QUALIFICATION OF CAST IRON WASTE CONTAINERS MANUFACTURED BY MELTING OF RADIOACTIVE SCRAP METAL

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SUMMARY

The CEA has recycled about 1000 metric tons of contaminated metallic waste by melting in the G3 steelworks to produce cast iron containers for use in conditioning waste for disposal at the Aube surface disposal site operated by the French national radioactive waste management agency (ANDRA). Cylindrical containers with a useful volume of 1.1 m^3 were manufactured from prefabricated steel molds into which the molten metal was poured. A metallurgical characterization and test program was defined to allow qualification by ANDRA as "durable containers" suitable for conditioning radioactive waste. Tests were conducted with simulated nonradioactive waste packages to assess the gas and water tightness, fire resistance, load and drop resistance of the cast iron containers, and the thickness of each container was verified by gamma scanning. All the results met the initial program criteria and objectives. Test specimens were taken from one container for metallurgical characterization, tensile and fracture toughness testing and corrosion testing. The mechanical test results were consistent with the expected values and confirmed the nature of the material. Corrosion test results obtained under conditions similar to those of the ANDRA disposal site (concrete water) showed satisfactory corrosion resistance. Based on the overall test results, these cast iron shells can be qualified as "durable containers" suitable for conditioning of radioactive waste for surface disposal.

INTRODUCTION

In decommissioning the Marcoule G2 and G3 reactors to stage 2, the CEA chose to melt the low-level contaminated scrap metal resulting from dismantling operations. The scraps were melted in an electric arc furnace installed in the G3 reactor building together with a filtration and ventilation system to recover particle matter and ensure dynamic containment of the steelworks. Approximately 4000 metric tons of scrap metal were melted down into cast iron ingots (Tachon et al. 1997).

The CEA subsequently undertook to recycle about 1000 tons of contaminated metallic waste from other CEA sites by melting them in the G3 steelworks to produce cast iron containers suitable for conditioning radioactive waste for surface disposal. A characterization and qualification test program (Bossy et al. 1994) was defined and implemented to obtain approval from ANDRA.

FABRICATION OF CAST IRON CONTAINERS

Several tons of contaminated scrap metal were melted in the furnace, and the melt was then poured into a prefabricated steel mold. The process, which offered the advantage of producing containers with no external contamination, included the following steps :

. prefabricated steel molds were first assembled, and then covered by a steel jacket that was filled with shot to ensure uniform cooling.

. when the melt reached the setpoint temperature, the pouring ladle was positioned over the mold and the molten metal was cast in a few minutes.

. after cooling, the shot was recovered and the jacket cover and shell were stripped from the mold.

Finishing operations included eliminating the feedhead and stiffening structures used to prevent deformation of the mold during casting. The shells and covers were then submitted to a radiometric examination to check the thickness and homogeneity of the cast iron by gamma scanning using a remote-controlled radioactive source. The cover and shell mating surfaces were then machined to form a recess for the lead seal used to ensure the tightness of the container after filling with waste.

A total of 150 cylindrical containers were produced with the following dimensions :

. outside diameter :	1400 mm
. height :	1300 mm
. useful volume :	1.1 m^3
. average weight (shell + cover) :	6800 kg
. thickness :	120 mm

The resulting gray cast iron contained about 3.4% carbon. Its chemical and radiological composition was systematically analyzed. The mean specific activity of the cast iron was 12 $Bq \cdot g^{-1}$, mainly attributable to ^{60}Co.

CONTAINER QUALIFICATION PROGRAM

Before authorizing these cast iron shells for use as radioactive waste containers, they had to be qualified for durability; this involved meeting standard requirements for mechanical properties, leaktightness and resistance to corrosion, impact and fire. No specifications exist at the present time for this type of metal container. A qualification program was therefore established jointly with ANDRA, comprising metallurgical and chemical characterization of the material as well as a series of tests including primarily the following :

. aqueous leach testing of the containers,

. mechanical (tensile and toughness) testing of material specimens,

. corrosion testing of material specimens,

. waste package fire resistance, leaktightness, loading and drop tests.

A quality assurance program was defined and implemented to ensure the quality of the containers selected for conditioning and disposal of radioactive waste.

CHEMICAL AND METALLURGICAL CHARACTERIZATION

Chemical characterization

Cast iron specimens sampled from the feedhead and from the melt bath were analyzed by inductively-coupled plasma atomic emission and mass spectrometry after the specimens were completely dissolved. The analysis results showed remarkable homogeneity among the specimens from different melts. Of all the elements determined, nickel and chromium showed the greatest variability. The measured concentrations of most elements were consistent with the expected values, except for sulfur, arsenic and lead, which was given particular attention.

Metallurgical characterization

A sampling, machining and characterization protocol (Bossy et al. 1996) was defined prior to metallurgical characterization, and was also used to prepare samples for mechanical and corrosion testing. Specimens were taken from various portions of a container: bottom, midplane and top of the shell, cover, feedhead, and lead seal mating faces on the shell and cover. The specimen surfaces were prepared using penetrants and developers in compliance with French standard NF A09-120 before microscopic characterization. The following results were obtained (Barbier et al. 1995) :

- the cast iron exhibited no visible cracking,
- no bonding was observed between the cast iron and the steel mold jacket or between the cast iron and the lead seal on the cover,
- the cast iron structure of the shell was characteristic of gray (type A) cast iron with a uniform graphite flake distribution ; only a few scattered graphite clusters were observed,
- the feedhead structure was identical to that of the shell; it may be considered and saved representative control sample,
- the cast iron structure of the cover was characteristic of oriented interdendritic graphite (type E) cast iron.

Chemical and metallurgical characterization revealed no serious anomalies in the material.

MECHANICAL AND LEACH TESTING

Tensile and toughness tests

Tensile and toughness tests were carried out on cast iron specimens taken from the same container used for metallurgical characterization. An experimental protocol was developed from ASTM Standard Test Method E8. During each tensile test the conventional 0.2% yield strength, the elongation at failure and the stress at failure were determined with respect to the initial gauge. The toughness was tested by fracturing the specimen with a single stroke of a pendulum impact test machine to determine the energy at failure *KG*.

The test results indicated that the tensile strength (ranging from 175 to 215 MPa) was slightly lower than the expected range (200–250 MPa) but revealed satisfactory material homogeneity. The toughness values (8–13 J) were more scattered and lower than the expected value of 15 J, probably as a result of the high Fe_3C content.

Aqueous leach tests

Three cast iron specimens were subjected to aqueous leaching to estimate the rate of radionuclide release. The test was conducted for a duration of two months in water with a composition similar to that recommended by Standard RFS III 2.c provide reference. The estimated mean release rate for ^{60}Co activity over a one-year period was very low : approximately 10^{-5} µm/day.

CORROSION TESTING

The test objective (Barbier et al. 1996) was to assess the cast iron corrosion resistance in two types of water representative of a surface disposal environment, and to check the homogeneity of the casting. An operating protocol was defined to estimate generalized corrosion, pitting corrosion and crevice corrosion. An electrochemical test setup based on the Stern-Maclides device was used to measure the free corrosion potentials, the instantaneous corrosion rates and the pitting corrosion sensitivity.
Two types of electrolyte were used :

- a synthetic solution representative of "concrete water", characterized by a very basic pH, constitutes the reference medium most representative of surface disposal conditions;
- a leachate solution obtained by rainwater runoff over gravel, with a pH near 7, was chosen to simulate particular disposal conditions.

Corrosion resistance in concrete water

After immersion for two months in the test medium, the mean instantaneous corrosion rate reached approximately 3 $\mu m \cdot yr^{-1}$ and the mean generalized corrosion rate was 4 $\mu m \cdot yr^{-1}$. Whitish plumbierite $Ca_5Si_60_{16}(OH)_2$ and/or xonolite $Ca_6Si_60_{17}(OH)_2$ deposits were observed on the specimen surfaces, probably protecting the material against corrosion.
The tests also showed that the cast iron was not sensitive to pitting corrosion in concrete water and that the corrosion rates were in no way accelerated by crevice effects. Finally, combining the cast iron with steel diminished the instantaneous corrosion rate by a factor of ten. As a result, the cast iron exhibits satisfactory resistance to generalized and localized corrosion in concrete water, attributable to its very basic pH ($\approx$ 13).

Corrosion resistance in leaching water

After immersion for two months in the leachate solution, the generalized corrosion rate was estimated at about 1 $mm \cdot yr^{-1}$, although this relatively high indicative value cannot be extrapolated over a period of several years. Conversely, the cast iron showed little or no sensitivity to pitting or crevice corrosion. The presence of steel had no effect on the cast iron corrosion rate. The material thus exhibits poor resistance to generalized corrosion in leaching water.
The cast iron shows satisfactory resistance to generalized corrosion and to pitting corrosion in concrete water, the medium most representative of a surface disposal site where numerous concrete containers are stored in concrete structures. After 300 years (the planned lifetime of a surface radioactive waste repository), the package comprising a steel mold and cast iron shell will therefore be only slightly corroded.

WASTE PACKAGE TESTING

Four types of tests were carried out on several cast iron containers: drop test, load resistance test, fire resistance test and air/water leaktightness test. Except for the load resistance test, the containers were filled with technological waste immobilized by cement grout; the container was sealed and the cover secured in place by lead sealing.

Drop test

A container filled with conditioned waste was dropped from a height of 1.2 meter at a 45-degree angle onto the circular rim around the cover. No dispersal of material from inside the package was observed after impact, although a small zone of lenticular deformation was found on the rim and the lead seal was partially damaged.

Load resistance test

A 35-ton load was applied onto an empty container for 43 hours. No dimensional variation was observed.

Fire resistance test

The fire resistance test (Plouzennec et al. 1997) was carried out more than 28 days after the waste was conditioned and the container was sealed. Several thermocouples were positioned inside and outside the container to measure the temperature variations. The container was placed for about 30 minutes in a fire where the temperature ranged from 300 to 1500°C.
The maximum temperature on the inside wall did not exceed 70°C, and decreased rapidly after 150 minutes. The maximum temperature in the cement grout (48°C) was reached only after 15 hours. The only damage observed concerned blistering and loosening of the lead seal around the cover and distortion on the bottom of the shell.
The fire resistance of the container may be considered satisfactory, particularly in view of the relatively low temperature rise inside the package.

Water and air tightness test

A container was filled with water and pressurized. No leakage was observed around the lead seal after 24 hours.
Helium was injected into a lead-sealed container filled with conditioned waste. No leakage was detected up to 0.3 bar. The pressure drop measured at 0.4 bar was limited to 6×10^3 bar per hour. The pressure limit (0.4 bar) beyond which leakage is detectable represents a force of 6150 daN on the cover.
The air and water leakage behavior of the container may be considered satisfactory.

CONCLUSIONS

The test and characterization results obtained under the predefined qualification program are conclusive enough to qualify the cast iron shells as durable containers suitable for conditioning radioactive waste for surface disposal. The advantage of these containers lies in the high degree of biological shielding provided by the cast iron. They provide an alternative to standard concrete packages, allowing more highly irradiating radioactive waste to be conditioned for surface disposal. Cast iron containers could also be used for interim storage of irradiating waste to allow for radioactive decay with minimum occupational doses. The fabrication of cast iron containers from low-level contaminated scrap metal provides a promising solution for recycling metallic waste, provided the residual radioactivity is limited and the production costs are suitably optimized.

REFERENCES

F. BARBIER *et al.*, *Metallurgical characterization and corrosion testing program*,
Note CEA/DTA/CEREM/DECM/SCECF/SECA, February 1995.
F. BARBIER *et al.*, *Qualification of cast iron containers : metallurgical characterization*,
Note CEA/DTA/CEREM/DECM/SCECF/LPCC 15471, July 1996.
R. BOSSY, *Cast iron waste containers : qualification program*,
Note CEA/DCC/DESD/BECC 94/147, July 1994.
R. BOSSY *et al.*, *Cast iron waste containers : synthetic report on qualification testing*,
Note CEA/BECC/XDO2/R501, November 1996.
PLOUZENNEC *et al.*, *Qualification of cast iron containers : corrosion resistance*,
Note CEA/DTA/CEREM/DECM/SCECF 424, June 1997.
M. TACHON *et al.*, *Report on decommissioning to stage 2 G2/G3reactors*,
Note CEA/DCC/DRDD/UDIN, 722/8/I/P/427, June 1997.