Design concept for maximized use of recycled scrap in the production of storage packages

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1. Introduction

In the decommissioning of nuclear plants large quantities of radioactively contaminated waste metal have to be disposed of. An economic alternative to final storage is the recycling of the scrap metal in the production of transport and storage containers for low and medium active waste made of nodular graphite ductile cast iron. In the particular case of the CARLA plant operated by Siempelkamp, scrap metal with an activity of up to 200 Bq/g is accepted for processing. This covers the vast majority of the metals of a plant to be decommissioned. The composition of the waste metals varies greatly, depending on the different origins like structural or stainless steels

After solidification of the high-carbon, high-silicon cast iron melt, the carbon has formed nodular graphite particles embedded in the metal matrix. Nodular cast iron has high strength and elongation. A further advantage of this material are its good radiation shielding properties.

Fracture toughness is an important material property in the design of containers for final storage. In the particular case of containers that have to meet the specifications for final storage these must withstand accident loadings from a height of up 5 m at temperatures of -20°C without crack initiation. Containers for final storage do not have the benefit of impact limiters. The fracture toughness of cast iron depends primarily on the microstructure of the metal matrix. A ferritic microstructure has a higher fracture toughness than a pearlitic microstructure. Carbides in the matrix lead to further embrittlement.

The metals to be recycled in the decommissioning of a nuclear installation have marked contents of elements like manganese (Mn) in structural steels, chromium (Cr), nickel (Ni) and molybdenum (Mo) in stainless steels and copper (Cu) in special steels. These elements lead to a pearlitic microstructure and to carbides, even at low contents in the melt. With a rising content of pearlite and carbides, the tensile and yield strength increase while elongation to rupture and fracture toughness decrease. In order to meet the requirements of sufficient ductility and fracture toughness, the specifications for the presently licensed containers limits the embrittling content of pearlite in the microstructure to 20% of the cross section of a metallographic specimen. In order to meet this limit on pearlite content, type and quantity of waste metal that can presently be recycled is very limited.

2. Goals and methods

Based on this background the research program FORM (<u>F</u>orschungsvorhaben zur <u>O</u>ptimierung der <u>R</u>eststoffverwertung von <u>M</u>etallen) [1], [3] investigates ways to substantially raise the amount of recycled metals in the production of containers while at the same time meeting the design requirements for transport and final-storage containers. A substantial increase would be doubling or tripling the amount of recycled metals. This requires the investigation of the mechanical and fracture toughness material properties under the influence of those elements that cause pearlites and carbides in the microstructure. In order to achieve this goal, the relations between chemical composition of the casting and manufacturing process (e.g. casting setup and solidification conditions, which influence the material properties of the product) on the one side and the resulting cast iron microstructure and material properties on the other side must be investigated.

As a further goal the design of the containers must also be optimized such as to reduce the stresses resulting from accident loading. This concerns details of the container design whose effects can be simulated in dynamic finite element calculations. By way of optimizing the structural design of the container, the material stresses resulting from a given external loading shall be reduced substantially. With respect to the dynamic precalculations of the full-size container drop tests, required for the licensing for use in final storage, the behavior of the target (solid rock simulated by concrete) as well as the interaction of target and container must be better understood than is presently the case.

Mastering of the casting process and of casting exactly to specification, in order to obtain the specified microstructure and material properties in the production casting, and the verification of material properties by quality control are central parts of the R&D program FORM. The material research will be done on relevant-sized castings followed by series of dummy containers as well as full size prototype containers. Both dummy and prototype containers receive large notches as artificial flaws and are drop tested after extensive precalculations. The requirements in the drop tests include those for the final repository, drops at a temperature of -20°C from heights of 0.8 m and 5 m respectively for Class I and Class II containers. It must be shown that there is no crack initiation under these loadings. The present phase of the research (FORM III) continues the work published under [1] and [3] and is in cooperation with the parallel research project EBER [4] at BAM (Bundesanstalt für Materialforschung und -prüfung) where BAM is responsible (among others) for the fracture mechanics evaluation and the instrumented drop tests.

3. Material investigations

A primary aim of the material investigations concerns metallurgy. For this purpose a large number of test plates were cast with a wide range of chemical compositions of the elements that lead to pearlite and carbides in the microstructure. These test plates had dimensions typical for the walls of cubic containers and MOSAIK®II containers. For a significantly higher recycling level than at present, the amount of pearlite will increase from below 20% to above 80%. This higher pearlite content results in more strength and less ductility. After investigating microstructure and mechanical properties in several positions of the test plates, these data were evaluated. Using the method 'design of experiments' (DOE) relevant information on the influence of chemical composition (impurity element concentration) on tensile strength, yield strength, elongation and content of pearlite and carbides were determined. It is important to know these mathematical correlations in order to create a software for optimizing the charge for melting. This software will be used in future production for reasons of quality-planning and quality-assurance. It must be pointed out that it is not the chemical composition of the impurity elements which will be invariable but rather the required mechanical properties of the material.

Besides the investigation of the microstructure and the tensile testing, the fracture toughness was investigated with three-point-bending specimens taken from selected test plates. Because for the pearlitic microstructure of the material, fracture toughness is at the lower shelf. The influence of the test temperature and the loading rate is relatively small. The measured dynamic fracture toughness of the pearlitic material at -20°C ranges from 20 to 35 MPa√m (Fig. 1). For predominantly ferritic material the fracture toughness under the same test conditions is expected to be >50 MPa√m (up to the present there is only limited information on fracture toughness of ferritic nodular cast iron at low temperature under dynamic loading. New research is under way to close this gap). There is also testing of specimens with machined notches without fatigue cracks. This will help improve the material model for better prediction of the stress level that leads to actual crack initiation in the containers.

Two rings with the same dimensions and setup as for MOSAIK®II containers were cast with two selected chemical compositions. As intended and expected this lead to castings with two differently high contents of pearlite and carbides in the microstructure. Specimens taken from these two rings provided the static and dynamic material properties that could be used one-to-one in the design optimization of the full-size cubic container type VII and the MOSAIK®II container and the precalculations of their drop tests [3], [4]. The figures for graphite shape and size shown in Table 1 indicate high quality nodular graphite, on the same level as for castings without waste metal.

Table 1. Content of waste metal (weight-%) and microstructure in the special nodular cast iron

	Mn %	Cr %	Ni %	Mo %	Cu %	ferrite %	pearlite %	carbides %	graphite shape ISO 945	graphite size
Ring 1	0.3	0.35	< 0.06	< 0.01	0.2	25	75	< 1	VI + (V)	6 - (5)
Ring 2	0.5	0.6	0.5	< 0.01	0.2	5	90	5	VI + (V)	6

The results of the static tensile tests show the high tensile and yield strengths and the low elongation to rupture, which was expected. Because of the carbides, there is a further drop in elongation for the ring 2 material. Under dynamic loading the yield strength rises markedly while tensile strength remains nearly the same. The elongation to rupture takes a further drop. The high yield strengths have the very advantageous effect that stresses in the container material remain in the linear elastic range even under accident loading. Table 2 shows the high strengths, the increases in yield strength and the decreases in elongations under dynamic loading.

Table 2. Mechanical properties of the special nodular cast iron at room temperature as function of loading rate

	Loading rate 1/s	Elastic Modulus GPa	Yield Strength MPa	Tensile strength MPa	Elongation to rupture %
Ring 1, test temperature -40°C	Static: 1	165	404	610	2.8
	Dynamic: 1,25*10^5	174	580	622	1.2
Ring 2, test temperature -20°C	Static: 1	168	452	602	1.5
	Dynamic: 1,25*10^5	168	613	621	0.3

With the high contents of pearlite, the reference container materials of rings 1 and 2 display a typical lower shelf fracture toughness. In ring 2, with carbides due to the higher content of Cr, the fracture toughness is not much lower than in the ring 1 material with much less Cr. The influence of the loading rate also is quite limited (Fig. 1).

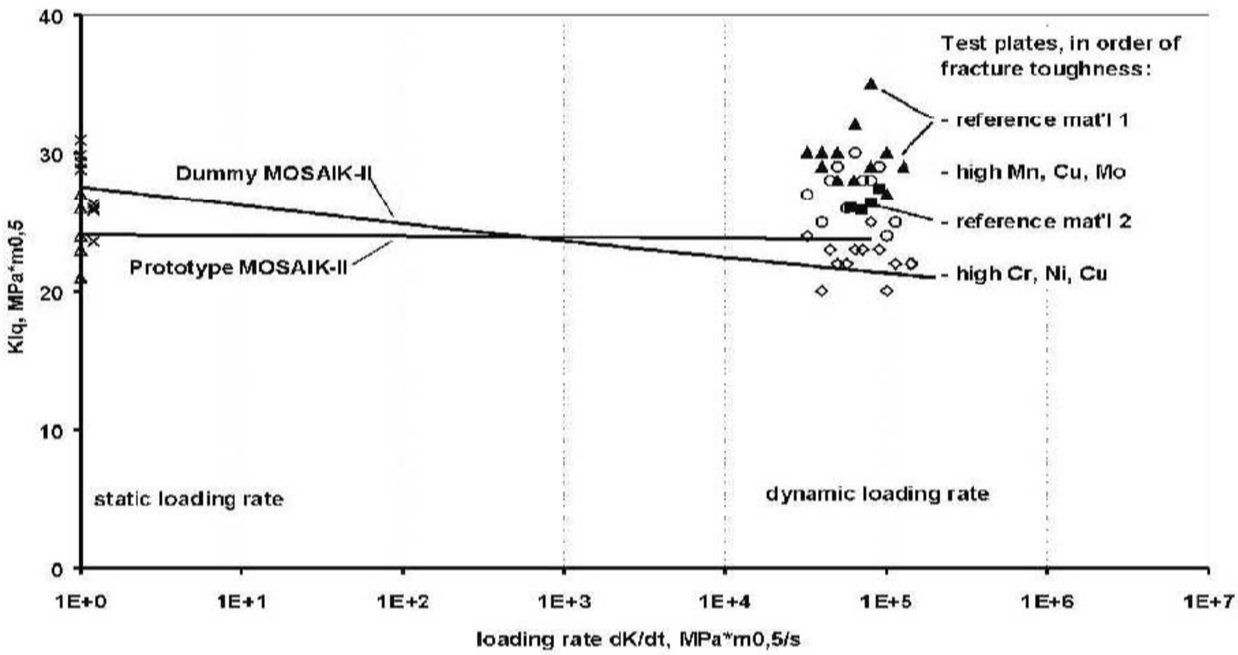
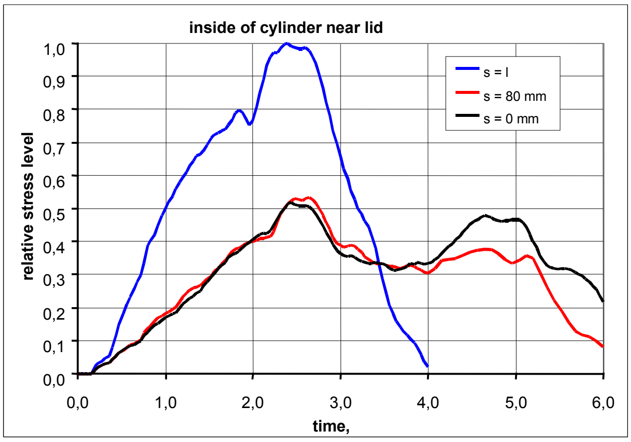


Fig. 1: Fracture toughness (incl. trend lines) at -20°C as function of material and loading rate

Besides metallographic aspects like pearlite and carbide contents in the microstructure, the upper limits of waste metal content are also set by manufacturing considerations like avoiding shrinkage defects and limiting hardness in order to maintain economic machinability.

4. Design concept and design optimization

The design concept calls for substantially lowering the maximum stresses from accident loads by structural optimization and by showing, through material and component testing, that the packages are safe against crack initiation despite their pearlitic microstructure. The structural design of containers was successfully optimized with the help of dynamic finite element calculations. Thanks to increases in the internal transition radii and by use of protrusions (Figs. 4 and 5) that act like built-in shock absorbers, the maximum stresses under impact loading could be reduced. In the case of the MOSAIK®II the highest stresses result from a side drop. For a 0.8 m drop, the tangential stress that governs the design was reduced, after several iterations, by a factor of about 2 from an initial level of 306 MPa to a mere 159 MPa (52 %) after optimization (Figs. 2 and 3). In the case of the box-shaped container type VII it is the flat bottom drop that causes the highest stresses in the structure. In a 5 m drop onto the rock target, the maximum computed deceleration, before optimization, was 1150 g. After optimization the maximum deceleration was reduced to 635 g.



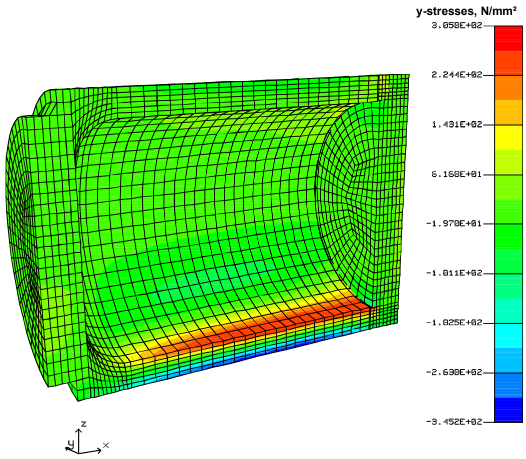


Fig. 2: Tensile stresses in MOSAIK®II container from 0.8m side drop before and after optimization

Fig. 3: Tangential stresses in MOSAIK®II container from 0.8m side drop before optimization

5. Prototype containers and drop tests

To prove the feasibility of pearlitic cast iron containers, series of test components are manufactured. Wall thickness and casting setup are identical to that of full size packages. The instrumented test components receive long and deep machined notches as artificial flaws at locations of high precalculated stress and will be drop tested from different heights onto concrete dummies of final storage targets. Extensive post-test investigation will determine whether crack initiation has occurred. Results will further sharpen simulation tools to predict stress levels and, above all, to predict the critical stress level leading to and the safety factor against crack initiation.

Prototype cubic and MOSAIK®II containers were manufactured with chemical compositions (and thus amounts of inactive simulated waste metal) corresponding to the preceding dummy container castings, rings 1 and 2. Specimens for tensile testing and determining microstructure were taken from cored samples, the standard quality assurance procedure. Microstructure and graphite nodules were found to be in good accordance with those for the two rings. The values for static strength and elongation in the container, measured at room temperature, are somewhat lower than in ring 1, measured at -40°C, which was to be expected. For the MOSAIK®II container the corresponding values are nearly identical to those of ring 2, which is a very good result (Table 2).

Table 1. Content of waste metals in melt and microstructure in the two prototype containers

	Mn %	Cr %	Ni %	Mo %	Cu %	Ferrite %	pearlite %	carbides %	graphite shape	graphite size
Cubic container	0,3	0,3	<0,06	<0,01	0,2	23	75	2	VI + V	5 - 6
MOSAIK®II container	0,5	0,6	0,5	<0,01	0,2	0	95	5	VI + (V)	6 - 5

Table 2. Tensile tests on cored samples from prototype containers under static loading at ambient temperature

	yield strength Mpa	Tensile strength MPa	Elongation %
Cubic container	354	579	4.8
MOSAIK®II container	443	591	1.8

After the drop tests sufficiently large samples can be taken from the containers for fracture toughness specimens. The results are already available for the MOSAIK®II container (Table 3) The results for the cubic containers will have to wait until after a further drop test scheduled later in the R&D program. The fracture toughness has been determined at dynamic loading rates on three-point bending specimens from samples taken from the regions that experience the highest stresses in side drop and bottom drop accidents. In both regions the fracture toughness was found to be higher than in ring 2, which is a reassuring result.

Table 3. Fracture toughness at -20°C under dynamic loading rate

	$K_{Iq,}$ MPa*m^0,5
Ring 2 (dummy container)	20
Wall of MOSAIK®II container	26
Bottom of MOSAIK®II container	23

The recesses in the outside surfaces of the prototype containers, respectively the radial and axial protrusions that act like built-in shock absorbers, can be seen in Figs. 4 and 5. Both containers received machined artificial flaws at locations of precalculated maximum stresses in drop test loading. Flaw size and orientation were selected such as to make applied stress intensity factors K_{appl} equal to the material properties K_{mat} measured on rings 1 and 2. There was no safety factor. The artificial flaws of the cubic container had a depth of 7 mm and a length of 90 mm. The two artificial flaws of the MOSAIK®II container were located at the 6 o'clock and 12 o'clock positions of the cylinder, each with a depth of 5 mm and length of 24 mm. All artificial flaws could exactly be located and their length safely sized by ultrasonic testing. Depth and width were widely overestimated in UT.

Both prototype containers have successfully passed their drop tests at -20°C onto concrete targets simulating final storage bedrock conditions (Fig. 5). The shock-absorbing design optimization did its precalculated work. The low dynamic fracture toughnesses of the pearlitic and carbidic nodular cast iron did not lead to crack initiation as was shown by microscopy of the notches of the MOSAIK®II container. The post-test investigations of the MOSAIK®II container have shown that fracture toughness was somewhat higher than anticipated (Table 3).

Evaluation of the drop test of the cubic container showed deceleration and stresses to have been lower than pre-calculated. Since it can be safely assumed that there was no crack initiation, the cubic container will tested in a second 5 m drop after machining of a new set of larger notches.

Fig. 4: MOSAIK®II container after drop test

Fig. 5: Cubic container after drop test

To improve precalculations of accident loadings, series of drop tests with dummy cubic containers will be used. Two series of dummies have been manufactured, one series without the stress reduction design features, the other with these features included. The two reference materials described above were used. The dummies are 1100mm x 1100mm x 820mm in size and weigh 4 t. They have artificial flaws 3.5 mm wide by 16 mm deep (10% of the 160 mm wall thickness) at locations of highest precalculated stress (Fig. 6). The flaws run the full width of the dummies. The fracture toughness of the dummies is determined on three-point-bending specimens machined from the containers The first set are standard specimens with fatigue cracks. The second set are specimens with the same notches as in the dummy containers. The knowledge of the respective fracture toughness will be applied in the evaluation of the drop tests and for precalculation of the stepwise increased drop heights in EBER by BAM (Fig. 7).

Successful drop tests will be another major argument justifying the use of a less ductile material. They will show the true stress levels in the castings under accident conditions for drops onto the solid rock specified as targets for final storage. The evaluation of the first series of the instrumented drop tests has already helped fine-tuning the simulation of casting and target response to impact.

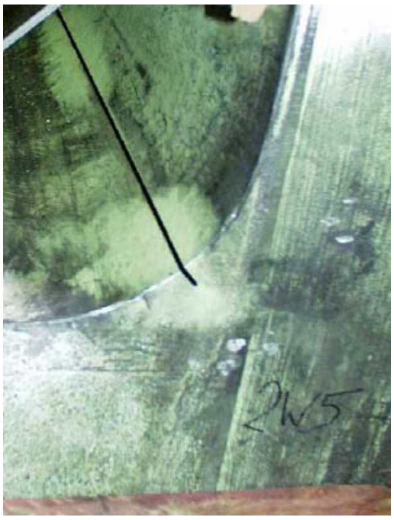


Fig. 6: Notch in dummy container

Fig. 7: Drop test of dummy container (Photo courtesy BAM)

6. Continuation of research and outlook

The preliminary drop tests of dummy containers without artificial flaws will be followed by the two drop test series with notched dummies. The drops will be from ever increasing heights until crack initiation is observed. After every drop test samples are taken at the notches and inspected for any crack initiation. The drop height of the following test, with a new dummy, will be determined in part by precalculations that are upgraded using the measurements of the drop test and of the results from the special bending specimens described above. Based on the testing of the container dummies, prototype MOSAIK®II and cubic containers will be manufactured. These prototypes will also be notched to 10% of wall thickness at those locations where the highest stresses are predicted. Both prototypes will be dropped from heights of 5 m. This will show the safety and reserves of the optimized storage containers. After the drop tests the dummies will be cut up and investigated for any crack initiation and for verification of the in-situ material properties.

7. Conclusions

Full size containers have passed the drop tests required for final storage. It has been shown that the use of pearlitic material is justified. The level of knowledge gained up to this point allows to confidently predict safety against crack initiation of pearlitic Class II MOSAIK®II casks and cubic containers under accident conditions for final storage: 5 m drop at -20°C. The optimization of the container design has reduced stresses resulting from accident loading by a factor of about two. Packages will no longer require ferritizing heat treatment. The quality of non-destructive inspection is not impaired. The amount of nuclear scrap used in the cast iron melts can be raised by an economically relevant factor of about three over the present level.

8. References

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