Kazakhstan: Treatment and Safe Disposal of Liquid Radioactive Waste from the BN-350 Reactor Unit at the LRW Processing Facility- 14016

*Dennis Kelley,**Artem Guelis, ***Anatoliy Ivanov, ***Igor Yakovlev, ****Anatoliy Galkin, ****Alexander Klepikov

*Pacific Nuclear Solutions, Indiana USA, **Argone National Laboratory, Illinois, USA ***Atomic Energy Combine, Aktau, Kazakhstan ****Nuclear Technology Safety Center, Almaty, Kazakhstan

ABSTRACT

The Mangystau Atomic Energy Combine (MAEC-Kazatomprom) located in Aktau (Shevchenko), Kazakhstan, began operations in 1973 and closed operations in 1999. The designed thermal power generating up to 1,000 MWt from the BN-350 sodium-cooled fast neutron reactor served three purposes: power generation for the city, power for the desalination plant and production of weapons grade plutonium. The reactor was decommissioned and spent fuel was sent to the Baikal – 1 site in northeast Kazakhstan with completion in 2010. Remaining on-site in Aktau at the LRW Storage facility are ten above-ground liquid storage tanks (currently five tanks are in operation) that are filled with a variety of mixed liquid waste. The waste consists of aqueous, sludge types and oil with an estimated 3,217 cubic meters of liquid. The total activity of Cs-137 nuclide is 9,618 Ci . It is the intent of MAEC to find suitable, low cost technologies to treat and dispose of the radwaste, to enable the completion of BN-350 decommissioning. At present, MAEC's plan is to store the radwaste on-site before the construction of the LRW processing facility (LRWPF), which will treat the main liquid waste streams.

In 2011 the U.S. Department of Energy's National Nuclear Security Administration (NNSA) and the Kazakhstani Nuclear Technology Safety Center (NTSC) began discussions about various potential projects under NNSA's Global Initiatives for Proliferation Prevention (GIPP) program. NNSA offered a joint project involving NTSC and MAEC to introduce and validate the use of high technology polymers for the purpose of solidifying a portion of MAEC's liquid waste which cannot be processed through the designed LRWPF. In addition NNSA's GIPP proposal took into consideration a cost effective process that would comply with final storage requirements in Kazakhstan.

In October, 2012, GIPP, NTSC and MAEC agreed to collaborate on a two year project under GIPP guidelines with Argonne National Lab as the lead U.S lab and Pacific Nuclear Solution is the lead U.S. industrial partner. The project formally begins on June 1, 2013.

The achievement of these objectives reduces the risk of proliferation and pursues the application of technologies for peaceful purposes.

The paper will introduce details of the two year project including the project structure, its participants, the two year project plan with objectives and milestones, a detailed description of the various LRW streams and details of the first year test program.

INTRODUCTION

Description of NTSC and MAEC

Nuclear Technology Safety Center (NTSC) was established in November 1997 as a result of an agreement between Department of Energy USA and Ministry for Science – Science Academy of the Republic of Kazakhstan "On Programs of Scientific-Research and Design Developments and Technology Exchange Programs for joint program on nuclear safety".

The main activity of NTSC includes:

- Scientific-technical support for the Kazakhstan Atomic Energy Committee
- Development and independent review of safety reports
- Project management in the area of nuclear safety and non-proliferation
- Independent review of the nuclear related projects and safety analysis reports
- Development of the regulations drafts in the area of nuclear safety and technologies
- Management of nuclear units designing
- Management of decommissioning of nuclear units
- Education and trainings in the area of non-proliferation and nuclear safety, including organization of international conferences and seminars

MAEC (MAEC-Kazatomprom LLP) is a united power and water producing complex whose main task is to provide electric power to the populated areas and enterprises of the Mangystau region and reserve electric power for the Atyrau region and to provide the city of Aktau with heat, power, drinking, hot and technical water and also to supply distillate to industrial enterprises.

The power complex structure includes three heat power plants HPP-1 HPP-2, HPP-3, and BN-350 reactor (since 1999 has been under decommissioning), a plant for distillate production and industrial water supply, heating mains and pipelines with central water supply units, a power transmission line with substations, a water intake structure, a repair works, a petroleum storage depot, a fuel-oil storage tank and other infrastructure facilities. The MAEC staff makes up 4,000 workers and specialists.

The history of the power complex is closely connected with the history of the exploration of mineral resources and oil deposits in Western Kazakhstan. The first units of HPP-1 were put into operation in 1962. As an independent facility the Mangystau Atomic Power Plant was established on July 1, 1968 and the first units of HPP-2 were put into operation in December of 1969. The BN-350 nuclear power reactor was launched in July 1973. In March of 1983 and October of 1984 the HPP-3 was launched. In those

years the electric loading achieved up to 750 megawatts at 1300 megawatts installed capacity of electricity generating facilities. Distilled water production created to 100 thousand cubic meters per day.

At present the power complex produces up to 40 thousand cubic meters of distillate per day and its electric loading is more than 500 megawatts. Operation of the power complex is supported by all required licenses for production activity. Due to a forecasted electric power deficit, the top-management of MAEC-Kazatomprom is now considering the construction of a modern gas-vapor power-generating unit with a capacity of 200 megawatts.



Fig. 1. BN-350 Building

Under the agreement the U.S. Department of Energy, National Nuclear Security Agency Office of Non-proliferation and International Security will provide funding and program management oversight to the partnership (Ref. 1).

The objectives of GIPP are:

- To engage former weapons of mass destruction scientists, engineers and technicians with U.S. national laboratories and U.S. industrial partners to jointly work on high technology commercial research and development projects.
- To create new technology sources and to provide long-term sustainable business opportunities for U.S. partners and host-country staff and institutions.

 To provide funding to the foreign scientists for implementation of the project and to the U.S. laboratory partner for technical and management oversight.

The two year project will be funded by GIPP with a total budget of \$ 700,000. Of the total \$ 490,000 will be distributed to participating MAEC scientists and staff based on the completion of quarterly milestones that are set by GIPP and the International Science and Technology Center (ISTC), Moscow. The remaining funds are allocated to ANL for its technical assistance and oversight.

Several departments and organizations will participate in the implementation of this project. The organizational structure is as follows:

Kazakhstan

- Kazakhstan Atomic Energy Committee
 - Provides government oversight as the Nuclear Regulator
- Nuclear Technology Safety Center
 - Provides technical and administrative oversight
- MAEC-Kazatomprom
 - o Participating scientists/engineers and staff:
 - 26 scientists and engineers
 - 35 assistants and staff

United States

- U.S. Department of Energy, Global Initiatives for Proliferation Prevention
 - o Provides funding for the project
- Argonne National Laboratory
 - o Project collaborator, technical and administrative oversight
- Pacific Nuclear Solutions, division of Pacific World Trade, Inc.
 - Industry collaborator, provides technology, training and technical assistance to MAEC

International Science and Technology Center, Moscow

• GIPP funding is provided to ISTC for distribution to the Kazakhstan participants; technical and administrative monitoring

Applied Technology and Waste Characterization

The solidification technology to be applied is Nochar's N series. Nochar's N series are a group of 3rd generation of polymers designed to immobilize all radioactive liquids including hydrocarbons, mixed sludge types, acids, alkaline, light alcohols and other aqueous waste for interim or final storage or incineration. The polymers absorb the liquid waste creating a solid form with little or no volumetric increase in a simple and safe process. The polymers are used in the U.S. Department of Energy complex and in international markets (Ref. 2 & 3).



Fig. 2. N polymer demonstration with 4 phase organic / aqueous waste stream

Table 1 shows the data on the amount of LRW, separated into phases, chemical composition and specific and total activity of LRW in the BN-350 storage tanks.

TABLE 1. Characterization of BN-350 LRW

N <u>o</u>	LRW volume in tanks			Salt	Chlorides	Specific	Total
				content		activity	activity of
						of Cs-	Cs-137
						137	nuclide
						nuclide	(error ±
						(error	30%)
						±30%)	
	m³	Phases	m³	g/l	g/l	Ci/l	Ci
1	753,5	Decantate	649,0	3,6	0,35	2,	13,629
						1x10 ⁻⁵	
		Sludge	104,5	5,0	0,71	1,2 x	1,254
						10 ^{- 5}	
2	370,8	Decantate	282,0	309,0	26,6	5,8 x	1635,6
						10 ⁻³	
		Sludge	88,8	467,0	19,5	1,7 x	1509,6
						10 ⁻²	

3	634,5	Decantate	591,0	267,0	17,8	1,8 x 10 ⁻³	1063,8
		Sludge	43,5	240,0	21,3	3,2 x 10 ⁻³	139,2
4	452,6	Decantate	400,0	128,0	14,2	8,9 x 10 ⁻⁴	356,2
		Sludge	52,6	139,0	23,1	9,9 x10 ⁻⁴	52,074
5	595,0	Decantate	492,5	146,0	15,9	3,3 x 10 ⁻³	1625,25
		Sludge	102,5	149,0	12,4	2,7 x 10 ⁻³	276,75
6	400,0	Oil	146,0			3,0 x10 ⁻²	2870,2
		Alkali	254,0	198,0	1,73	1,13 x 10 ⁻²	2870,2
7	10,6	Sludge	10,6				74,5
		Decantate	2414,5				4694,484
		Sludge	402,5				2053,378
		Oil	146,0				0,438
		Alkali	254,0				2870,2
	Total: 3217.0m ³						9618.5 Ci

Notes:

- 1. The results can be regarded as estimates.
- 2. The main activity in all sample forms radionuclide Cs-137.
- 3. Decantate means aqueous, sludge free.

BN-350 LRW is stored at Building 157 in above ground tanks. There are ten tanks of which five are in operation now. The tanks became operational in 1972. The tanks hold

a variety of waste with a mix of sludge types and gravel on the floor of the tanks. Approximately 400 cubic meters of sludge distillation residue exists with large amounts of chloride. A uniform layer of sludge covering 182 square meters will require the installation of hydraulic guns with video monitoring to dislodge and washout the sludge, thus increasing the volume of LRW.

In other tanks approximately 2,400 cu. meters of waste is in the form of decantate of distillation residues with high salt content and a large amount of chloride and oxalates which has caused increased corrosion of equipment and pipelines. Lubricating oil and a water-alkali-oil emulsion created from Na-K alloy processing are stored in other tanks. The estimated total volume of LRW is 3,217 cubic meters with total activity of 9,618.5 ci (35,588.5* 10-4 MBq).

Experiments

Various experiments were conducted in April, 2013, in the MAEC central laboratory using active (LLW) waste streams. Four waste streams were tested: machine oil, sodium hydroxide and 2 saline solutions containing sodium nitrate, sodium oxalate and sodium chloride with different salt content.

Test No. 1 – Machine Oil

The oil is a standard type with 1-2% of silica organic compound. Polymer N910 was placed in a glass beaker with a net weight of 50 grams. At a 1:1 ratio by weight, 50 grams of oil were poured slowly into the center of the polymer with the solidification process occurring immediately with no mixing. A significant amount of unused polymer remained, as seen in Figure 3. Additional oil was added to the polymer to fully utilize the polymer's capacity, at a 2:1 ratio (2 parts oil: 1 part polymer) and at a 3:1 (3 parts oil: 1 part polymer) ratio, as shown in Figure 4. Slow and short-term mixing was required at the 2:1 and 3:1 ratios. A 3:1 ratio is the outer limit for an acceptable and safe solidification for oil.



Fig.3. Machine oil solidification process

Fig.4. Solidified oil at 3:1 ratio

Test No. 2 – Sodium hydroxide

Sodium hydroxide is a highly caustic solution at 14 pH. The tests conducted on the active solution included the N960 polymer, for aqueous solutions. A 1:1 bonding ratio by weight was created with 50 grams of sodium hydroxide and 50 grams of N960. Figure 5 illustrates the immediate solidification of the solution at the top layer of the white polymer. As a result of the immediate solidification, a skin or solid layer of material formed, thus restricting the flow of the solution to the remainder of the polymer. Light mixing was required to combine the solution with the polymer. The solidification result of the 1:1 test was excellent. Further tests were conducted at 2:1 and 3:1 bonding ratios. These tests show the polymers capacity to absorb a large amount of solution while retaining a firm solidified mass, without any loose liquid or liquid release, as shown in Figure 6. Also, by extending the solidification ratio, the process is attractive economically and reduced the over volume of waste for final storage.



Fig.5. Sodium hydroxide in liquid form 3:1 ratio

Fig.6. Solidified sodium hydroxide at

Tests 3 & 4: Saline solutions with high salt content

Tests were conducted with two saline solutions that included sodium nitrate, sodium oxalate and sodium chloride. The first test included a solution with 150 g / liter of salt at 1:1 ratio, then again at 2:1 and 3:1 ratios. As seen in Figure 7, a skin developed immediately because of the immediate solidification of the solution. Light mixing was required to combine the solution with the N960 polymer with the result shown in Figure 8. A second set of tests were conducted with the saline solution and 450 g / liter of salt at 1:1, 2:1 and 3:1 bonding ratios. The final results on all five tests indicate excellent solidification models.



Fig.7. Saline solution with N960

Fig.8. Saline solution at 2:1 ratio

Final Waste Form for Disposal

Extensive work is underway to determine the final waste form for disposal that conforms to Kazakhstan waste management regulations. Various cement formulas have been tested with waste loading percentages of solidified materials to be determined. The best one or two encapsulation formulas will selected for testing under Task 3, as outlined below. Fig. 9 illustrates cementation test work.



Fig. 9. Cementation tests

Two Year Project Plan

The project is highly structured with tasks and milestones clearly defined. The scope of activity is categorized into three Task Groups. Rigorous data analysis and reporting is a part of each Task group. Quarterly reports are sent from MAEC-Kazatomprom to NTSC for review and translation, then to ISTC for review and administrative action, then to ANL and PNS for technical review and approval.

Task 1 includes the following activity:

- Development of the work program and schedule
- Review the polymer technology, its application experience and its application to BN-350 LRW
- Procurement and delivery of materials and equipment to MAEC

Task 2 includes the following activity:

- Sample testing of LRW streams: machine oil, sodium hydroxide and other aqueous solutions using N910 and N960 polymers
- Preparation of simulant streams for testing with cement and geo-cement stone materials
- Select the optimal ratio of LRW-oil: polymer solidification and dry cement mixture for encapsulation of LRW into a homogeneous matrix
- Select the optimal ratio of LRW-aqueous: polymer solidification, with coating material, and dry cement mixture for encapsulation of LRW into a homogeneous matrix
- Extensive testing of the LRW-oil solidified material combined with cement slurry to determine compatibility and final solid matrix
- Select the optimal ratio of LRW-sodium hydroxide solidification and combine with geo-cement components (kaoline clay, diatomite, blast furnace slag) for encapsulation of LRW into a homogeneous matrix
- Extensive testing of LRW-sodium hydroxide solidification combined with geocement to determine compatibility and final solid matrix
- Large scale testing of LRW simulants in a 10 liter container:
 - Oil and aqueous solidification materials in a cement slurry to determine the final matrix
 - Sodium hydroxide solidification in a geo-cement material to determine the final matrix
 - Determine large scale production processes for each type of LRW

Task 3 includes the following activity:

- Sample testing of the solidified cement forms and solidified geo-cement forms in compliance with GOST R 50926-96 and GOST R 51883-2002 requirements:
 - o Freeze-thaw resistance
 - Thermal cycling resistance
 - Radiation stability, requirement of 1 million gray irradiation tests on final waste form using Cobalt 60, gamma source irradiator
 - o Chemical stability, leach test in water

- Water stability test, 90 days in water
- Mechanical strength, compression and bending; compression tests conducted after each of the above tests are completed
- Mass ratio of fission isotopes
- Cut large scale cement and geo-cement forms, followed by mechanical tests
- Determination of final packaging: carbon steel drums

At the conclusion of the project a final and comprehensive report will be issued by MAEC.

CONCLUSIONS

Experiments carried out to date indicate that polymers N910 for organic waste and N960 for aqueous waste have shown good results for the immobilization of BN-350 LRW, especially with oil sludge.

Aqueous waste treatment at MAEC may have several options including geocement stone, a process tested by Kazakhstan's NTSC. Extensive testing with N960 polymer and its encapsulation with geocement will be conducted to determine if the total waste package is viable, meets the long-term storage requirements and is a cost effective option for the treatment of the sodium hydroxide waste stream.

U.S. technologies offered to MAEC are proven in the radioactive waste treatment sector and provide viable options to MAEC for the treatment and disposal of some of the BN-350 waste streams.

The GIPP program is a government to government mechanism to engage former weapons scientists in the peaceful use of new commercial technologies from U.S. industry and encourage joint collaboration of both parties to pursue the development of new designs and applications. The technologies developed may be applied in the U.S. Department of Energy as well as international markets.

Technical results of the first year's project work will be published and presented at future international waste management conferences.

REFERENCES

- 1. Argonne National Laboratory, Nuclear Engineering Division, Non-proliferation and National Security Program (NPNS). www.ne.anl.gov
- 2. "Proven Technologies for the Treatment of Complex Radioactive Liquid Waste Streams; U.S. Department of Energy and International Case Studies", Dennis Kelley, Pacific World Trade, and Dennis Campbell, Nochar, Inc. Conference on Alternative Materials for Radioactive Waste Stabilization and Nuclear Materials Containment, Barga, Italy, March, 2007

3. "Russia: Results and Prospects of Liquid Solidification Experiments at Rosatom Sites", Yury Pokhitonov, Vasiliy Babain, Vladislav Kamachev, V.G. Khlopin Radium Institute, Dennis Kelley, Pacific Nuclear Solutions. The 14th International Conference on Environmental Remediation and Radioactive Waste Management, Reims, France, September, 2011