Clearance Measurements prior to the Shut-Down of ERAM - 8230

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ABSTRACT

Low and intermediate level radioactive waste was emplaced in the Morsleben Repository (ERAM), a former salt mine, until 1998. Now the German Federal Office for Radiation Protection (Bundesamt für Strahlenschutz – BfS) has applied for the decommissioning of the plant. The ERAM's own radioactive waste requires clearance as far as it may not remain in the repository.

The German Mining Law also demands all non-mining typical waste to be removed from the mine when being decommissioned and the Waste Avoidance, Recovery and Disposal Act demands waste to remain within the economy cycle if this is economically justifiable.

Therefore new clearance measuring devices have to be applied using gamma and beta radiation to determine the contamination of objects and substances regarding the special nuclide spectra in the ERAM. Even if a very multifaceted radionuclide spectrum is to be expected due to the fact that the emplaced waste results from various applications such as nuclear engineering, medicine, research, and military, numerous investigations have confirmed that only a few radionuclides have to be considered. To facilitate this procedure the controlled area of the ERAM is to be classified according to different contamination categories. Assumptions about the radionuclide mixture are permanently controlled by routine measurements and the evaluation of possible contamination events.

The considerations about the radionuclide mixture and the structure of the controlled area will be described in this presentation. Furthermore a new methodology to measure Sr-90 is discussed which uses the Cerenkov effect and the low level of background gamma radiation in the salt mine.

INTRODUCTION

The Morsleben repository (Endlager für radioaktive Abfälle Morsleben – ERAM) is an abandoned salt mine located in the Federal State of Saxony-Anhalt. It was licensed by the former German Democratic Republic. After German reunification the German Federal Office for Radiation Protection (Bundesamt für Strahlenschutz – BfS) became responsible for the ERAM. From 1971 to 1998, 36,754 m^3 LLW and ILW and 6,623 spent sealed radiation sources with a total activity of 520 TBq (reference date end of 2005) were disposed of in the ERAM. Now, BfS has applied for the decommissioning of the plant.

During the decommissioning phase the ERAM will produce radioactive waste of its own until the repository will be finally sealed. In the early decommissioning phase this waste may be disposed of as radioactive waste. Once the emplacement areas are closed or in cases where a recycling or reuse of the material is preferable a clearance procedure is necessary when such material is going to leave the repository.

This material will be released as non-radioactive material and be deposited as conventional waste. In order to be released it is subject to either clearance according to § 29 of the German Radiation Protection Ordinance (RPO) [1] or contamination control according to § 44 RPO. An alternative to clearance is the so-called removal which is described in [2]. Of course, this waste is, if at all, only weakly contaminated and, therefore, represents a special challenge to clearance measurements.

There are both technical and legal reasons for the necessity to remove radioactive or potentially contaminated substances from the repository, which seems to be paradoxical at first sight. The most important technical reason is based on the nature of the former rock salt mine. The rock salt forms a barrier around the emplaced radioactive waste. This barrier may be damaged by gas-generating substances, depending on the amount of gas generated. Therefore, gas-generating substances that cannot remain in the ERAM as a result of safety assessments must be removed from the emplacement areas before they are backfilled. This also applies to substances which may release gases in the post-closure phase. Apart from organic substances, metals are therefore concerned, too.

The German Mining Law points in the same direction. It demands all non-mining typical waste to be removed from the mine when the mine is decommissioned. Furthermore, the German Waste Avoidance, Recovery and Disposal Act (Kreislaufwirtschafts- und Abfallgesetz, KrW-/AbfG) demands that waste remains within the economy cycle if this is economically justifiable. Otherwise it must be deposited. For waste resulting from practices as stated in the Radiation Protection Ordinance clearance is required in any case.

CLEARANCE AND BOUNDARY CONDITIONS

Legal conditions and the radionuclide spectrum

To give consideration to this task the permanent operating license for the ERAM as of April 22, 1986 was modified within the scope of its 27th amendment. Accordingly, a clearance measuring device can be applied using gamma radiation to determine the contamination of objects and substances. The previous regulations allowed only for surface contamination measurements. For objects to be cleared according to § 29 RPO or released from the controlled area according to § 44 (3) RPO, the regulations set out in Annexes III and IV to the Radiation Protection Ordinance will now be applied instead of the limits stipulated by the permanent operating license, including the 10 % rule for radionuclides which are negligible in summation (cut-off criterion). For mass and surface specific contaminations the respective sum rules are given as follows:

$$\sum_i \frac{C_i}{R_i} \leq 1 \text{ and } \sum_i \frac{A_{s,i}}{O_i} \leq 1 \qquad \text{(Eq. 1)}$$

C_i is the mass specific activity (Bq/g) and $A_{s,i}$ the surface specific activity (Bq/cm^2) to be cleared while R_i and O_i are the respective clearance values and i indicates the respective radionuclide. According to the cut-off criterion mentioned above, those radionuclides i can be neglected that do not add more than 10 % within the sum of equation 1. The clearance values R_i and O_i apply to both, § 29 and 44 of the Radiation

Protection Ordinance. However, for a clearance procedure according to § 29 RPO a separate permit from the competent authority is necessary.

These extended options of the 27th amendment take into account the special nuclide spectrum which can be found in the ERAM. Due to the operational history of the ERAM a large variety of radionuclides may be expected from the emplaced waste resulting from various fields of activity, such as nuclear engineering, medicine, research and the military. It is also noteworthy that different disposal techniques were tested and applied bearing different risks of contamination. For this reason, numerous investigations were performed. Finally, it could be confirmed that only a small number of radionuclides needs to be considered for clearance and release measurements.

To this end, gamma spectrometric measurements were performed at a number of samples from the ERAM which are representative with regard to possible contaminations. The samples included material deposited on exhaust air paths, in waste water systems and in tanks for liquid radioactive waste as well as traces of contaminations detected. Some of the samples were then analyzed with the help of supplemental radiochemical measurements since beta and electron capture emitters cannot be detected with gamma spectrometric methods. Nevertheless, they may considerably contribute to the level of radiation within the summation criterion as given in equation 1.

Our investigations have shown that only the radionuclides Co-60, Sr-90, Cs-137 as well as Eu-152 and Eu-154 would be relevant for clearance and release measurements. The remaining radionuclides always summed up to less than 10 % according to the cut-off criterion and can therefore be neglected. This is illustrated in Table I.

The following condensed summation formula shall therefore be applied to clearance measurements aimed at proving that the limits set out in Annex III, table 1, column 5 of the RPO are respected:

$$2 \cdot C_{Cs-137} + 10 \cdot C_{Co-60} + 0.5 \cdot C_{Sr-90} + 5 \cdot (C_{Eu-152} + C_{Eu-154}) \leq 1 \text{ Bq/g} \qquad \text{(Eq. 2)}$$

Since it could be shown that the activity of Sr-90 was, except in very few cases, lower than the activity of Cs-137, we can assume conservatively for gamma measurements which cannot detect Sr-90 that the activity of Sr-90 equals the activity of Cs-137. With this assumption, the summation formula can be condensed even further and is now:

$$2.5 \cdot C_{Cs-137} + 10 \cdot C_{Co-60} + 5 \cdot (C_{Eu-152} + C_{Eu-154}) \leq 1 \text{ Bq/g} \qquad \text{(Eq. 3)}$$

The assumptions relating to the existing radionuclide vector are constantly verified with the help of routine measurements and through the evaluation of possible contamination events.

Table I. Selected Measuring Results of Mass-Specific Contaminations

Radionuclide	Activity Concentration, mBq/g			
	Clearance Value	Deposition on Old Exhaust Air Path	Deposition in Old Specialized Waste Water System	Contamination above Room 3
Co-60	100	65 (0.65)	252 (2.52)	925 (9.25)
Cs-137	500	361 (0.722)	704 (0.141)	6,328 (12.7)
Cs-134	200	2 (0.01)	5 (0.025)	21 (0.105)
Eu-152	200	8 (0.04)	242 (1.21)	31 (0.155)
Eu-154	200	3 (0.015)	149 (0.745)	22 (0.11)
Eu-155	8,000	N/A <3 (< 0.001)	7.5 (< 0.001)	N/A < 30 (< 0.004)
Sr-90	2,000	30 (0.015)	66 (0.033)	4,840 (2.42)
Ni-63	300,000	74 (< 0.001)	17 (< 0.001)	N/A
Fe-55	200,000	45 (< 0.001)	200 (< 0.001)	N/A

Values in brackets: factor in relation to the clearance value according to Annex III, table 1, column 5 of the German Radiation Protection Ordinance.

N/A: not available; in case of gamma spectrometry: < detection limit.

Segregation of Radiation Protection Areas

The permanent operating license, which has recently been modified by the 27th amendment, stipulates that there are no supervised areas in the ERAM. Only controlled areas and exclusion areas are present. It must be noted, however, that the ERAM's exclusion areas do not correspond to exclusion areas as defined by the Radiation Protection Ordinance. In the permanent operating license the term "exclusion area" is used differently in terms of a restricted access with special demand on radiation protection measures. Whereas the Radiation Protection Ordinance requires an exclusion area in cases where the local dose rate may exceed 3 mSv/h, a value of 10 µSv/h is applied in the ERAM.

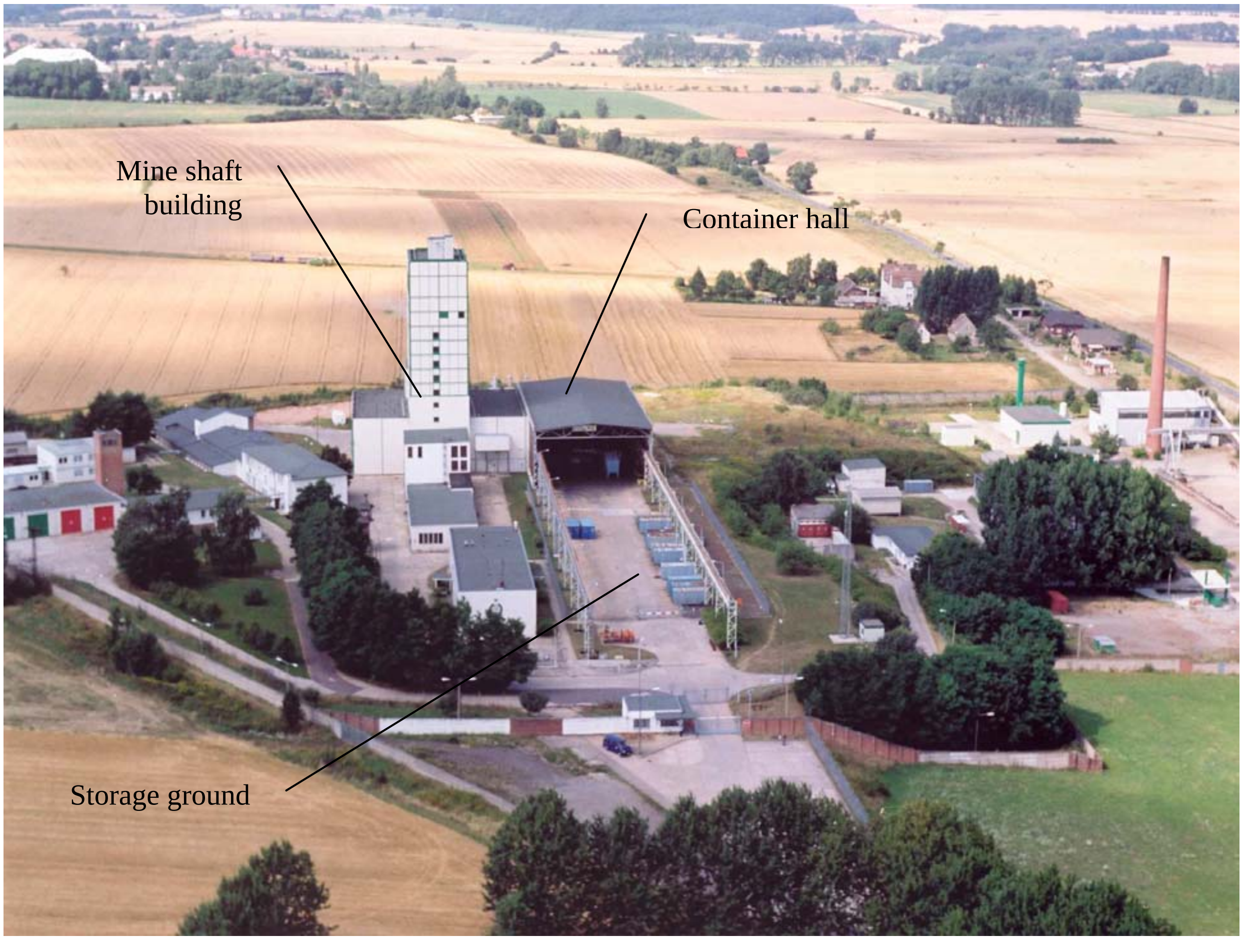

Fig. 1. Aerial view of the above surface installations of the ERAM with controlled area

In order to facilitate clearance and release measurements the controlled area of the ERAM is to be classified according to different contamination categories. The segregated areas are largely characterized by the prevalent local dose rates delivered by the emplaced waste. The controlled area above ground comprises

- the fenced open-air storage ground,
- the handling installation for binding material,
- the container hall and
- parts of the general purpose building (mine shaft building), such as the active laboratory, the mine by-pass, the active workshop and the personnel lock.

Figure 1 shows an overview of the above surface installations. The controlled area below ground comprises

- the lower level of the conveyor cage,
- the mine shaft and
- the 4th level (emplacement area) including the levels below (see Figure 2).

The three emplacement rooms in the Southfield are examples for exclusion areas according to the definition mentioned above.

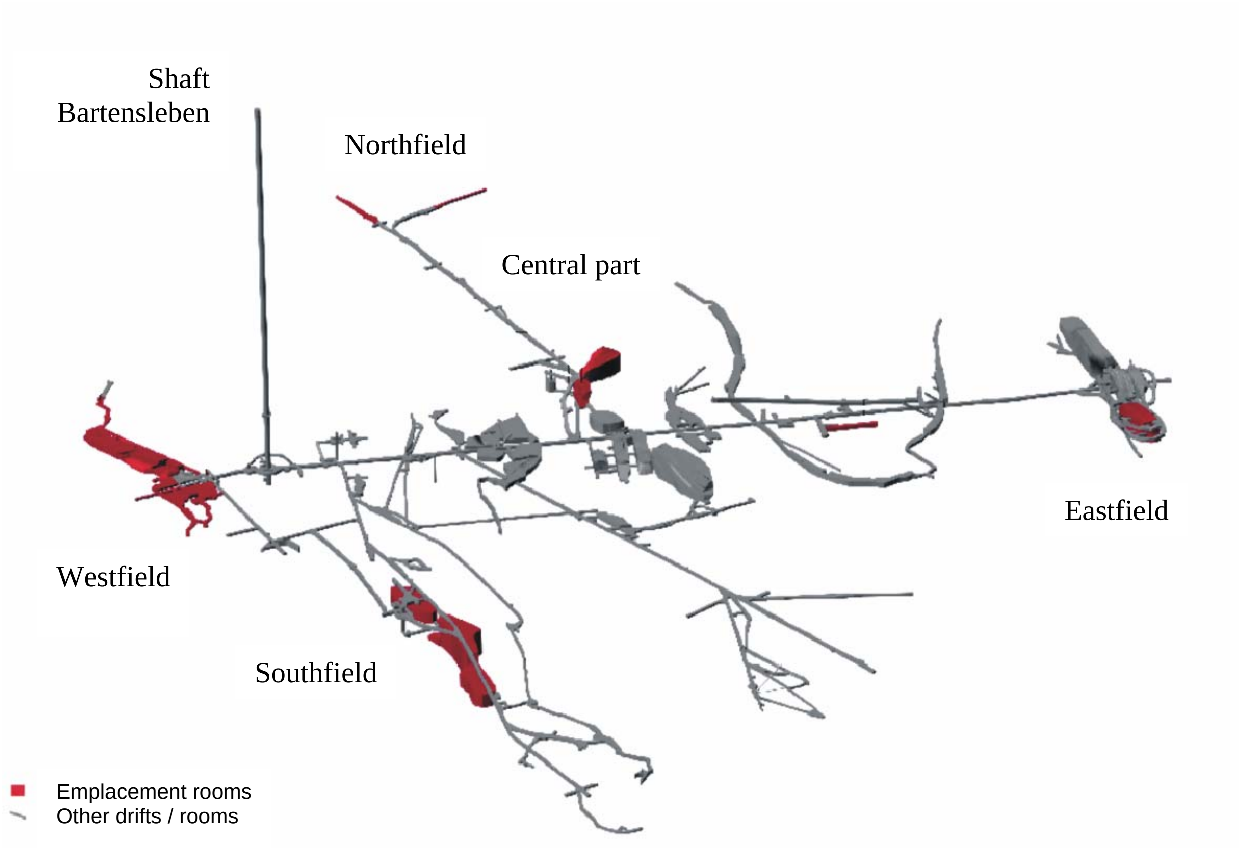

Fig. 2. Emplacement rooms and the 4th level (controlled area) in the ERAM

The emplacement of waste delivered by external waste suppliers was abandoned in 1998. The waste emplaced has largely been backfilled. Today only such waste is handled which is produced in the ERAM itself and which is supposed to be transported to the western field for disposal. A contamination risk thus only exists for the handling and transportation of the remaining waste. Even if the current controlled area on the 4th level does not need to remain in its original limits from the radiological point of view, a considerable effort and numerous clearance measurements would be required to cut back the controlled area below ground. Considering the envisaged decommissioning of the ERAM, it does not appear reasonable to implement such a measure for the remaining period of operation of the repository.

The implementation of the 27th amendment of the permanent operating license shall make it possible to divide the controlled area into two categories for which different requirements need to be respected. These requirements shall apply to both clearance measurements according to § 29 RPO and measurements governing the release of objects according to § 44 RPO. Contamination category I shall include such areas in which contaminations are extremely unlikely and where no hints for a spreading of contamination are given. This needs to be proved taking into account the operational history. Contamination category II shall comprise permanent exclusion areas and areas in which open radioactive sources are handled, such as the active laboratory above ground. If required, contamination category II can be assigned to certain areas for an indefinite or a limited period of time.

Clearance, Removal, and Release

In the ERAM clearance, removal, and release of objects or substances from the controlled area (contamination control according to § 44 (3) RPO) can be applied. These terms are defined in the following paragraphs. The procedures described are well established in the field of nuclear engineering, in particular in the context of decommissioning projects [2].

1. Clearance according to § 29 RPO is an official act which results in relieving a radioactive substance or movable object from the obligations set out in nuclear law in Germany. A clearance is required if two conditions are fulfilled: Firstly, the substance or object is activated or contaminated, and secondly, it stems from practices as defined in the Radiation Protection Ordinance and is therefore subject to the obligations set out in nuclear law.

2. The removal has to be distinguished as a subdivision of a clearance procedure which operators of installations are allowed to perform on their own responsibility provided that the substance or object is not activated or contaminated as defined in the Radiation Protection Ordinance. For this procedure the term "removal" has been established in practical applications.

3. In the case of a release of objects from a controlled area, contamination control is mandatory according to § 44 (3) RPO irrespective of the above criteria. After such contamination control has been performed, the substances or objects may be used without restriction provided that they do not stem from practices as defined in the Radiation Protection Ordinance. Objects stemming from such practices, however, continue to be subject to the obligations set out in nuclear law and may only be used with the relevant restrictions.

Vehicles which have been constantly employed in the ERAM may serve as an example for objects which may be subject to a clearance or removal when they are no longer utilized, as well as machines and parts of machines which have been used in the controlled area within the limits of the license. Contamination control according to § 44 (3) RPO is, for example, required for tools and machines owned by subcontractors which have been employed in the ERAM for a limited period of time and for other objects which may be reused such as cable reels.

Measuring Methods for Clearance Measurements

Except for Sr-90, the radionuclides to be considered can be well detected with regard to their mass and surface specific clearance values by measuring their gamma radiation. Due to the high decay energy of its daughter nuclide Y-90, Sr-90 can be easily detected with the help of liquid scintillation and can be differentiated from the other beta emitters by using pure water instead of the scintillator liquid, making use of its Cerenkov effect.

It is intended to measure materials, provided that their volume is appropriate, with a completely shielded clearance measuring device, which encloses the medium to be measured on all sides with plastic scintillators. Such clearance measuring devices are described in the German standard DIN 25457 "Activity measurement procedures for the clearance of radioactive residual substances and parts of nuclear installations" and are customary in nuclear engineering.

The clearance measuring device will be calibrated for use in the ERAM to detect values exceeding the criterion given by the summation formula (2) irrespective of the actual radionuclide spectrum and its composition provided that the Sr-90 activity does not exceed the Cs-137 activity. The measurement period will be defined such that the device reports a significant measuring value (probability of error of first type and second type is < 5 % each) if the result of summation formula (2) reaches 10 % of the maximum value of 1 Bq/g. This procedure exceeds by far the requirements for the detection limit set out in the Radiation Protection Ordinance. Since the above criterion might be required for a removal later on, however, this is the only way to confirm that an object or substance is not activated or contaminated.

The favorable conditions, in particular the low level of background radiation in the mine, generally allow to reach very low detection limits in the ERAM. Yet there may be other problems with applying such measuring devices in the salt mine, such as the corrosiveness of the air due to its high salt content. Currently investigations are carried out in order to find out if it is practicable to install the clearance measuring device below ground.

In the ERAM gamma-ray spectrometers for in-situ measurements are available for materials which cannot be examined with the clearance measuring device because of their large volume. This method has already been tested on areas above ground and for the segregation and exemption of smaller parts from the controlled area.

In the case that routine supervision or contamination events lead to suspect a contamination with Sr-90, additional measurements shall ensure that the contribution of Sr-90 does not produce values exceeding the limits given by equation 1. The extent and type of measurements will be individually determined for the particular case.

Besides the procedure of beta measurements with proportional counters (direct measurements and evaluation of wipe tests), a method is currently elaborated to measure Sr-90 specifically without radiochemical analysis. Due to the high decay energy of its daughter nuclide Y-90, Sr-90 can easily be detected with the help of liquid scintillation spectrometers and can be distinguished from other beta emitters by using pure water instead of the scintillation liquid and exploiting the Cerenkov effect. Lower-energy beta emitters do not contribute to the Cerenkov radiation.

The detection sensitivity of this method, however, is limited by the background effect, which is affected by the so-called quenching from the substrate used for the wipe test. Quenching describes the impact on the measuring signal from the extent to which the photons produced through the Cerenkov effect are absorbed depending on the level of staining of the substrate. A high level of staining reduces the light output and thus the measuring signal. Yet this also applies to the measuring effect produced by the background radiation.

The Cerenkov method was used in test measurements under the conditions prevailing in the active laboratory above ground. For the above reasons, a stained, uncontaminated substrate from a wipe test yielded a lower measuring signal which corresponded to an apparently negative activity of Sr-90 of up to 3 Bq on the filter as compared to an unused substrate. Thus, up to now, this procedure can only be used to detect Sr-90 contaminations of more than 0.3 Bq/cm², if the removal factor is assumed to be 10 for a wiped surface of 100 cm².

Current work is focussed on improvements to this measuring procedure by reducing the above explained effect. With respect to the conditions of use in the ERAM there are three options to be investigated:

- Performing the underground measurements at places with a lower level of background radiation;
- Reducing the quenching effect by adding bleaching agents;
- Determining the quenching effect through measurements with an external source of sufficient gamma-ray energy

The objective is to make the Cerenkov method a suitable tool for the specific determination of Sr-90 both in routine investigations and in clearance measurements.

SUMMARY

In advance of the decommissioning of the ERAM it becomes increasingly necessary to release potentially contaminated substances or objects from the controlled area or to perform a clearance according to § 29 RPO on this material. In order to create the required boundary conditions the permanent operating license for the ERAM was modified in its 27^{th} amendment. Under the new terms it is now possible to segregate the controlled area into areas of different contamination categories and to obtain a clear structure with regard to the contamination potential and the radionuclide vector of the ERAM. This structure shall be based on simple regulations and routine measurements. Contamination controls according to § 44 RPO and clearance measurements according to § 29 RPO with the help of a clearance measuring device can now be performed more easily and will provide a higher level of detection certainty. In order to ensure proper detection of contaminations which cannot be detected with gamma-ray measurements, such as Sr-90, a multi-stage procedure of evaluating contamination events, routine measurements, as well as measurements on materials subject to clearance or release is envisaged. The instruments and procedures to be applied in this process are subject to continuous development.

With regard to measuring, there are particularly favorable conditions in the areas of the rock salt deposits of the ERAM as a result of the lacking cosmic radiation. Thus, better detection limits can be achieved for the total gamma measurements and also for the aforementioned Cerenkov measurements. Due to the conditions in the salt rock, non-stochastic influences of the zero effect are reduced as well, such as the effect of self-shielding and quench effects.

REFERENCES

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