Reworking of Legacy Radioactive Wastes at Al-Tuwaitha Nuclear Site

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Abstract

Seven rusty barrels of legacy radioactive wastes contained debris and located in Fuel Fabrication Facility (FFF) at Al-Tuwaitha Nuclear Site. Identification of legacy wastes comprised characterizing, classifying, over pack and relocating into specified accumulation zone of radioactive wastes inside (FFF) location. The results show that the radiological characterization of legacy wastes are contaminated with natural Uranium and have radioactivity of Low Level Waste (LLW). Over pack technique made the deformed packages agreed with the requirements of radioactive waste acceptance criteria, which reduced the surface contamination and the touched dose rate for each barrel by (99.97-99.99)% and (55.23-73.64)%, respectively, as well as, it will be very helpful in futuristic waste treatment by exploiting the open space for encapsulation processes to increase mechanical and shielding properties. The manner implemented was simple, economic and capable to make any releases to the environment to be restricted and subject to regulatory control.

Key Words: Over Pack; Legacy Wastes; Radioactive Waste Characterization and Al-Twaitha Site.

اعادة معاملة تركة من النفايات المشعة في موقع التويثة النووي

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وزارة العلوم والتكنولوجيا /
مديرية تصفية المنشآت والمواقع النووية المدمرة
بغداد–العراق

الخلاصة

سبعة براميل صدأة لتركة نفايات مشعة تحتوي على الانقاض وموضوعة في العراء في منشأة تصنيع الوقود الواقعة في موقع التويثة النووي.شملت عملية تحديد تركة النفايات على توصيف وتصنيف واعادة تغليف ونقل تركة النفايات الى منطقة التراكم المخصصة للنفايات المشعة داخل منشأة تصنيع الوقود. اظهرت نتائج التوصيف الاشعاعي ان تركة النفايات ملوثة باليورانيوم الطبيعي وواطئة النشاط الاشعاعي. ان تقنية اعادة التغليف جعلت الرزم المشوهة موافقة لمتطلبات معايير قبول النفايات المشعة وخفضت التلوث السطحي والجرعة الاشعاعية الملامسة لكل برميل بنسبة (99.97–99.99)% و(55.23–73.64) % على التوالي، بالاضافة الى انها ستكون مفيدة جدا في المعالجة المستقبلية للنفاية من خلال استغلال الفراغ المفتوح في عمليات التغليف المحكم لزيادة التدريع. ان الاسلوب الذي طبق كان سهل واقتصادي وقادر على جعل الانبعاثات الى البيئة مقيدة وخاضعة للسيطرة التنظيمية.

الكلمات المفتاحية: اعادة التغليف؛ تركة نفايات؛ توصيف النفايات المشعة وموقع التويثة.

Introduction

Reworking has had a fairly narrow definition, comprising specific processes such as over packing, or cutting up of the waste package and repacking…….etc. This identification gives it a wider definition as a process involving physical intervention to packaged waste arising from deviation from the planned storage, treatment, or intended disposal process for that packaged waste. Reworking might become necessary if a waste package were to deteriorate or be damaged, or become out of specification due to external factors (Environment Agencies, 2003).

A waste package can be deformed from the effects of gas pressurization. Also, chemical changes in the package can damage the waste container, e.g. from chemical reactions leading to expansion of the waste form, or corrosion of the container. Under these conditions, over packing or other more invasive methodologies will be necessary in order to meet the ongoing storage, transport or disposal waste acceptance requirements (HSE, Environment Agency 2005).

An outer package is used to prevent leakage and to provide additional shielding or handling features. This outer package is referred to as an over pack. The IAEA recommend that packages with serious container structural problems must always be over packed. Over packs may mitigate mechanical corrosion connected problems associated with waste packages. It can also help reduce surface dose rate levels to acceptable levels. Various types of over pack are currently in use. As an example, 200 liter drums containing radioactive waste can be inserted into either 400 liter drums or a large rectangular container manufactured of steel or reinforced concrete. The open space in these over packs may be grouted with cement to increase the mechanical and shielding properties (Krumbach, 2005).

There are many sites in Iraq which have been used for nuclear activities and which contain potentially significant amounts of radioactive waste. The main nuclear site is Al-Tuwaitha, the former nuclear research center. Many of these sites suffered substantial physical damage during the Gulf Wars and have been subjected to subsequent looting (Jarjies, *et al.* 2007). Al-Tuwaitha Nuclear Research Center served as the foundation of Iraq's nuclear research and development from 1967 until its final closure by U.S.-led coalition forces in 2003. Secret operations at Al-Tuwaitha, combined with the bombing of nuclear facilities and the subsequent looting by local residents, have contributed widespread radioactive contamination (Ronald, *et al.* 2009).

Rusty barrels wastes considered a legacy waste which located at Fuel Fabrication Facility (FFF); one of the Al-Tuwaitha Site Facilities. Rusty barrels need reworking and over pack because of packages deterioration due to bad storing circumstances which became out of specification (Environmental Agency, 2005). On April 2013, meeting was held between the IAEA experts and Iraqi Decommissioning Directorate (IDD) managers to discuss decommissioning plan for the FFF. One of the meeting recommendations was identification and removes the legacy wastes (Kurt and Jeremy, 2013).

The aim of this work is to identify the legacy wastes by concepts of characterizing classifying, over pack, and relocating into specified accumulation zone of radioactive wastes. At IDD, the generated radioactive wastes are packaging according to waste package specifications devised by Radioactive Waste Treatment Directorate (RWTD) which derived from IAEA specifications of waste packaging (IAEA, 2006). Therefore, the waste package specifications must be compatible with the RWTD plans for wastes repository.

Experimental work

Site description:

The work has done at Fuel Fabrication Facility (FFF) which considered as one of the nuclear facilities in Al-Tuwaitha Site, 20 Km south-east of Baghdad. It used to manufacture the nuclear fuel (natural UO_2) with requirements of the nuclear technology in laboratory scale. The facility designated to prepare 4.1 m pins and to assemble fuel element of maximum 4.5 m. It operated in 1981 and destroyed in 1991 in the Second Gulf War (Qusay, 2013).

A waste from unknown stage of the FFF life was generated and packaged in seven-200 liter carbon steel barrels. The main reason for this may be to stabilize wastes at the lowers risks by reducing the likelihood of wastes leakage into the environment. Barrels were outdoors located for many years (Fig. 1) and had subjected to severe weather variations.

Fig. (1) Rusty Barrels

Factors of the bad Storing and the Existed Radionuclide Nature, Created theH Barrels Corrosion.

Materials and Methods

Materials used to fulfill reworking of legacy waste were:-

1. LUDLUM (2241) used in field measurement and has a prop of (NaI) detector (type 44-10) used for measuring dose rate by micro-Sievert per hour (µSv/h) unites.
2. LUDLUM (3030) scalar meter for measuring alpha/beta emitters with ZnS (Ag) scintillation detector used to measure surface contamination of each barrel by smear tests.
3. Vacuum cleaner to suck up and containerize the generated dust and aerosols.
4. Gamma spectrometer with semiconductor detector of high purity germanium used for laboratory analyzing of homogeneous contaminated debris samples.
5. Barrels of 400 liter in volume, made of carbon steel, painted with brightly colored (yellow) and have closed sealed, used for over pack and containerize the rusty barrels wastes.
6. Freight container has dimensions $6^m \times 2.5^m \times 2.5^m$ and have closed sealed, used as accumulation zone for radioactive waste.
7. Thick nylon bags to ensure work accomplishment with a safe manner, used to contain the rusty barrels with their contaminated debris.
8. Nylon stratum was used as ground cover. The nylon stratum was thick and waterproof type material capable of withstanding the work activities involved without tearing or ripping.
9. Assistant Instruments like forklift, digital camera, 500 Kg maximum load scale, cleaning dustpan, shovel and cleaning brushes.

Held detector (LUDLUM (2241)) and laboratory detector (LUDLUM (3030)) have subjected to a daily calibration before work. The calibration has done through surveying of standard sources have a unique radioisotope of Sr-90, Am-241 or Cs-137 by the detectors then compare the results with the attached certificate for each detector from its manufactured company. While a standard source consists of mixed radionuclide had used to calibrate the Gamma spectrometer laboratory analyzer before sample measuring.

Experimental procedure

Reworking of legacy wastes comprised characterizing classifying, over pack, and relocating into specified accumulation zone of radioactive wastes.

All rusty barrels were opened and the waste was inspected for characterization purposes. Radioactive waste characterization was an important obligation in waste management (Savidou, *et. al.* 2007) to categorize waste and to compare with the limitations of national criteria (Radiation Protection Center, 2012) which agreed with the international limitations (IAEA, 2004 and IAEA, 2005). Each rusty barrel was served from all sides by (LUDLUM (2241)) to measure the touched dose rate then, swipes were taken from the outer sides and sent to the IDD laboratory to identify the amount of surface contamination. Ultimately, a sample was taken from different heights of each rusty barrel and mixed to make 1 Kg of homogeneous sample. Samples were sent to the Central Laboratories Directorate (CLD) to recognize the type and concentration of radioisotopes in the waste material by using gamma spectrometer with a semiconductor detector of high purity germanium. Laboratory measurements indicate ground contamination with U-238, which mainly an Alpha emitting isotope, hence hard to detect and investigate. Field and laboratory detection for U-238 was mainly conducted through Pa-234m at Gamma energy line 1001keV. The analyzing procedure was according to the IAEA-TECDOC-1092 standards (IAEA, 1999).

The workers under control of personal radiation protection equipment implemented the simple and economic procedure to overpack the rusty barrels. The work began by carpeting the area around the rusty barrels by thick nylon stratum. The boundary of the surrounded area was as small as possible but large enough to allow persons and equipment to access the work area for accomplishment work in a safe manner. Each rusty barrel covered by two thick nylon bags and overpack by 400 liter barrel. The combination was rotated and slipped inside the over pack barrel, and then remove the contamination from the nylon stratum carefully. After that, the nylon bags were twisted and the barrel was sealed. Contact dose rate, surface contamination and weight was measured for each waste barrel then, the barrel marked with suitable attention signs (radiation trefoil) and tags (Fig. 2). All repacked containers were transferred into transitional accumulation zone (freight container) until completion of submitting's administrative to RWTD which is the responsible for the final radioactive waste status.

Carpeting the surrounding area and covered the rusty barrel by nylon bags

Over pack the rusty barrel by 400 liter barrel.

Rotated and slipped the combination inside the over pack barrel.

Measuring of weight and dose rate of each barrel

Sealing, marking and relocating barrels in 6m freight container

Fig (2) Indicates Steps of Identification and Remove of Rusty Barrels

Results and Discussion

The results are listed in Tables (1 and 2)

Table (1) Results of Gamma Spectrometer Analyzer*

Barrel code	Weight (Kg)	Activity Concentration (**Bq/Kg)**
D1	**185**	**16064.4**
D2	**185.8**	**53552.8**
D3	**170**	**24260.6**
D4	**202.2**	**81867**
D5	**185.7**	**15504.4**
D6	**194.1**	**10480**
D7	**119.1**	**77610.3**

***The physical status is debris.**
***The radioisotope is natural Uranium.**

Table (2) Characteristics of Legacy Waste before and after Reworking

Barrel code	**Before over pack**		**After over pack**		**Reduction ratio %**	**Dose attenuation %**
	Contact dose rate µSv/h	**Surface contamination Bq/cm^2**	**Contact dose rate µSv/h**	**Surface contamination Bq/cm^2**		
D1	0.411	38.68	0.184	0.0054	99.98	55.23
D2	0.837	45.19	0.275	0.0067	99.98	67.14
D3	0.648	43.11	0.200	0.0058	99.98	69.13
D4	1.529	52.82	0.425	0.0052	99.99	72.00
D5	0.375	33.08	0.154	0.0045	99.98	58.93
D6	0.246	23.67	0.086	0.0056	99.97	65.04
D7	1.385	50.20	0.365	0.0050	99.99	73.64

The touched dose attenuation percentage and reduction ratio percentage were calculated by using the relationships (1) and (2) respectively.

$$\text{Dose attenuation } \% = \frac{\text{dose rate before over pack} - \text{dose rate after over pack}}{\text{dose rate before over pack}} \times 100 \quad \text{.....(1)}$$ (IAEA, 2006)

$$\text{Reduction ratio } \% = \frac{\text{surface contamination before over pack} - \text{surface contamination after over pack}}{\text{surface contamination before over pack}} \times 100 \quad \text{.....(2)}$$ (IAEA, 2006)

Table (1) showed that legacy wastes of rusty barrels were debris and contaminated with natural Uranium exceeds the limitations mentioned in national criteria (1000 Bq/kg). The waste was classified as low level waste because the touched dose rate was less than 2mSv/h and the maximum specific activity less than 4GBq/t for isotopes have alpha emitters (IAEA, 2009).

While Table (2), illustrated the over pack manner contribute to contain and mitigate the contamination of rusty barrels wastes. Over pack is a term normally used to describe the placing of a smaller non-compliant package inside a larger package. It was caused reduction ratio in surface contamination (99.97-99.99)%, attenuated the radiological dose rate (55.23-73.64)% and made the deformed packages to be adequate for requirements of radioactive waste acceptance criteria. As well as, the open space in these over pack barrels may be helpful for encapsulation (during waste treatment in RWTD) to increase mechanical and shielding properties.

In theoretical stage, the waste package performance is affected by repository conditions. The legacy waste barrels located in unclosed area, therefore, that leads to undergo barrels to wide fluctuations of temperature and humidity. Consequently, it caused corrosion of barrels. Which agreed with the interpretation mentioned in reference (King, 2007) that corrosion could be initiated if the surface wets non-uniformly during saturation. Deliquescence of water on salt crystals or other contaminants on the barrel surface will lead to formation of macro and micro water droplets connected by a thin adsorbed water layer. The cathodic reduction of O_2 will occurs on and around the micro-droplets and the periphery of the macro-droplet, since these regions experience the highest O_2 flux. The anodic reaction will be focused in the center of the macro-droplet where O_2 replenishment is limited. This spatial separation of anodic and cathodic processes will only operate during the aerobic phase.

Another factor, the interaction between waste barrel and waste form might be contributed barrels corrosion and that agree with opinion of (Shoesmith and King, 1999) which mentioned that gamma radiation leads to an increase in corrosion rate.

Conclusion

The manner implemented to rework rusty barrels was simple, economic and makes any releases to the environment to be restricted and subject to regulatory control.

Over pack manner is successful for containing and mitigating contamination and it will be very helpful for waste treatment in RWTD in the future by exploiting the open space for encapsulation processes to increase mechanical and shielding properties.

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