STMI'S INDUSTRIAL EXPERIENCE IN LOW ACTIVITY STEEL WASTE PROCESSING BY MELTING

J. P. Lacroix
STMI
91190 Gif Sur Yvette, France
M. Tailliez
EDF-SPT-UTO
95008 Paris, France

ABSTRACT

STMI has been requested by EDF to produce a pilot cast, made from contaminated carbon steel. The contaminated metal came from heat exchangers from Chinon A2. The purpose of the cast was to find the behavior and final distribution of the contamination radionuclides after melting. It also was aimed to evaluate the potential contamination of the surrounding environment due to the melting and casting processes.

The pilot cast was produced in April 1984 in an existing industrial facility using full scale industrial facilities and methods. A melting arc furnace of four ton capacity was loaded with 3.6 tons of cut iron pieces (such as pipes and vessels) showing a 20 mCi/ton total activity. Atmospheric contamination monitoring was conducted during all steps of the activity. Atmospheric contamination monitoring was conducted during all steps of the operation: furnace feeding, melting, oxydation, reduction and casting. Monitoring results were summarized and reported to the SCPRI (Central Agency for the Protection from Ionizing Radiation of the French Public Health Ministry).

The operation included a gas and smoke filtration step. In general, all produced wastes were recovered and analyzed: filtration dust, furnace fire-clay, slag and sand from the mold and casting ladle(?). The distribution of radionuclides was determined experimentally both in the cast and in the waste materials.

The final products were: a cylindrical cask (D = 600 mm; H = 800 mm; 40 mm thickness); two shielding plates (D = 500 mm; H = 12500 mm; 100 mm thickness); and four cylindrical ingots (D = 380 mm; H = 500 mm). All of 7.85 G/CM3 density.

In summary, the end products of a low activity waste metal melting industrial plant can be handled in two ways: (1) reuse in the nuclear power field, with proper case to minimize the activity present in the products; (2) storage of the low volume waste. It should be considered as the best and most cost efficient process for low activity metallic waste.

It is STMI/EDF's intention to cast an additional four tons of stainless steel by the end of 1985.

PRESENTATION

In pursuit of a policy to reduce the costs of managing radioactive waste from its nuclear power plants, the French Electricity Board (EDF) has launched a design study and test program to define the best technical and economic conditions applicable to the management of metallic waste.

Among the solutions to be visualized, the fusion of drums can be justified by:
- a reduction in the volume of waste to be stored;
- the dilution of very contaminated, or high-level waste with other metallic waste of low radioactivity;
- the manufacture of finished products suitable for recyling in the nuclear industry.

It appeared indispensable to make a full-scale examination of the conditions required to overcome the personal and environmental risks of radioactivity throughout a fusion operation.

The EDF therefore commissioned the Societe des Techniques en Milieu Ionisant (STMI) to perform an experimental fusion operation on contaminated carbon steel in an existing industrial foundry, for modifications to the process employed, but taking all the precautions necessary to overcome the previously assessed risks.

Prior to this operation, a file which included, in particular, a detailed description of the arrangements for measuring contamination and for radiological testing, was submitted for comments and approval to the Ministry of Health's Central Bureau for Protection against Ionizing Radiation.

DESCRIPTION OF OPERATIONS

The experimental operation consisted of:
- the selection, preparation and radiological evaluation of drums from the storage yard of a nuclear power plant;
- transport of the drums to the foundry;
- the melting, casting and production of ingots and molded parts.

Selection, Preparation and Radiological Evaluation of Drums

The materials were taken from a rejected heat exchanger from the EDF 2 Reactor (GCR) from CHINON A power plant and; more precisely, *from its lower part* (low-pressure vaporizer) which appeared to have surface activity allowing the specific activity to be restricted to 3.7 to 7.4 10^8 $Bq.T^{-1}$ (10 to 20 $mCi.T^{-1}$).

This evaluation has been made, on one hand, using measurements of surface radioactivity or estimation of the developed surface and, on the other hand, using calculations based on dose rate measurements and geometric dimensions.

The weight planned for the operation was restricted by the size of the furnace, i.e., 3 to 4 metric tons.

Starting with the dimensions of the furnace and the melting technique used, it was necessary to cut the material into sections not exceeding 1 meter long. This has been achieved with a flame cutter, in a ventilated area equipped with filters, by operators wearing protective clothing to avoid contamination. This precaution was made necessary by the particularly unstable nature of the contamination. The preparatory work has been performed with 211 man-hours.

Transport of Drums

Once cut, the metallic pieces have been placed in the internal basket of a steel vessel of the "5 cu m injectable vessel" type approved by ANDRA (Agence Nationale pour la Gestion des Dechets Radioactifs--National Agency for the management of radioactive waste). This vessel is used here as a carrying container.

Melting and Casting

The melting, casting and molding of the ingots and other pieces took place on the facilities of Fonderies et Acieries de Feurs Company (in the Loire department).

This industrial establishment has two plants located in the South of Feurs. The work was done in the Northern Hall of the South plant, where furnace No. 2 is located. This zone offered the advantage of being practically isolated, due to its operational self-sufficiency and its handling and casting areas.

The work area has a system of ventilation on and around the furnace to collect fumes and gases and transfer them to a filtration plant located at the foot of a 16-meter stock, which discharges about 50,000 cu m per hour when in operation.

Furnace No. 2 is a graphite electrode arc furnace with a capacity of 4 to 5 metric tons. It has a refractory lining (natural ramming mass from UZES--QUARTZ--Silica sand) and can be tilted for casting. Practically all the gases and fumes are collected by a mobile hood above the furnace. Any emissions not collected by the hood are drawn into a suction device located below the roof of the buildings.

Prior to starting the work, insulating equipment was installed to prevent any transfer of radioactive elements in the event of material being dropped during handling.

On the eve of the operation, an inactive casting was carried out in order to determine the background noise and to check the monitoring devices. On this occasion, the ventilation ducts were checked and no dust deposits were found.

The melting operation concerned a weight of about 3.8 metric tons loaded into the furnace using a traveling crane equipped with a lifting magnet and a weighing system. After melting the metallic waste, oxygen was injected, followed by deoxidation by the addition of manganese salt. The subsequent casting lasted about twenty minutes in previously prepared molds.

These molds were of a standard type, with a metal flask and a filling of sand solidified by the reaction of sodium silicate to an ester, resulting in the release of silica. The finished products consisted of: four ingots;
two armored plates;
one armored drum shell.

During these operations, the following activities took place:

- checkings of the transfer of activity in the atmosphere and on the surfaces, as well as dose rate measurements;
- periodic sampling of metal and slag;
- containuous sampling of dust in the gas and fume discharge circuit.

Overhaul of Installations

On completion of the melting operation, certain findings were noted prior to overhauling the equipment:

- Furnace: A check of the furance found metallic inclusions in the refractory lining and these were removed by stripping one layer. Waste matter from these operations was packed into drums. This work has been performed under controlled atmospheric contamination.
- Pouring ladles: The metallic deposit at the bottom of the ladles was removed.
- Slag: The slag, poured using a special ladle and comprising metallic inclusions, was packed in drums.
- Sand: The molding sand that had been in contact with the metal was found to have an equivalent activity about 3.7 Bq cm^{-2} (10^{-4} u Ci.cm^{-2}).
- Dust: The dust gathered by the dust collector was bagged, placed in a drum and removed as waste.
- Miscellaneous: No significant contamination of the hood was found.

The metal foil protecting the lifting magnet was removed and added to the waste material.

RESULTS

Weight Ratios

The total weight of materials used was determined by means of partial weighing and was proportioned as follows:

Molded parts	3660 Kgs	94%
Slag	200 Kgs	more than 5%
Dust collected	30 Kgs	less than 1%

These are the normal proportions found in standard melting operations.

Distribution of Radioactive Elements

Measuring Techniques:

The level of radioactivity was determined using standard means of measuring radiation:

- measurements of $\beta\gamma$ on both samples and work surfaces;
- spectrometry γ on samples;
- measurement of dose rate on samples and in situ.

The latter were used, in particular, to cross-check the direct measurements of radioactivity.

Radioactivity of finished products:

Of the total activity involved, 99.5% was found in the cast metal, i.e., 2.81 10^{9} Bq (76 m Ci).

This radioactivity was distributed as follows:

^{60}Co	99.7%
^{54}Mn	0.2%
^{65}Zn	0.1%

It was found that the level of radioactivity was fairly even throughout, although there was an increasing rate of ^{60}Co activity between the surface and the core, up to a factor of 2. Later experiments using inactive cobalt failed to confirm this factor, which would therefore appear to be a result of statistical fluctuations.

Radioactivity of melting waste:

The total waste activity was determined as follows:

- filtration dust	5.5 MBq (148 uCi)
- slag	8 MBq (217 uCi)
- sand	1.4 MBq (38 uCi)

(Where the sand is concerned, this activity is due to metallic particles which are recovered during purification of the sand.)

Note should be taken of the fairly high amount of cesium in the dust. This was expected, given the melting technique, the surface-type contamination and the low density of the cesium.

Atmospheric radioactivity:

Atmospheric discharges were checked in the immediate vicinity of the furnace during these operations and in the discharge stack, by sampling.

Checks in the vicinity of the furnace produced the following results per operation.

Type of Operations	Activity Concentration Bq m^{-3} (Ci m^{-3})
Loading of furnace	4.8 (1.3 10^{-10})
Melting	4.10^{-2} (1.1 10^{-12})
Oxidation-Deoxidation	0.6 (1.7 10^{-11})
Casting	0.5 (1.3 10^{-11})

These activities did not exceed the acceptable public level of 0.4 maximum permissible concentration (NPC).

Sampling was made in the immediate vicinity of the materials during handling and at the fume generating point during periods of emission.

The discharged activity, determined by sampling of filters was as follows, during the various work phases:

Type of Operations	Activity Concentration (Bq.m^{-3}(Cim^{-3})	Total Discharged Activity Bq (Ci)
Loading of Furnace	0.2 (5.2 10^{-12})	7400 (2.10^{-2})
Melting	0.2 (5.2 10^{-12})	11100 (3.10^{-7})
Oxidation-Deoxidation	0.06 (1.5 10^{-12})	1480 (4.10^{-8})

The maximum instantaneous discharged activity was:
2.6 Bq.S^{-1} (7 10^{-11} Ci.S^{-1})

Taking into account an atmospheric transfer coefficient of about 3.10^{-6} s.m^{-3}, maximum ground activity in the area under windy conditions was 6.8 10^{-4} Bq m^{-3} (2.10^{-14} Ci.m^{-3}) for a total discharge in the order of 18500 Bq (0.5 uCi). The isotopes discharged were identical to those found in the dust. Nevertheless, measurements made outside the plant failed to reveal any radionuclides other than those present in the natural state.

Dose Rate Measurements of Finished Products:

The dose rate measurements were carried out at a distance of 10 cm from the surface of parts:

Ingots (4 measurements)	0.2, 0.22, 0.22, 0.26 mGy.h^{-1}
Armored Plate	: 0.46 and 0.48 m Gy.h^{-1}
Armored shell	: 0.26 m Gy.h^{-1}

ANALYSIS AND CONCLUSION

From the industrial point of view, this operation demonstrated that metals with very low radioactivity can be melted using standard equipment only slightly modified for this purpose. The quality of the steel produced is such that it is possible to envisage a wide range of finished products with several grades of steel to suit requirements (mild steel; weldable mildsteel; hardened steel; alloyed steel). It is also possible to envisage the manufacture of steel with a high carbon content (semi-steel) where higher resistance to corrosion is required.

From the radiological point of view, the experimental casting has demonstrated that the initial unstable surface radioactivity is transformed into radioactivity that spreads through the steel and settles at a level of 99.5%. Most of the remainder is found in the generated waste and can be recovered in a fairly stable form.

The atmospheric activity around the furnace did not exceed a public level of 0.4 maximum permissible concentration (MPC) during about one-sixth of the operating time, with a level of 3.7 10^{-3} to 5.7 10^{-2} MPC for the whole population the rest of the time, in very limited areas in the vicinity of the sources.

The discharge through industrial filtration systems not designed for very high level purification was in the order of only 18500 Bq (0.5 uCi), i.e., 0.3% of the total activity found in the dust.

The activity of the waste generated by the melting operations was essentially in the form of steel particles encased in the slag, the molding sand and the dust in the filtering sleeves.

Based on the results of this experiment, extrapolation to the scale of an industrial operation was envisaged for 4,000 metric tons of metallic waste from CHINON A (EDF reactor No. 3) and with a specific activity a thousand times lower than that involved in the experimental operation.

It can be considered that the dose rate measured at a distance of 10 cms from the finished products would not exceed 0.4 u Gy.h^{-1}, i.e., equivalent to the dose rate generated by natural radioactivity. In the same way, the total specific activity of the finished products would be in the order of 7.4 10^{5} Bq.T^{-1} (2.10^{-5} Ci.T^{-1}) where-as the overall specific activity of the generated waste would be broken down as follows:

Slag : 3.7 10^4 $Bq.T^{-1}$ (1.10^{-6} $Ci.T^{-1}$)
Sand : Not applicable (steel particles recovered during purification of the sand prior to re-use)
Dust : 1.85 10^6 $Bq.T^{-1}$ (5.10^{-5} $Ci.T^{-1}$)

For an operation involving about 4,000 metric tons, the melting waste would comprise about 200 metric tons of slag and 300 metric tons of dust, with respective overall activity levels of 7.4 10^5 Bq (2.10^{-4} Ci) and 5.6 10^7Bq (1.5 10^{-3} Ci).

Finally, there would be no significant increase in atmospheric radioactivity outside the plant.

Consequently, the dose rates involved are insignificant enough to allow the achievement of a large-scale operation in complete safety. In France, the only obstacle would come from current regulations for, in the absence of a threshold allowing definition of a publicly-acceptable level of specific contamination of a metal, a technical safety file has to be established for each operation and submitted to the ministerial authorities for approval. However, attempts are currently being made to define this threshold and this would allow the melting of contaminated metallic wastes on a truly industrial scale.