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PERFORMANCE OF A POLYMER SEALANT COATING IN AN ARCTIC MARINE ENVIRONMENT

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ABSTRACT

The feasibility of using a polymer-based coating, Polibrid 705, to seal concrete and steel surfaces from permanent radioactive contamination in an Arctic marine environment has been successfully demonstrated using a combination of field and laboratory testing. A mobile, self-sufficient spraying device was developed to specifications provided by the Russian Northern Navy and deployed at the RTP Atomflot site, Murmansk, Russia. Demonstration coatings were applied to concrete surfaces exposed to conditions ranging from indoor pedestrian usage to heavy vehicle passage and container handling in a loading dock. A large steel container was also coated with the polymer, filled with solid radwaste, sealed, and left out of doors, exposed to the full annual Arctic weather cycle. The 12 months of field testing gave rise to little degradation of the sealant coating, except for a few chips and gouge marks on the loading bay surface that were readily repaired. Contamination resulting from radwaste handling was easily removed and the surface was not degraded by contact with the decontamination agents. The field tests were accompanied by a series of laboratory qualification tests carried out at a research laboratory in St. Petersburg. The laboratory tests examined a variety of properties, including bond strength between the coating and the substrate, thermal cycling resistance, wear resistance, flammability, and ease of decontamination. The Polibrid 705 coating met all the Russian Navy qualification requirements with the exception of flammability. In this last instance, it was decided to restrict application of the coating to land-based facilities.

INTRODUCTION

The Arctic Military Environmental Cooperation (AMEC) forum was launched in 1996 with the aim of supporting a dialogue and joint activities in the Arctic region among U.S., Russian, and Norwegian military and environmental officials. Of the seven project areas approved under AMEC, one, Project 1.4, is designed to address the development and implementation of technologies relevant to the storage of solid radioactive wastes generated by the Russian nuclear submarine dismantlement program. The first program under this Project involved developing and qualifying a technology designed to seal concrete and metal structures from permanent radioactive contamination in an Arctic marine environment [1]. This paper describes the development of a technology for depositing coatings of a polymer-based sealant, and the performance of the coating under both actual field conditions and in laboratory qualification tests.

THE TECHNOLOGY AND THE SPECIFICATIONS

The technology selected for this task derived from the interest expressed the Russian Federation's Northern Navy in preventing permanent contamination of structural materials (such as concrete and steel) during the handling, processing and storage of radioactive wastes. Such contamination poses several problems, such as increases in worker exposures, and processing and disposal cost at the end of the life of these materials. The most promising solution appeared to lie in the use of a polymer-based sealant, so the decision was made to focus on demonstrating the viability of such a coating when deposited on concrete and metals surfaces exposed to the environmental and working conditions found at the RF Northern Navy bases. These are located mostly in the Murmansk region in northwest Russia, inside the Arctic Circle. The conditions in this region dictated that the coating should be applied during the relatively mild weather of the summer in that region (high temperatures usually in the range 10-15°C).

The first steps involved the preparation of a set of technical specifications and identification of a suitable supplier for the equipment and sealant material. The specifications assembled by the Russian Navy personnel are summarized in Table I.

Table I. Equipment and Sealant Specifications

Equipment	Sealant
• Must be a stand-alone unit • Must be mobile • Must have own power supplies	• Must be resistant to a marine environment • Must be resistant to Arctic temperatures • Must be resistant to acids, bases, organic solvents • Must be resistant to permanent contamination • Must be easily decontaminated • Must have flammability resistance

The requirement that the equipment be both mobile and self-sufficient was critical because it was likely that it would be used in places where utilities such as electricity and compressed air were not readily available. The sealant requirements were commensurate with those that might be expected for use in any solid radioactive waste handling facility located within the Arctic Circle. With regard to the sealant, enough material was to be supplied to cover an area of at least 1500 m^2 to a thickness deemed sufficient to meet the needs of the application (2 to 3 mm, depending on the substrate). The selected vendor was also required to provide equipment manuals in Russian and to train Russian personnel in the use of the equipment.

TECHNOLOGY DESCRIPTION AND DEMONSTRATION PLAN

Promatec Technologies, Cypress, TX, was contracted to supply the specified equipment and sealant materials. The Promatec technology uses an airless spray system to deposit coatings of a polymer named Polibrid 705 [2]. The process involves preheating two components, a resin and a catalyst (called Compounds A and B), using a hydraulic system to pump them to a mixer manifold, then spraying the resultant mixture onto the object surface. Polibrid 705 coatings have the advantage that they cure (harden) very rapidly and can be put into use within a very short time of being applied. This quick-setting ability also means that, at the conclusion of each spraying operation, the mixer manifold and the spray gun must be flushed with a solvent (methyl ethyl ketone, MEK). If this is not done within a few minutes of ceasing to spray, the residual polymer will solidify in place and block this part of the delivery system. The conventional Promatec spray unit normally relies on external sources for utilities. However, in order to meet the Northern Navy specifications, two gasoline engines were added, one for an electrical generator and the other for the hydraulic pump. Figure 1 shows the final unit being set up at the Russian site prior to spraying operations.

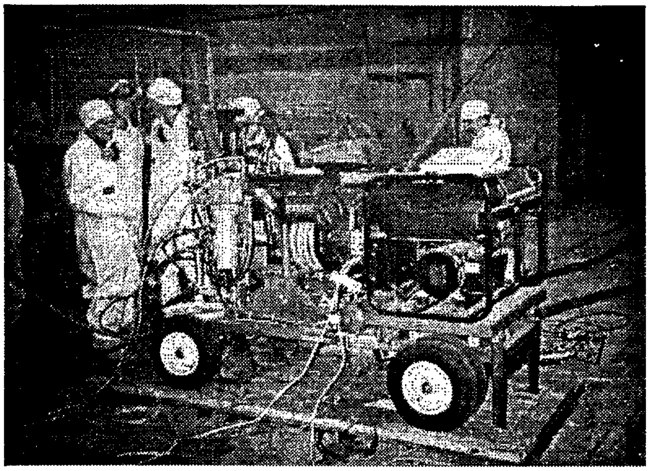

Figure 1 Polymer spray equipment at the RTP Atomflot site in Murmansk

The Promatec technology has been proven to be effective in sealing concrete and metal structures in several non-nuclear applications. However, the polymer had not been subjected to environmental conditions as severe as those experienced at the Russian Northern Navy bases. Consequently, the Russian Navy personnel met with the technical staff at the Interbranch Coordination Scientific-Technical Center of Nuclide Production (ICC Nuclide) to develop a plan to demonstrate that to qualify the coating for use in the Arctic marine environment. This plan comprised both testing under actual field conditions at the RTP Atomflot site in Murmansk and an extensive laboratory qualification test program, to be conducted at ICC Nuclide's research facility in St. Petersburg. The field testing involved coatings deposited on concrete surfaces exposed to both light and heavy duty use, and on the outside surfaces of a steel radioactive waste container. The laboratory test program focussed on various physical and chemical properties of the Polibrid 705 coating after deposition on both concrete and steel substrates. The properties of interest, specified by the Russian Navy personnel, were: coating adhesion strength, moisture and decontamination resistance, ease of decontaminability, wear resistance, resistance to low temperatures and to thermal cycling, isolation properties, and flammability resistance.

COATING APPLICATION

The surface designated for the initial demonstration of the spraying equipment was an approximate 50 m^2 concrete area located just inside the main entrance to a high-bay area used for handling and storing solid low-level radioactive waste. Later, the coating was applied to an internal corridor and staircase, some room floors, some concrete and metal coupons (to be used in laboratory testing in St. Petersburg), and a 1 m^3 steel container and its steel plate lid. This container was to be filled with solid radioactive waste, the lid put in place, and the unit positioned on the roof of the building, exposed to the elements for one year.

Actual coating of the surfaces was completed in August 1998, while the ambient outside temperatures were generally just above 10°C, and was performed by Russian personnel after training by the Promatec engineers (Figure 2). Figures 3 and 4 show, respectively, the loading dock floor after spraying and the coated solid radwaste container.

Figure 2. Russian technical expert spraying the concrete floor of the loading dock in the high-bay area.

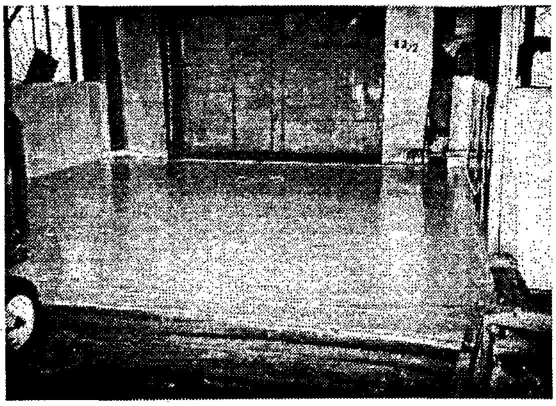

Figure 3. Concrete loading dock floor in the high bay after spray coating with Polibrid 705.

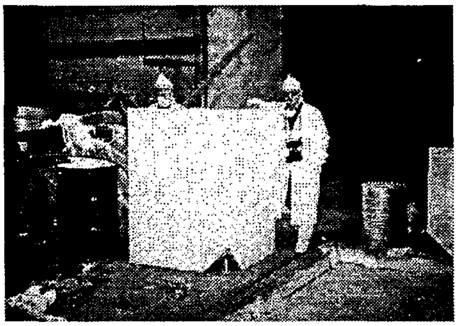

Figure 4. Solid radwaste container after spray coating with Polibrid 705.

FIELD TEST OBSERVATIONS

The loading dock floor is exposed to vehicular use on a daily basis and represents possibly the most severe environment to which the coating is likely to be subjected in normal circumstances.

Over the 12 months following the spray coating, the temperature in the area varied from +15°C to -15°C and the coating was subject to mechanical wear (from constant vehicle usage and handling operations), radioactive contamination, and treatment with decontamination solvents. It was also exposed to gamma radiation at an estimated 3 μGy/h and β contamination to a maximum level of 167 kBq/m^2. The rubber-tired vehicles caused negligible wear on the coating but damage was sustained from the use of sleds with metal runners and from the impact of metal objects during crane lifting operations. Some contamination occurred but this was removed in a single decontamination cycle. The decontamination process itself had no effect on the properties of the coating.

The conditions experienced by the coatings on the internal corridors and stairway, and on the room floors were less severe, with temperatures always above 0°C. Some damage was caused by the constant passage of personnel using industrial-type footwear.

The coating on the container was exposed to temperatures as low as -45°C, daily temperature cycling, atmospheric precipitation (rain and snow), solar radiation, and gamma radiation (from the contained waste, estimated at about 1.2 mGy/h). However, it was not subjected to the kind of mechanical abuse suffered by the other coating surfaces and its durability was judged to be high.

LABORATORY TESTING

Laboratory testing, which was required to qualify the sealant for use in Russian Northern Navy installations, was carried out at ICC Nuclide's research center in St. Petersburg. The program, summarized in Table II, involved coated samples of concrete (100 mm x 100 mm x 20 mm) and steel (100 mm x 100 mm x 2 mm), some of which had been spray-coated at RTP Atomflot and the remainder brush-coated at ICC Nuclide. With the exception of the determination of bond strengths and isolation characteristics, the tests were conducted in accordance with applicable Russian Navy specifications, Bond strength tests followed a procedure proposed by BNL while ICC Nuclide developed a specific procedure to determine the isolation properties of Polibrid 705.

Table II. Property and Test Specifications

Property	Test Specification
Bond strength	BNL procedure
Decontaminability	GOST 27708-88 RD 95.10366-89
Wear resistance	GOST 20811-75
Thermal cycling	GOST 6128-81
Isolation	ICC Nuclide test
Flammability	GOST 21227-75 ICC Nuclide test

Bond Strength Results

The experimental data from the bond strength tests are summarized in Tables III (as-deposited strengths) and IV (strengths after immersion in various media).

Table III. Polibrid 705 Bond Strengths

Substrate	Strength (MPa)
Steel (dry surface)	3.2-3.6
Steel (corrosion on surface)	2.5
Steel (acid-etched surface)	2.4
Steel (old paint)	3.1
Concrete (dry)	1.1-1.5
Concrete (wet)	<0.05
Concrete (old paint)	1.2

Table IV. Bond Strengths after Immersion

Substrate/Medium	Strength (MPa)
Steel/sea water	3.3
Steel/SF-3K	3.1
Steel/ZPS-1M	3.2
Steel/Redox solution	2.1
Concrete/sea water	0.5
Concrete/SF-3K	1.4
Concrete/ZPS-1M	1.6
Concrete/Redox solution	1.1

At first glance, it appears that the Polibrid 705 coatings bonded better to steel than to concrete. However, this is a deceptive observation because, in the case of the concrete, failure always occurred in the concrete itself rather than at the concrete/coating interface. This can be attributed to the low quality of the concrete. Thus the true concrete/coating adhesion strength is unknown although it would be higher than that recorded in the current tests. In a similar context, the bond strength is slightly less when the coating is deposited on painted steel. Again, this is mostly a reflection of the strength of the bond between the steel and the paint. The results also confirmed the importance of surface condition prior to coating application, dry clean surfaces being most desirable for good adhesion.

Immersion in sea water and three types of decontamination solution (SF-3K, ZPS-1M, Redox) usually caused a reduction in the bond strength of steel samples but a similar effect was not so obvious with the concrete samples, again presumably because of the problems associated with using a low quality concrete. Generally, the immersion resistance was considered to meet the Russian Navy requirements, provided that the coating was applied to a clean dry surface.

Decontamination Capability

Decontamination tests involved five radionuclides and three decontamination methods, ZPS-1M, SF-3K, and a redox solution (ZPS-1M is a polyvinyl spirit that, after application, evaporates to leave in place a strippable coating; SF-3K is an oxalic acid-based decontamination solution). The Russian Navy requires (in OTT 6.1.40-90 and OTT 6.1.38-90) that materials used at sites with a contamination potential must be easily decontaminated. By this, it is meant that the decontamination coefficient (the ratio of the initial to the final level of contamination) should be approximately 10 or higher after the first decontamination cycle. An additional requirement is that the materials must be able to withstand up to 3 cycles of decontamination without degradation.

The results of the tests conducted by ICC Nuclide are summarized in Table V. The decontamination coefficient data indicate that these criteria were met by both materials for all radionuclides using the ZPS-1M method and for all except Cs-137 using the SF-3K method. However, when the redox solution was used, decontamination coefficients were low (2 to 4.5) in all cases and the surface became visibly porous.

Table V. Coefficients of Decontamination (first cycle) of Polibrid 705

Radionuclide	Bonded to steel		Bonded to concrete	
	SF-3K	ZPS-1M	SF-3K	ZPS-3M
Cs-137	5.2	21	3.5	10
Co-60	33	30	9.3	9.3
Eu-152	163	---	162	---
Sr-90	165	---	80	---
Pu-239	84	---	400	---

Considering that the predominant contaminating radionuclide is usually Co-60, the test results indicated that the decontamination capability of Polibrid 705 is satisfactory when either SF-3K or ZPS-1M are used. However, if Cs-137 is the predominant contaminant, then only the ZPS-1M solution should be used.

Wear Resistance

The wear resistance tests involved making 2000 abrasive strokes using a special "pin-on-plate" type device, followed by visual examination of the coating surface. The Polibrid 705 coatings showed no damage and the average mass loss was only 0.0021 g. Thus the coatings were judged to meet the requirements of the Russian Navy.

Thermal Cycling (Cold Weather) Resistance

Three tests were performed at ICC Nuclide's laboratory in St. Petersburg while a fourth test was done at the National Institute of Standards and Technology (NIST) in the U.S.A.

Two of the ICC Nuclide tests involved a similar thermal cycle sequence: 6 hours at -40°C followed by 8 h at 20°C, repeated 5 times. The first of these tests was performed on the initial ingredients (Compounds A and B). After the sequence the two compounds were mixed to determine if any coagulation had taken place. In the second test, the thermal cycles were applied to coatings of Polibrid 705 on concrete and on steel, after which the coatings were examined visually then coating adhesion tests were performed. The results of the two kinds of tests indicated that neither the initial ingredients nor the coating had degraded. The third test at St. Petersburg involved the application of 25 cycles between -20°C and +20°C to coatings on both concrete and steel, after which coating adhesion strengths were measured. In this case, bond strengths decreased significantly from those of the as-deposited coatings. However, it was concluded that the coatings still met the Russian Navy requirements.

The NIST tests [3] used coatings on both concrete and steel, and a free-standing film, and involved a thermal cycle of one h at -21°C followed by at least one h at +25°C. Specimen surfaces were examined with a special optical-digital microscope daily until the prescribed number of cycles had been accumulated (50, 100, 150, 200). No visible (<0.1 μm) cracks were observed, leading to the conclusion that the coating could be successfully applied in a climate with frequent sharp temperature drops.

The results from the thermal cycling tests on the Polibrid 705 coatings (as opposed to the test on initial ingredients) are summarized in Table VI.

Table VI. Thermal Cycling Tests on Polibrid 705 Coatings

Location	Temps. (°C)	Cycles	Test	Results
ICC Nuclide	-20 to +20	5	Visual	No change
			Adhesion	No change
		25	Adhesion	Reduced bond strength
NIST	-21 to +25	200	Visual	No cracking

Isolation Characteristics

The isolation tests were performed to determine the feasibility of using Polibrid 705 to isolate fixed radioactive contamination on containers and other surfaces. To this end, samples of steel and concrete were surface contaminated with appropriate radionuclides (200-300 Bq for each radionuclide) then coated with Polibrid 705 (thickness 1-1.5 mm). Alcohol smears were subsequently taken at specified intervals after application of the coating, and the smear activity measured using a spectrometer and a radiometer. Estimates were thus possible of the proportion of radionuclides that had diffused through the coating to the surface. The results from the first three months of testing indicated that the highest proportion recorded was only 10^{-3} (for Cs-137 and Co-60); that is, after three months only 0.1% of the radioactivity had migrated to the surface of the Polibrid-coated area. Based on this, the Russian Navy concluded that Polibrid 705 had good isolating characteristics.

Flammability

The two tests used by ICC Nuclide to assess flammability are unlike any of the standard methods used in the U.S. (e.g., NFPA 255 and ASTM D 3806). The GOST 21227-75 required coating a piece of aluminum foil, holding it over a naked flame for 10 s, and observing how much is burnt. In the case of Polibrid 705, the coating burned completely and it thus failed to meet the Navy requirement for the charred area not to exceed 60 mm. The second (and non-standard) test performed at ICC Nuclide used a gasoline-induced fire around a 100 mm cubic container coated with 2mm of Polibrid 705. In this case, the coating was burnt almost completely through on two sides. The results of these two tests raised some concern among the Russian Navy personnel about the flammability resistance and threatened to be a "show stopper." The Russian tests were examined in depth and compared with the results of a flame spread test (ASTM D 3806) performed in the U.S. on an aluminum sheet (10 cm x 60 cm) with a 1 mm thick Polibrid coating. These latter results had indicated that Polibrid 705 self-extinguished after the flame source had been removed. After much discussion, it was decided to limit the application of Polibrid coatings to shore-based facilities.

CONCLUSIONS

A mobile, self-sufficient spraying device was developed to specifications provided by the Russian Ministry of Defence Northern Navy and deployed at the RTP Atomflot site, Murmansk, Russia. The equipment has been used to apply demonstration coatings of a polymer, Polibrid 705, to concrete surfaces that are exposed to conditions ranging from indoor pedestrian usage to heavy vehicle passage and container handling in a loading bay. A large steel container was also coated with the polymer, filled with solid radwaste, sealed, and left out of doors for twelve months, exposed to the full Arctic weather cycle. The field tests were accompanied by a series of laboratory qualification tests carried out at the research laboratory of ICC Nuclide in St. Petersburg.

During the 12-month field tests, the sealant coating showed little sign of degradation except for a few chips and gouge marks on the loading bay surface that were readily repaired. Contamination resulting from radwaste handling was easily removed and the surface was not degraded by contact with the decontamination agents.

In the laboratory testing Polibrid 705 met all the Russian qualification requirements with the exception of flammability. In this last instance, it was decided to restrict application of the coating to land-based facilities.

ACKNOWLEDGEMENTS

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