Optimization of Material Recycling During the Decommissioning Process of Nuclear Power Plants – 16123

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ABSTRACT

The German Government decided in 2011 to phase out nuclear power. Therefore, in the near future there will be several nuclear power plants queued up, in parallel, for dismantling. In nuclear decommissioning there is no standardized procedure available, so that a high level of optimization potential can be assumed. We present the results of a preliminary study, in which the optimization potential of metal recycling during the dismantling process should be estimated. The project can be roughly divided into four sections:

- Determination of the metal inventory of an exemplarily selected plant
- Analysis of the recycling routes of scrap metal fractions and assessment of the potential for optimization of each process step during the dismantling
- Conduction of activation calculations to uncover the potential of interim storage allowing for radioactive decay
- In collaboration with the Institute of Process Metallurgy and Metal Recycling at the RWTH Aachen, appropriate process steps and process chains of an optimized metal recycling for each of the resulting scrap metal fractions are identified.

The paper and the presentation summarize the status and the results of the project implementation and will address for the optimization potentials for decommissioning projects.

INTRODUCTION

With the amendment of the German Atomic Energy Act, the termination of the peaceful use of nuclear energy has been decided. Thus, the existing 17 operational power reactors will be finally shut down and transferred into the decommissioning process within the next 11 years. An interesting question is, in what extent the valuable metals of strategic economic importance can be recycled within the scope of decommissioning. In this respect valuable bulk metals like copper, aluminum and lead, but also rare metals such as indium, niobium, vanadium, cobalt, or tin and rare earth metals are of particular interest. Due to the high demands on the components installed many valuable materials are used in nuclear power plants. It is therefore of particular interest to increase the recycling rates in the nuclear decommissioning and dismantling.

Individual components follow specific paths in the dismantling process, which end up in clearance or disposal. Depending on the decommissioning strategy this paths may be influenced at various processing points, so that the releasable portion of the corresponding material is maximized. This optimization potential is to be assessed in the course of this preliminary study for a typical PWR.

The work plan for the pre-investigation is scheduled for 18 months and can be summarized as follows:

- Acquisition of design, operational, and material data
- Data from a sample facility: detailed specification of used components, material composition applied and data needed for activation calculations, fluence-values and contamination
- Setting up a database to assign metals and components with additional data like activation and decay time possibly needed, concentration, distribution, total mass and recyclability
- Determination of the activity distribution to evaluate the activity level of the component for processing and recyclability, thus: preparation of a sophisticated neutron transport model for the calculation of neutron fluence and subsequent activity analysis
- Classification of recyclability considering the following criteria: total mass, concentration, distribution in structural material, and extent of impurities

DETAILED APPROACH

At the start of the process, the assumption is that the system is in new condition in order to determine the overall potential for an optimized scrap metal recycling after decommissioning. The determined total inventory is then gradually reduced based on various criteria such as activation, contamination, recyclability according to the state of the art, and economy.

In the first step, the entire inventory of metallic components is determined. For this purpose, the system is considered component-wise starting from the RPV to the outside in order to quantify the material content later on. In parallel, the contamination of components in the controlled area which are either exposed to the neutron flux or directly connected to such components, so basically components of the primary circuit and auxiliary circuits, is qualitatively assessed based on averaged empirical data. Furthermore, the current best practice of the dismantling process and the disposal of residuals is determined to finally make a comparative assessment of the recovery rates with the actual potential.

To sum up, the work processes within the preliminary study is described as follows:

- Data inquiry with power plant operators and dismantling companies
- Creation of a component dependent database
- Performing Monte Carlo simulations to determine the activity distribution in the reactor building
- Identification and analysis of the recyclability of relevant components under confinement of technically feasible recovery rates
- Final evaluation of the economic potential of an optimized dismantling

DATA ASCERTAINMENT

In the course of the preliminary study, a typical light water moderated pressurized water reactor of the power class 1300 MW_{el} was selected. Material flow balances of related metals were collected in two different ways. On the one hand, the power plant has been divided into different areas, whose components were individually

analyzed for the contained metals or alloys. Subsequently, the researched materials were combined into scrap metal fractions which are steel, high-alloy stainless steels, aluminum, copper / non-ferrous metals, nickel and lead. This classification is metallurgically motivated based on the value of the substances and the associated melting processes.
In another data query balance sheets of metallic material flows, which occur during the dismantling, were acquired at various dismantling companies [1]. These data are consistent with the research results acquired with the first approach. Fig. 1 exemplarily depicts the material flow of the copper fraction. Values are normalized to be per 1000 kg Copper.

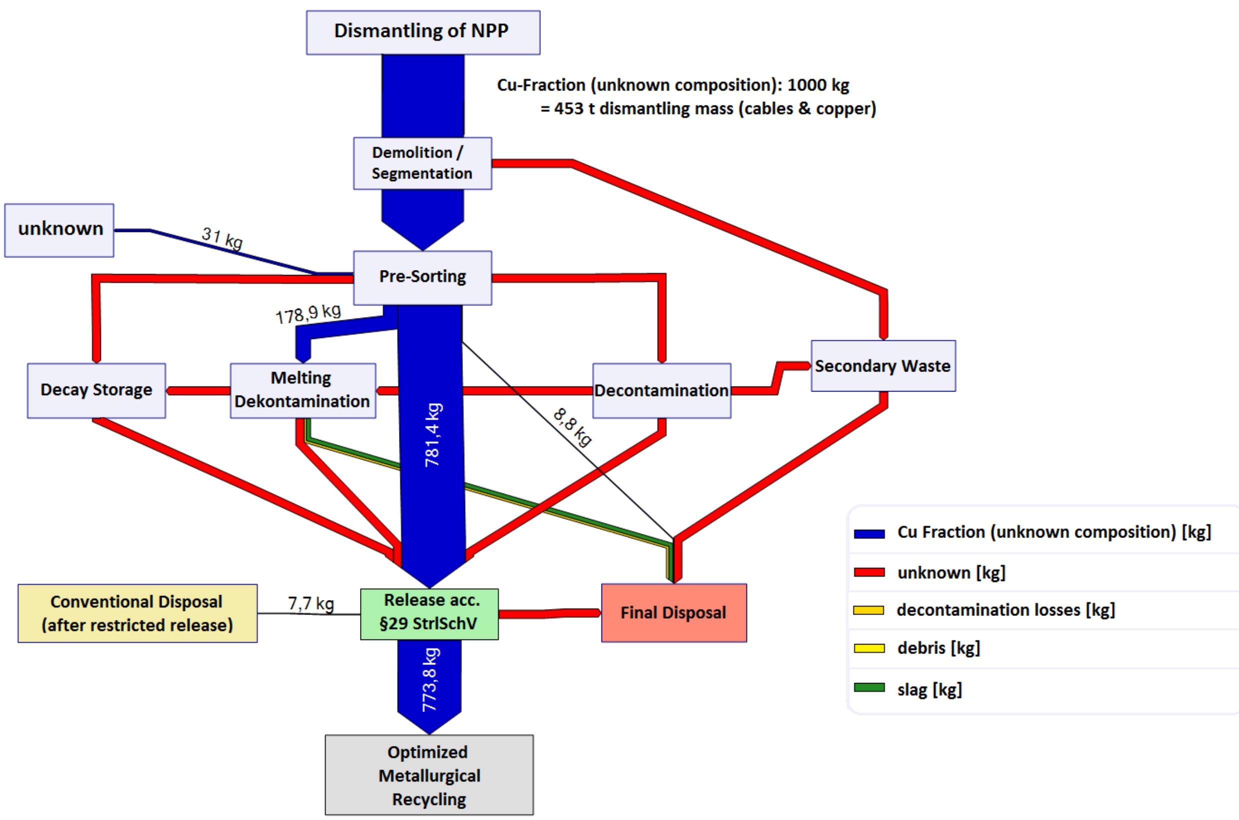

Fig. 1. Material flow chart of the copper fraction without electronic motors and e-waste. Masses are normalized to 1000 kg copper.

ACTIVATION CALCULATIONS

Essential for assessing whether a component is recyclable or not, is the knowledge of the present activity distribution in the reactor. To determine the distribution of the activity the program ORIGEN2, a code for burnup and activation calculation, was used. ORIGEN2 is able to calculate the entire nuclide vector at any time during the operation of the nuclear reactor as well as after the shutdown. Input variables are the isotopic composition of components, their neutron capture cross sections, the absolute neutron flux in the component, and the respective neutron spectrum. Trace elements with a high neutron absorption cross section have to be considered in particular. ORIGEN2 works with the single group cross section, which is the average of the energy-dependent cross section over the entire energy range, and the absolute neutron flux.

In order to take into account the energy dependence of the cross sections and the neutron flux, the single group cross sections must be calculated in advance from the energy-dependent cross sections and neutron spectrum. The single group cross section is isotope-specific and component- or location-dependent. With the absolute neutron flux and number density ORIGEN2 calculates the reaction rate, which describes how many neutrons of each isotope are captured and how strong each component is activated. With this it can be determined which components are actually recyclable and what time it takes in the decay storage.

Simulations by means of Monte Carlo

MCNP (Monte Carlo N-Particle) is a code, developed at the Los Alamos National Laboratories, for analyzing the transport of neutral particles i.e. gammas and neutrons, by the Monte Carlo method. The used MCNP model represents an example of a German Pressurized Water Reactor (PWR). A CAD model of a typical PWR was converted into an MCNP model (see Fig. 2 and Fig. 3) and was herein used for the neutron transport simulations.

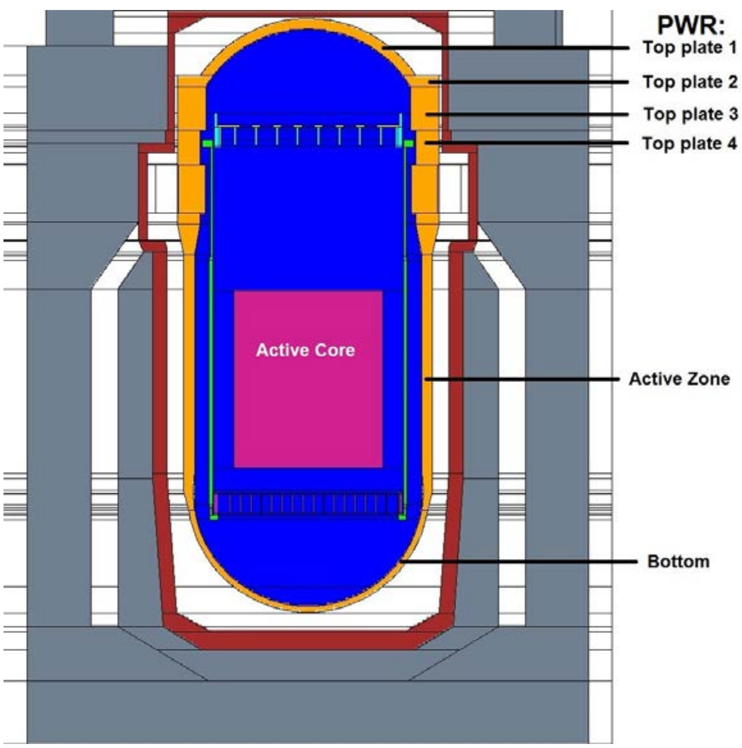

Fig. 2. YZ cross sectional view of the MCNP model.

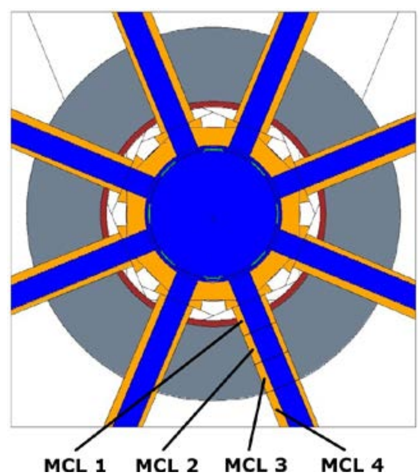

Fig. 3. XY cross sectional view of the MCNP model with main cooling lines (MCL)

Since only the neutron fluxes and activation in the external structures, in particular the pressure vessel wall, are important a homogenized radioactive core was used instead of a detailed heterogeneous fuel-model. This step led to a more efficient simulation of the neutron distribution in the region outside of the core. Here all the volumes and densities of existing materials, such as fuel cladding tubes and water were included into the core and reduced to a single composition in a cylinder.
To further increase efficiency, the variance reduction method FW-CADIS (Forward-Weighted Consistent Adjoint Driven Importance Sampling) of the SCALE framework (Standard Computational Analysis for Licensing Evaluation by Oak Ridge National Laboratory) was used to prepare an optimized weighting map for the neutron transport in the MCNP simulation. This increased the count rate of neutrons in the reactor pressure vessel and the neutron spectrum can be simulated with a smaller relative error. Fig. 4 shows the calculated neutron flux distribution in the MCNP geometry.

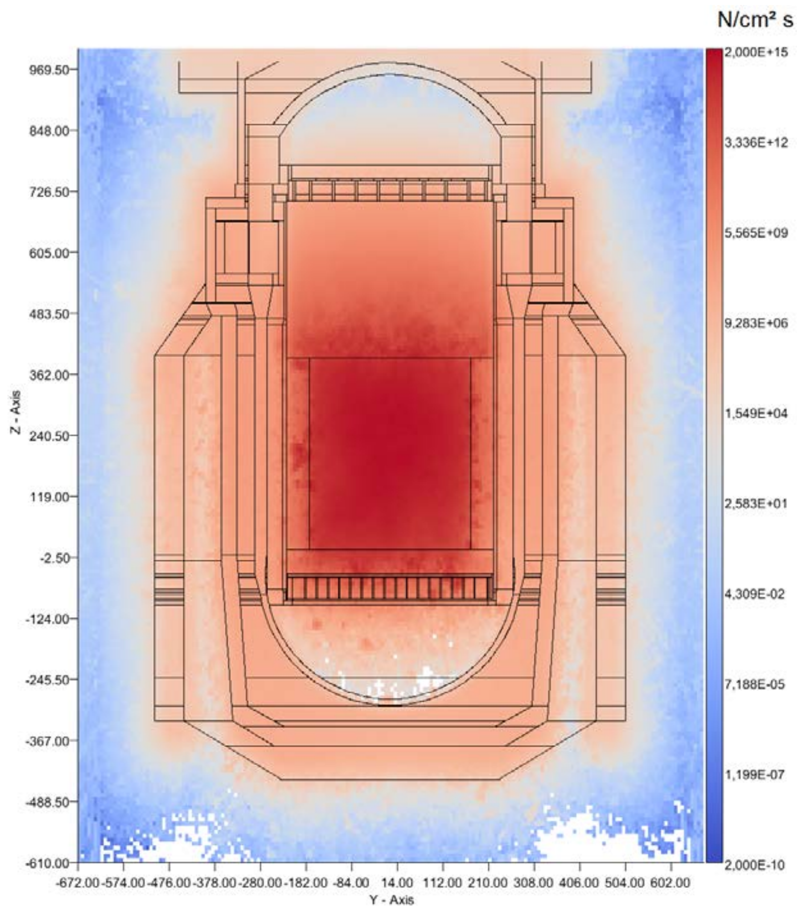

Fig. 4. Total neutron flux in the entire MCNP geometry.

Assessment of the decay behavior
With this neutron transport simulation we were able to determine the neutron flux and spectrum of each component of the pressure vessel. With the neutron

spectrum we generated a one group cross section library for ORIGEN2 and calculated the decay behavior after 32 years operation time of the nuclear power plant. The time for cooldown covers a period of 2 to 150 years. In the following all available limits of the nuclides in the German Radiation Protection Ordinance [2] are considered and compared.

Furthermore, for clearance of a mixture of radionuclides the German Radiation Protection Ordinance stipulates that the sum rule

$$\sum_i {C_i}/{R_i} \leq 1 \quad \text{(Eq. 1)}$$

is satisfied, wherein C_i denotes the specific activity and R_i the respective clearance value of radionuclide i. Once all relevant nuclides are below their clearance values and equation 1 is satisfied, a further utilization of the metal is possible.

Fig. 5 and Fig. 6 show the activity curves for the top plate and the pressure vessel wall on fuel level. Depicted are the main elements for the overall activity and in addition, the sum of the activity and release value ratios.

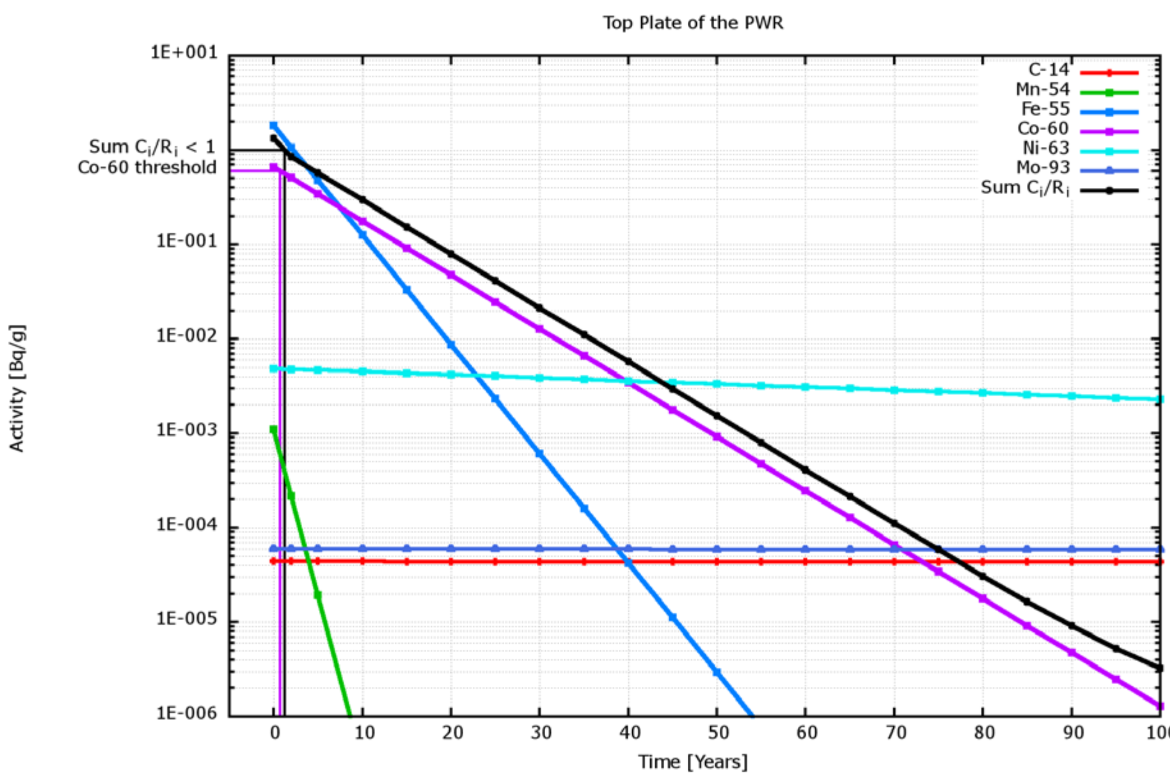

Fig. 5. Activity of the pressure vessel top plate 1, cf. Fig. 2.

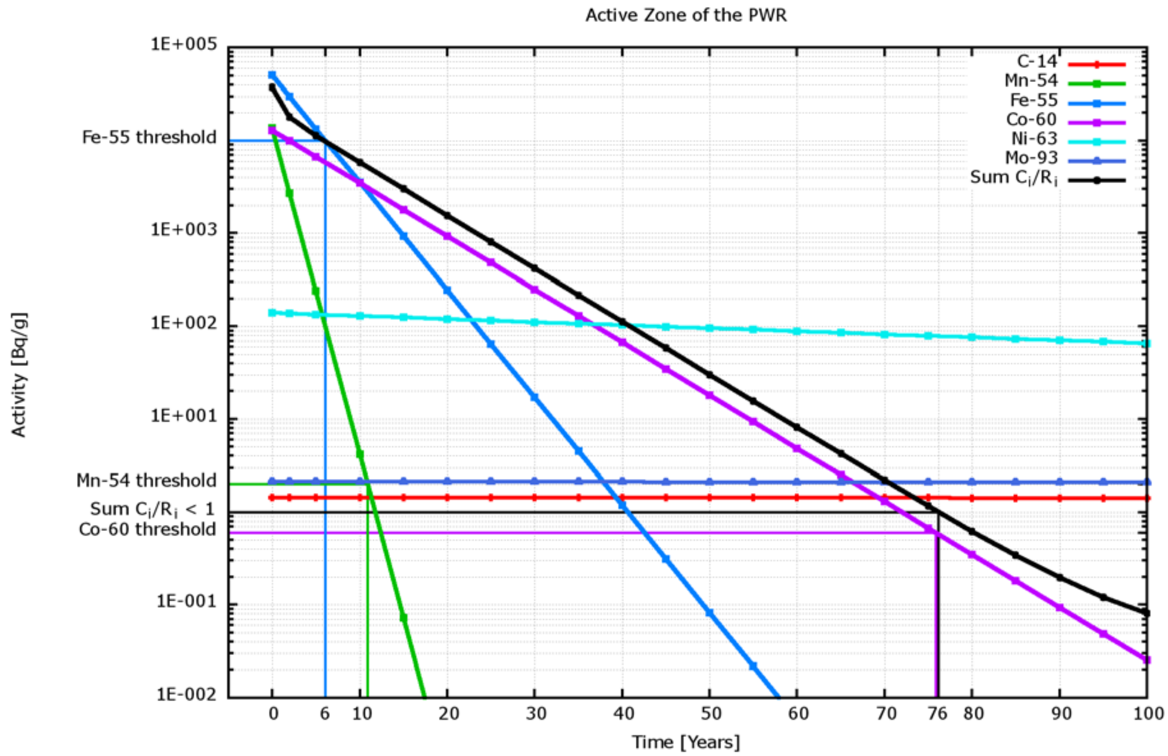

Fig. 6. Activity of the RPV at fuel level, cf. Fig. 2.

The calculated activities are consistent with measurements and calculations performed by the German power supply companies RWE [3]. Masses, Neutron-Fluxes, and decay times for the considered components are summarized in TABLE I.

TABLE I: Mass, Neutron flux and decay time needed for clearance of the RPV and main cooling line (MCL) components labeled according to Fig. 2 and Fig. 3.

Component	Mass [Mg]	Neutron-Flux [n/s cm²]	Clearance after
Top Plate 1	36.8	2.00E+04	1.26 a
Top Plate 2	22.1	1.16E+05	0.00 a
Top Plate 3	76.4	6.08E+05	25.00 a
Top Plate 4	50.3	3.62E+06	37.58 a
MCL 1	1.86	5.89E+06	40.09 a
MCL 2	7.68	1.23E+06	28.19 a
MCL 3	5.55	1.29E+02	0.00 a
MCL 4	5.67	1.14E-01	0.00 a
Active Zone	172.9	9.17E+08	76.19 a
Bottom Plate	37.9	2.46E+07	53.77 a

As expected, the activity is higher in the component closer to the active region of

the fuel zone. The cooldown time of the pressure vessel on fuel level to the potential release for recycling is about 76 years. The base plate can be released after about 54 years.
The part of the RPV, which is located at the level of the main cooling lines, and the first segment of main cooling lines require a cooldown of approximately 40 years. The highest components such as top plate 1 and top plate 2 could be released after about a year assuming a complete decontamination.

QUALITATIVE POTENTIALS FOR OPTIMIZATIONS

From the results of our enquiries, it comes clear that the potentials for increasing the residual material retrieval from the dismantlement of nuclear facilities for recycling are not only to be found in the improvement of decontamination measures and further processing.
Organizational and social aspects must be considered as well in this regard. These are summarized and described in this chapter as 'qualitative' potentials. According to several employees of various disposal service providers for the NPP operators currently exists no coherent strategy for the release or in the process of accumulating residues. Even for conceptually identical systems such as the "convoy fleet" of Siemens, provided as the default type for future reactor systems, a common standard will probably not be realized. The reason for this is the ongoing employment of the power plant staff for decommissioning and a lack of exchange between the power supply companies in decommissioning matters.
This does not mean that experiences from earlier decommissioning projects of the same company will not have any influence on their follow-up projects. Thus one of the objectives stated, to reduce the number of the nuclide to be tested as much as possible.

Waste prevention

An important potential for cost savings through an optimized dismantling process involves the reduction of waste that must be stored in a final repository.
However, economic incentives for safe recycling are not alone the proceeds realizable from the sale of recovered recyclables. Especially the dropping out costs for interim storage, which cannot be reasonably estimated due to the uncertain timing of the commissioning of the Konrad repository, is another significant cost factor. Furthermore, the masses of the metals lost through the disposal decrease, which is advantageous, both from the perspective of sustainability as well as applied financial resources [1, 4].

Decontamination procedures

The previous research revealed few indications that significantly higher recycling rates or waste reduction could be achieved by improved decontamination measures. However, some apparatuses are currently in development that promise a significant improvement in the decontamination, e.g. of the primary side of the steam generator tube surfaces [5].

Very promising in this respect seems the specific combination of chemical and mechanical methods, so that several orders of magnitude lower activity must be reckoned. But the effort necessary for this purpose is expected to exceed the economic benefit for the plant operators by far [4]. According to this fact it is obvious that without legal provisions in this area innovations can hardly be expected.
However, insofar as the efforts are economically justifiable, such combinations are already used [1].

Knowledge of the activity inventory

Experience from previously conducted decommissioning projects as well as the results of our research suggest that large parts of the RPV are not as strongly activated as previously assumed due to the intense radiation exposure and could even partially be released after precedent decontamination [6].
So for example, a part of the lid from the RPV of the BWR Würgassen was placed as an exhibit in front of the administration building after precedent decontamination.
The results of our own activation calculations indicate that, also for PWRs, either an immediate release of individual RPV parts is possible or even components that were exposed to the highest radiation, such as the RPV wall on fuel level, can be released after appropriate interim storage (a few decades) and do not necessarily need to be disposed [3]. A previous decontamination is of course necessary.
Overall, this shows that a precise knowledge of the activity inventory, coupled with exact calculations of the required decay time, can dramatically reduce the repository capacity needed considering absence of surface decontaminations. At least it can be considered that significant financial savings from an overpass in a lower waste class can be considered; either because the radioactivity is far subsided that recycling within the nuclear energy is possible, or because short-lived, heat-generating nuclides are disintegrated far enough so that they can be classified as minor heat developing.
Outside of Germany, this procedure is common practice.
Especially in light of recent developments concerning the delayed start-up of German repositories this provides potential to reduce the cost for waste to be stored. According to the estimate of a contact person the cost for interim storage should be lower by a factor of 2-3 compared to the cost of final disposal [1].

Release of radioactive remnants

The release represents an administrative act of the competent authority and is carried out in accordance with the German Radiation Protection Ordinance (StrlSchV) [2]. Substances of lower radioactivity can be released only when an effective dose of not more than 10 µS per year can occur for members of the public by them [2]. This is considered by the authorities to be met if the conditions laid down in the StrlSchV clearance levels are met. At times, the evidence that the protection will be respected for an intended release path can be performed by a separate process.
The release procedure for radioactive waste metals, as they occur during the

dismantling of the controlled area, was identified to potentially be an essential lever for the clearance measurement, thus for influencing the disposal route, since opportunities to redirect the incurred material flows also approach in the sequence of the individual process steps.
As our research showed, there are several release strategies for each class of material, as there is currently no standardized procedure for this. Accordingly, it is expected that in many cases sub-optimal strategies are pursued and thus a difficult to quantify amount of recyclable material is lost. Choosing the release strategy depends on many factors. Several conversations with employees of the nuclear industry revealed that in Germany it is a direct manner of the location since a clear north-south divide in the intensity of the cooperation with the supervising evaluators can be found. An Example is waste electrical & electronic equipment (WEEE), including portable IT devices, on which various approval procedures could be used and several opinions exist:

- Opinion 1: Printed circuit boards (PCB) are not released for recycling; since the release procedure was too costly (proof of absence of surface contaminations also necessary from under Surface Mounted Device (SMD) components, e.g., capacitors, resistors, ICs, etc.) [1]
- Opinion 2: See no problem in the release of such components; e.g., shredding and mass-related activity measurement are adequate and already practiced [7]

Remarkably about that is that both contacts support their statements by the fact that those contaminations are airborne and therefore homogenously distributed.

Classification in homogeneous material batches

The classification of scrap arising from the dismantling takes place in the following groups:

- Copper
- Non-ferrous metals (bronze, brass)
- Galvanized metals
- Black Metals (Ferrous Metals)
- White Metals
- Black / White Metals austenitic
- Lead
- Aluminum
- Silver & Alloys
- composite material (WEEE)

The assignment is carried out mainly by the operating personnel of the plant based in visual inspection. Furthermore, the magnetic properties are checked for identification.
According to the statements of a contact person the review is carried as follows:
Non-ferrous metals yes / no? If not, then check with a magnet. Magnetic yes: ferrite / no: austenite
By that way of sorting, it may easily lead to confusion (magnetized austenite being identified as less valuable ferrite). Possibly quality materials lose their properties being downcycled, or the plant operator loses higher revenues.

In one case e.g. a demolition worker has incorrectly identified austenite as a less valuable ferrite; the proceeds would have amounted to only 1/9 of the actual scrap metal value.
If the executive staff was specially trained, this source of error could be eliminated; accordingly provides the contact person there is optimization potential within "Sampling / sorting"
It should be forwarded, that the degree of sorting during the dismantling of nuclear facilities greatly depends on the will of plant operators, or the companies contracted. Thus, for example the remnants at the site Würgassen were only separated in steel, non-ferrous metals and plastics, and afterwards handed over to recyclers as mixed fractions. If a finer sorting would have been carried out the sales revenues would have been significantly higher.

Release strategy as a location-based "philosophical question"

Since the operational staff is also used for D&D, it takes part in developing the decommissioning strategy, with the result that the respective "clerk in the decision-making with regard to waste disposal and recycling is often untrained on that field", as a contact Person once stated.
Further examples are the German Konvoi reactors. The design of these PWRs is standardized to a high degree and a common concept for D&D is therefore suggestive. Nonetheless the idea of a common concept for D&D was abandoned due to different dismantling crews and the federal structure of the German licensing law [1, 4].

Review of the term “waste” in nuclear law

The term "radioactive waste" was originally defined according to objective criteria, but with the seventh amending to the Atomic Energy Act (AtG) this objective definition of waste has been transformed in a subjective order. The hitherto required priority of recovery over disposal of residual radioactive material was lost. Specifically, this meant that only those residues that could not be safely recovered, their recycling is not practicable, economically justifiable or incompatible with §1 No. 2 - 4 AtG were declared as waste. After the entry into force of the amendment of primacy priority of recovery was no longer considered.
Since then, it is up to the operators of nuclear fission plants to utilize residual material after decontamination or decide that the radioactive waste is to be eliminated, which corresponds to the direct disposal.
A clear distinction between the terms “residual radioactive material” and “radioactive waste” does not exist; critical here is the plant operator’s will.
The resultant subjective definition of waste should have considerable impact on the quantities of recycled residues. In the eyes of the plant operators’ "merchants" recyclables are only seen as valuables, if profit could be achieved with them. This underlines a statement of an informant. "Even non-performed decontamination measures are mostly result of a lack of awareness to raw material value ". In this context, he noted also that many plant operators expose the old replaced turbine generator "in their front yard,", because they shy away from "disposal costs", not

aware of that components raw material value.

Acceptance shared for recycling steel scrap

Further problems arise from the acceptance of exploiters and landfill operator for released scrap metal. The decreasing willingness to accept released materials is, for example, driven by political campaigns which fan fear of radiation exposure even if the material is not radioactive [8].
This was confirmed by an informant who told us that German recyclers want to pay less than market price for the materials in order to minimize their business risk.

CONCLUSIONS

We have presented the approach and the results to date of a preliminary study which assesses the potential for optimizing the metal recycling during the dismantling of NPPs. A large part of the project encompasses the inquiry and collection of data regarding the components implemented in a nuclear power plant and their material composition. To uncover optimization potentials intensive discussions with power plant operators, dismantling companies, consultants and approval authorities were conducted. The outcome of these discussions is a qualitative list of suggestions, which may engage within the decommissioning process at various points. An intensely investigated aspect is the decay storage of activated components. Monte Carlo simulations and activation calculations for metallic components within the biological shield were conducted. Thus the decay time for each component could be determined after which clearance according to the German Radiation Protection Ordinance is feasible. It was found that after proper decontamination the entire reactor pressure vessel and the main coolant lines can be unconditionally released after a period of approximately 80 years. Large parts of the pressure vessel can be released even much earlier. We find that the dismantling does not follow a standardized procedure and therefore holds potential for optimization at various points. Furthermore, at a higher level, for example, legislation or public acceptance, there are some possibilities to increase the mass flows in the direction of release and recycling or re-use without diminishing the radiation protection of personnel and population.

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