OPTIMIZATION OF WASTE CONTAINERS MADE OF DUCTILE CAST IRON BY USING RECYCLED SCRAP FROM THE NUCLEAR INDUSTRY

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ABSTRACT

Activated and radioactively contaminated steel scrap from service and increasingly from decommissioning of nuclear facilities has to be managed.

As commissioning of final storages is delayed in most countries which operate nuclear power plants, recycling of radioactively contaminated scrap to products for further application in the nuclear cycle is an economic alternative.Recycling in the production of transport and storage containers for low and medium active waste has been developed in Germany since the early 80ies. These containers are made of ferritic nodular cast iron. As a central part of the design requirements for storage containers, crack initiation from drops without impact limiters onto a solid rock target at low temperatures must be excluded. In order to guarantee a sufficiently safe level for the fracture toughness, the share of the cast iron microstructure leading to brittle behaviour (pearlite and carbides) is limited to 20 % in the currently licensed containers. To produce ferritic nodular cast iron that meets this criterium the quantity and quality of elements that lead to a pearlitic microstructure (i.e. Chromium, Nickel, Manganese, Copper and Molybdenum) must be kept very low. Therefore present manufacturing rules limit the amount of scrap steel with high content of these elements, because their presence leads to a pearlitic microstructure that in turn results in lower fracture toughness.

Against this background, a research programme investigates ways to increase the amount of recycled metals in the production of cast iron containers by allowing highly pearlitic nodular cast iron while at the same time observing all design requirements. Casting exactly to specification, in order to obtain the material properties and their verification by quality control and drop testing of containers with machined artificial flaws are central parts of the R&D programme. The structural design of 2 representative container types were optimized in a way to reduce the maximum stresses under impact loading by nearly half. Presently, a series of drop tests to investigate the behaviour up to actual failure under impact is under way using dummies cast with selected inactive reference recycling materials.

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.

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 as low as -40°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 matrixlead to 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, already even at low contents in the melts. 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 on 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 type and quantity of waste metal that can presently be recycled is very limited.

GOALS AND METHODS

Based on this background the research program FORM [1] and [3] investigates ways to 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. 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. By way of optimizing the structural design of the container, the material stresses resulting from a given external loading shall be substantially reduced. 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. Drop tests of full-size and dummy containers with machined artificial flaws, after extensive precalculations, are central parts of a complementing R&D program administered by BAM (Bundesanstalt für Materialforschung und -prüfung).

MATERIAL INVESTIGATIONS

In the first phase of the research, testblocks were manufactured that were cast with a wide range of simulated (inactive) waste metal compositions. Taking specimens revealed insights on the influence of the chemical composition on the microstructure and the feasible upper limits for the relevant elements Mn, Cr, Ni, Mo and Cu. One important result was finding out that the decomposition of the high pearlite content in the microstructure in the as-cast condition by heat treatment, was not economically feasible for the desired elevated quantities of waste metal recycling. This ferritising treatment can be successfully employed only for the low amounts of waste metal used in the present manufacturing technology of containers [1] and [2].

In the 2nd phase of the research 2 dummy containers with significant dimensions and casting setup (in the following referred to as ring 1 and ring 2, the cylindrical part of the MOSAIK®II container) were cast with the 2 chemical compositions selected from the wide bandwidth of phase one. As intended and expected this lead to castings with 2 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 prototype box-shaped container type VII and the prototype MOSAIK®II container and the precalculations of their drop tests [3] and [4].

The simulated waste metal content of the ring 1 material resulted in a highly pearlitic microstructure. There were practically no carbides. Ferrite content is relatively high (in the context of this research) at 25 %. Content is defined as portion of metallic matrix (i.e. without graphite) of a metallographic specimen. The higher contents of Mn and Ni in the ring 2 material result in an almost fully pearlitic microstructure. In addition, the higher contents of Cr results in carbides. The numbers shown in Table I on shape and size of the graphite stand for the good quality of the nodular graphite, on the same level as for castings without waste metal.

Table I 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 II shows the high strengths of the 2 special cast iron materials selected for reference within the FORM program, the increases in yield strength and the decreases in elongations under dynamic loading.

Table II 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: 125x10^5	165	404	610	2,8
	Dynamic: 125	174	580	622	1,2
Ring 2, test temperature -20°C	Static: 125x10^5	168	452	602	1,5
	Dynamic: 125	168	613	621	0,3

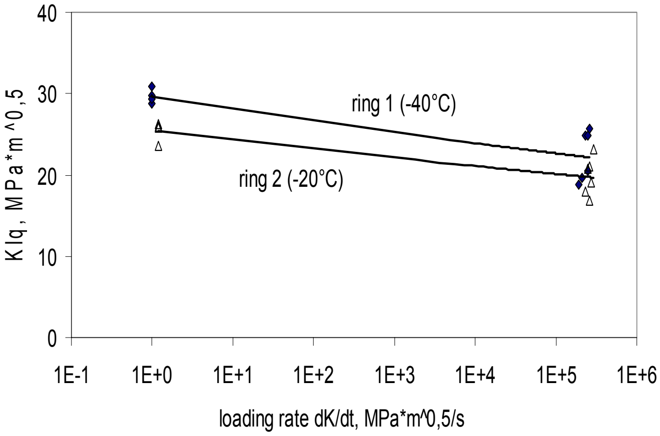

Fig. 1 Fracture Toughness (Incl. Trend Lines) Static And Dynamic.

With the high contents of pearlite the reference container materials of ring 1 and ring 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 less Cr. The influence of the loading rate also is quite limited (Fig. 1). Fracture toughness is measured on 3-point bending specimens.

Testing of the material was continued by casting and taking specimens of upright plates 800 mm wide x 1000 mm high x 160 mm thick. One half of the plate was cast against chills on one surface. With respect to cooling and solidification conditions these test plates represent full size walls for all types of containers for low and medium active waste covered by this research. The amount of waste metals (again as an inactive simulated recycling material) were varied in such a manner as to permit evaluation of the test results in a regression analysis later on. Besides metallographic aspects like pearlite and carbide contents in the microstructure, the upper limits of waste metal content are also given by manufacturing considerations like avoiding shrinkage defects and limiting hardness in order to retain economic machinability. In the future the amount of waste metal in the melt can be planned with the help of the regression analyses to meet the particular requirements.

Tensile specimens are taken from five locations on each of the test plates. As long as the melt contains none or only little chromium (≤ 0.35 weight-%) from waste metal, ductility varies widely (depending on the contents of Mn, Ni, Mo and Cu) and can be as high as 11%, despite the pearlite. With a chromium content of 0.6 % to 0.7 % the range of ductility becomes small and drops to less than 2 %, which is caused by the carbides in the otherwise almost purely pearlitic microstructure.

DESIGN OPTIMIZATION

Parallel to the material investigations the structural design of containers was successfully optimized with the help of dynamic finite element calculations. Thanks to increases in the transition radii between adjoining walls and between walls and container bottom and by use of extensions (Figs. 3 - 4) that act like built-in shock absorbers, the maximum stresses under impact loading could be reduced by almost half.

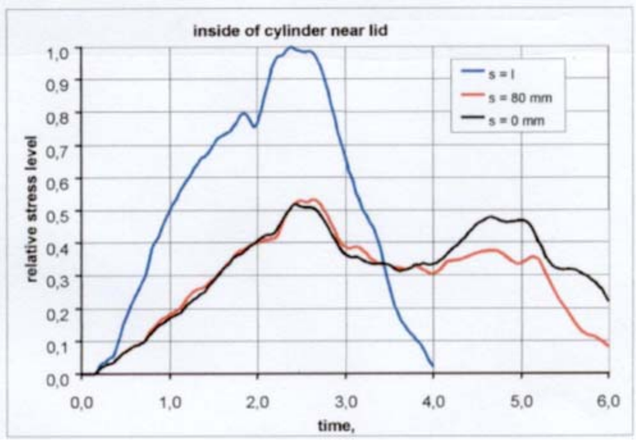

Fig. 2 Tensile Stresses in MOSAIK®II Container
From 0.8 M Side Drop Before and After Optimization

In the case of the MOSAIK®II, one of the two container types used in the optimization, the highest stresses result from a side drop. For a 0.8 m drop, which corresponds to the requirements on the waste container class II for final storage, the tangential stress that governs the design was brought down by a number of iterations from an initial level of 306 MPa (corresponding to 100 %) to a mere 159 MPa (52 %) after optimization (Fig. 2).

In the case of the box-shaped container type VII, the second reference container in this project, it is the flat bottom drop that causes the highest stresses in the structure. In a 5 m drop onto the rock target, the design requirement for waste container class I for final storage, the maximum computed deceleration, before optimization, is 1150 g. After optimization the maximum deceleration is down to 635 g.

PROTOTYPE CONTAINERS AND DROP TESTS

Both prototype containers were manufactured with chemical compositions (and thus amounts of inactive simulated waste metal) corresponding to the preceding dummy container castings, ring 1 and ring 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 moderate level of 0.3 % of chromium in the melt led to 2 % of carbides in the microstructure of the box-type container (Tables I and III). 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 is to be expected. For the MOSAIK®II container the corresponding values are nearly identical with those of ring 2, which is a very good result, again in consideration of the different temperatures RT and -20°C (Tables II and IV).

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

	Mn %	Cr %	Ni %	Mo %	Cu %	Ferrite %	Pearlite %	Carbides	Graphite	Graphite
Box-type 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 IV Tensile tests on cored samples from prototype containers under static loading at ambient temperature

	Yield strength MPa	Tensile strength MPa	Elongation %
Box-type container	354	579	4,8
MOSAIK®II container	443	591	1,8

Fracture toughness cannot be determined from the small cored samples. However, after the drop tests sufficiently large samples can be taken from both containers. The results are already available for the MOSAIK®II container (Table V, those for the box-type containers will have to wait until after a further drop test scheduled later in the program). The fracture toughness has been determined at dynamic loading rates on 3-point bending specimens from samples taken from the wall (this corresponds to ring 2) and from the bottom of the MOSAIK®II container. The regions selected are those 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. This is a very good result.

Table V Fracture toughness measured on specimens from dummy and from full size prototype container.

	Fracture toughness K_{Iq}, MPa*m^0,5 at -20°C under dynamic loading rate
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 extensions that act like built-in shock absorbers, can be seen in Figs. 3 and 4. Both containers received machined artificial flaws at all locations of calculated maximum stresses from drop test loading. Flaw size and orientation were selected such as to make precalculated applied stress intensity factors K_{appl} equal to the material properties K_{mat} measured on rings 1 and 2. The calculated safety factor was such set equal to one. The artificial flaws of the box-type container had a depth of ca. 7 mm and a length of ca. 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 depth safely sized by ultrasonic testing. Depth and width were widely overestimated in the UT.

Both prototype containers have successfully passed their drop tests at -20°C onto concrete targets simulating final storage bedrock conditions (Figs. 3 and 4). The shock-absorbing design optimization did its precalculated work. The successful drop tests of the prototypes are an important step towards proof of feasibility of containers with elevated content of waste metal. 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 artificial cracks of the MOSAIK®II container. The basis for this success lies in the high material strength, further increased by dynamic loading and low temperatures, and the dramatically reduced stress levels after design optimization. Also, the post-test investigations of the MOSAIK®II container have shown that fracture toughness was somewhat higher than anticipated (Table V). The low fracture toughness nevertheless requires an exact fracture mechanics analysis.

Evaluation of the drop test of the box-type container showed deceleration and stresses to have been lower than precalculated. The reasons for this were traced to a test target that was less stiff at its surface than intended and to an incomplete understanding of the interaction of container and target at impact. This shows the need to improve the finite element simulation of the drop test onto the bedrock target of final storage (that is not infinitely stiff like the IAEA target). In any case, the box-type container will be machined again for a second set of larger cracks and be subject to a second 5 m drop test.

Fig. 3 Prototype MOSAIK®II Ready for Drop Test at -20°C.

Fig. 4 Box-type Container After 5 m Drop Test onto Simulated Target for Final Storage.

CONTINUATION OF RESEARCH AND OUTLOOK

There are a number of goals for the third phase (FORM III) of the R&D program. For one, work is under way to establish the statistically verified relationships between the major manufacturing and material parameters like chemical composition of the melt, content of metal waste, casting setup, microstructure and mechanical properties in the container. A series of test plates with dimensions and setup relevant for container manufacturing has been cast, inspected by the standard nondestructive ultrasonic and penetrant testing and then cut into specimens for destructive testing. The statistically verified relationships just mentioned in the future will permit to select melt composition and casting setup such as to meet the required material properties as well as to economically optimize the amount of recycled waste metals. An (inactive) reference material for the manufacturing of further dummy and full containers has already been selected.

In several series of drop tests of the dummy and full containers (carried out by BAM in the scope a parallel R&D program) the behavior of the castings under ever increasing loads up to crack initiation and even failure will be investigated. Using dummy containers economically allows to do a wide range of testing. The dummies have dimensions, casting setups, material properties and drop test behavior that are relevant for the complete containers. The first series of drop tests has served to establish and verify the behavior of the dummy containers and the interaction with the target as well as to benchmark the computer calculations. For the following two series of drop tests large artificial flaws will be machined in those locations with the highest stresses due to impact. After the drop tests (with stepwise increased drop heights exceeding standard requirements) the dummies will be cut up and investigated for any crack initiation.

Based on the results of the drop tests with the dummy containers and further numerical optimization a second MOSAIK$^{®}$II container will be manufactured and drop tested from the increased height of 5 m that is required for a class II container. The existing box-type container will be machined with a second set of flaws that will be much larger set than the existing first set. Then there will be a second 5 m drop test for this container followed by post-test investigation of the flaw for crack initiation and verification of the in-situ material properties.

CONCLUSIONS

Recycling metal wastes causes the cast iron container material to have a pearlitic microstructure with low ductility. The optimization of the container design has reduced stresses resulting from accident loading by almost a factor of 2. Full size containers have passed the drop tests required for final storage. Compared to the present level of recycling the amount of metal waste in the containers can be increased by a factor of 3 to 4. The quality of non-destructive inspection is not impaired.

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