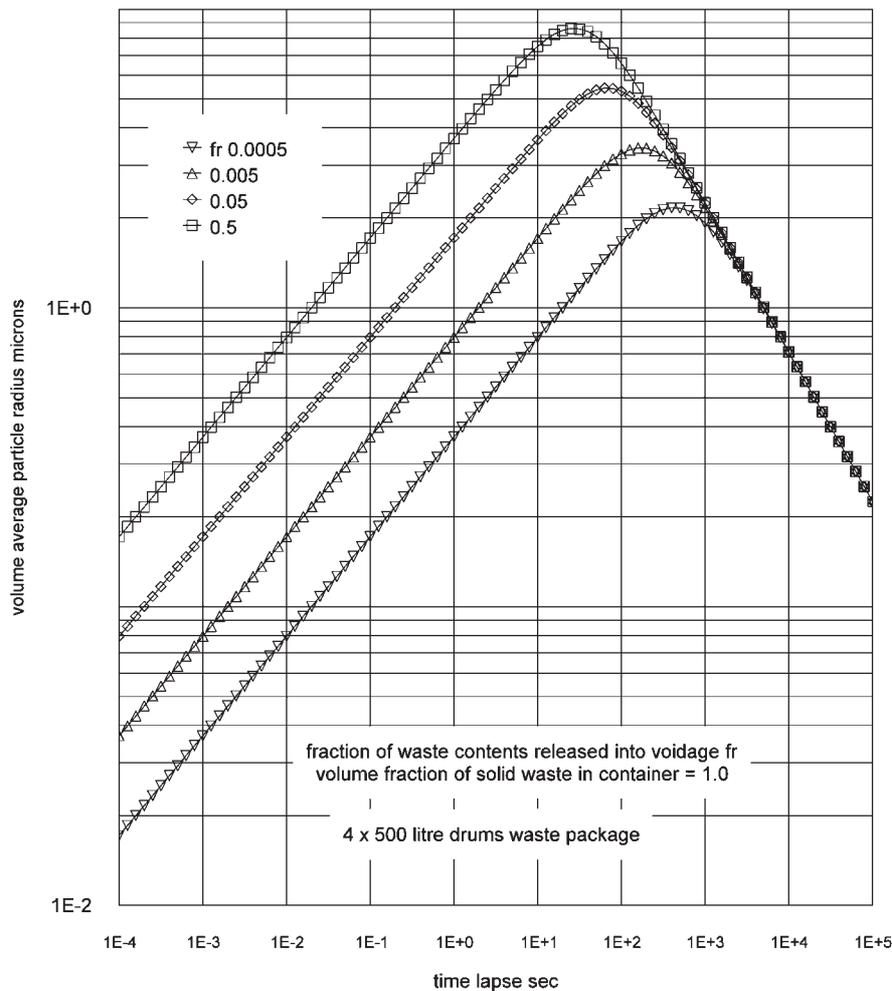
The waste packages will retain their integrity under the impact tests representing normal conditions of transport¹ and hence there would be no release of radioactive material from the waste packages. However, the impact energy from these impact tests and the acceleration and deceleration forces during normal conditions of transport could be sufficient to cause the release of contamination on the external surface of the waste packages. Analysis was carried out to determine the detachment of fine particulate material adhering to the external surfaces of the waste packages by aerodynamic forces (generated by the recirculating flows in the SWTC cavity) and by inertial forces (impulsive forces) experienced during the impact tests.

Surface roughness can significantly reduce adhesion as well as produce a lognormal distribution of adhesive forces with a very broad geometric standard deviation and this was addressed by taking a value of 0.01 for the reduction in adhesion compared to that for perfect smooth contact and a geometric standard deviation (spread) of 10 based on measurements of adhesive forces

from a nominally smooth stainless steel surface.⁴ Furthermore it was assumed that the external surfaces of the waste packages were covered with a monolayer of fine particles and that their size was the cutoff size for removal of particles from surfaces by the recirculating flows in the SWTC cavity. Even with these pessimisms for the particles surface mass and size, the fractions released from the SWTC were estimated to be $<0.4 \times 10^{-14}$ for removal by aerodynamic forces and $<0.5 \times 10^{-13}$ by inertial forces, which are negligible and any uncertainties in these fractions are essentially not significant.

Accident conditions of transport

Taking account of a period of one week after the impact and fire tests representing accident conditions of transport, in line with the requirements of the IAEA Transport Regulations,¹ the variation of decontamination factor (DF) across the SWTC containment boundary with the fraction of material released from the waste packages into the SWTC cavity is shown in Fig. 1. These results take account of deposition in the SWTC cavity



2 Particle volume average radius as function of time for range of values for fraction of material released from waste package(s) into SWTC cavity under accident conditions

by gravitational settling and Brownian motion, together with a changing particle size distribution determined by Brownian agglomeration. The DF value represents the ratio of the quantity of radioactive material released into the SWTC cavity to the quantity of material outside the SWTC. For example, if the total activity within four 500 L Drum waste packages is $10^8 A_2$ and a fraction of 10^{-6} of this was released into the SWTC cavity, i.e. $10^2 A_2$, the DF is given as 9×10^2 . Consequently, the total activity released from the SWTC is given by $10^2 A_2 / 9 \times 10^2$, which equals $0.1 A_2$, which is well below the regulatory limit of one A_2 .

Results are shown in Fig. 1 for four 500 L Drum waste packages and for a single 3 m^3 square cornered Box waste package, as these correspond to the maximum and minimum net empty volumes within the cavity SWTC respectively, and hence the maximum and minimum releases respectively for a given fraction of waste material released into the cavity volume. Results for the round cornered 3 m^3 Box and the 3 m^3 Drum waste packages would lie within these results.

It should be noted that the DF actually increases with the fraction of the material in the waste package(s) that is released into the SWTC cavity. This is because as the quantity of material is released into the cavity increases, this leads to an increase in the agglomeration rate and hence an increase in deposition within the SWTC cavity at any given time.

There will also be an influence of the cavity temperature on the release of material from the SWTC and this is shown in Fig. 1, where DFs for the 3 m^3 square cornered Box waste package are presented at two extremes of temperature, namely 160°C , which corresponds to the maximum SWTC cavity temperature during the fire test, and -10°C . It can be seen that there is not a large difference between the two extremes, with the lower temperature giving the higher DF, i.e. a lower release. This is because the agglomeration rate depends on the Brownian diffusion coefficient, which in turn depends on two quantities which both depend on temperature, but in opposing ways, namely thermal energy and the dynamic viscosity of the air in the cavity. While the thermal energy reduces with reducing temperature and hence reduces the agglomeration rate, the viscosity increases with reducing temperature and hence increases the agglomeration rate. It turns out that the increase in agglomeration rate due to a reduction in viscosity outweighs the reduction in the agglomeration rate due to a reduction in thermal energy.

The rate at which the average particle size would grow from the pessimistically assumed starting point of zero particle size is shown in Fig. 2, with this being driven by both agglomeration and deposition within the SWTC cavity and showing a maximum in the volume average radius being reached before subsequent reductions in

size. This occurs because deposition rates are weighted towards increasing particle sizes so that, as time progresses, deposition will tend to reduce the particle size distribution in the SWTC cavity, as well as reducing the volume fraction which reduces the agglomeration rates. This process occurs beyond the maximum in the volume average radius. Below the maximum value, the deposition is not sufficient for the reduction in volume fraction to reduce the agglomeration.

Results from drop tests⁵ have been used to obtain best estimates for the fraction of particulate <10 µm diameter released into the SWTC cavity. However, no assumptions were made about the particle size distribution <10 µm, as that information was not available, and an extremely pessimistic assumption was made that all particles were of zero particle size when released from the waste package. Typical release fractions from waste packages into the SWTC cavity are of the order of 10^{-6} to 10^{-5} under the severe impact and fire tests representing accident conditions of transport, and it can be seen from Fig. 1 that the DFs corresponding to these release fractions are between 10^3 and 2.5×10^3 and this leads to values of between 10^{-9} to 4×10^{-9} for the fraction of the contents of the waste packages that is released from the SWTC. Given that the permitted release under accident conditions of transport is one A_2 per week, this means that the permitted activity within waste package(s) carried in the SWTC is of the order of $10^9 A_2$.

If a starting particle size greater than zero was assumed, there would be an increase in the DF that would be consistent with the DF increasing with increasing release fractions into the SWTC cavity due to increasing agglomeration rates. The degree of pessimism in choosing zero initial particle size can be seen by the DF increasing by a factor of between 1 and

100 by taking an initial particle size of between 1 and 10 µm.

Conclusions

This paper sets out a methodology for the calculation of releases of waste material in the form of gas-borne particulate through leakage via the lid and valve seals of an SWTC under normal and accident conditions of transport. The separate methodologies for calculating releases in either case are similar but there are important differences which reflect the way fine particulate are released into the container cavity. Both methodologies illustrate the important role that particle size plays in determining both types of release.

While this analysis has been carried out specifically for the SWTC carrying waste packages, it is applicable to other arrangements and its use would reduce the high degree of pessimism used in typical containment assessments, while still giving conservative results.

References

1. 'Regulations for the safe transport of radioactive material'; 2005 edn, Safety Standards Series no. TS-R-1, IAEA, 2005.
2. S. K. Friedlander: 'Smoke, dust and haze: fundamentals of aerosol dynamics', 2nd edn; 2000, Oxford, Oxford University Press.
3. C. Thornton and Z. M. Ning: 'A theoretical model for the stick/bounce behaviour of adhesive, elastic-plastic spheres', *Powder Technol.*, 1998, **99**, (2), 154–162.
4. M. W. Reeks and D. Hall: 'Kinetic models for particle resuspension in turbulent flows: theory and measurement', *J. Aerosol Sci.*, 2001, **32**, 1–31.
5. F. Lange, O. Martens, O. Nolte, H. Lodding, W. Koch and E. Hörmann: 'Testing of packages with LSA materials in very severe mechanical impact conditions with measurement of airborne release', *Packag. Transport Stor. Secur. Radioact. Mater.*, 2007, **18**, (2) 59–71.