## Study of the Polymer Coating for Detecting and Surface Decontamination of Uranium- 11494

Pham Quynh Luong Institute for Technology of Radioactive and Rare Elements (ITTRE) Vietnam Atomic Energy Institute (VAEI)

#### Abstract:

Strippable polymer coating is one of the methods for effective surface decontamination. It has been developed in both detecting and removing the radioactive isotope and heavy metal elements from contaminated surfaces. A polymer coating is produced to be sprayed or brushed on contaminated material of uranium. The places of U contamination is shown by color change of polymer coating. As the polymer coating is dried up to form a strong film, the contaminations are absorbed in to the coating and contaminated surfaces are cleaned by removing the film.

#### Introduction.

Radioactive materials are used extensively in heath care, industries, agriculture, geology and research activities...etc, particularly in nuclear industries. The mining of uranium ore and its processing are activities which give rise to significant quantities of contaminants. The main representatives of naturally occurring radioactive elements are uranium, radium, thorium, radon and their daughter decay products. The nuclear fuel cycle encompasses all sectors of nuclear energy, such as uranium ore mining and milling, uranium hexafluoride production, uranium enrichment, production of fuel rods and assemblies, operation of nuclear reactor, storage and reprocessing of burn out fuel assemblies, decommissioning of nuclear power plants and reclassification, and treatment and storage of radioactive waste. During the operation and handing surface of facilities, equipments, glove boxes, floors, walls may get contaminated with radioactive isotopes. Surface decontamination must be treated in time to protect workers who contact with it and finally it may become airborne and may enter in to body of workers due to inhalation and ingestion. Thus any effective decontamination process for this environment is very important. Various surface decontamination techniques are used in nuclear industry. There are many decontamination techniques. Each one has advantage and disadvantage. Decontamination is only beneficial where the value of the recovered object or advantage gained is greater than the cost of the decontamination process and the treatment, conditioning, transport and disposal of the secondary waste produced. Decontamination of solid waste (e.g. tools, instruments and plant items) is most commonly employed with the objective of reuse. Decontamination may also be employed to reduce contamination to levels acceptable for disposal as non-radioactive waste, to minimize personnel exposure during subsequent waste treatment operations or for product recovery.

Strippable polymer coating is one of the methods for effective surface decontamination. Strippable polymer coating are polymeric solutions or gel that can applied to a contaminated surface by spraying or pasting. These coating consist of strippable polymetric compositions containing blends of polymers, copolymers, plasticizer, decontamination reagent and colorimetric indicator. They are mixed fully in to a decontamination gel, this gel can be sprayed or pasted on to contaminated surface. After some hours, these coating were dried and form strong film. This film showed contaminated areas on the surface by colour change of film (from pink to green). Contaminants are absorbed or chemical chelating in to film that can easily be peeled off

the surface. Thus the surface is decontaminated by removing the film. In this study, we present the results obtained in our laboratory from product the decontamination coating for detection and removal of uranium from stainless steel, mild steel, painted cement and nickel surfaces

# Experiment

# 1. Preparation of strippable polymer solution

In our experiments: 170g of polyvinyl alcohol was dissolved in 774 g distilled water in a 1- L beaker at  $50^{0} - 60^{0}$ C. They were good stirred mechanical using a stainless steel. After all of polyvinyl alcohol (PVA) were dissolved fully that formed white heterogengeous solution. Added 50 g glycerine, 5g ethylenediaminetetraacetic acid disodium salt (EDTA), 0.005g arsenazo III (in 1ml distilled water) to the polymer solution (all were laboratory reagent). Followed by 45 min of additional stirring. The red – violet, viscous gel solution was formed. Sited the beaker for overnight at room temperature without stirring for defoamed. Then this mixture was transferred to a clean polyethylene bottle for storage.

# 2. Preparation of polymer coating for the study of decontamination agent effectiveness.

The aqueous  $PVA/H_2O$  blend was prepared as described in preparation of strippable polymer solution section above. Added 50 g glycerine, 5g potential decontamination agent, 0.005g arsenazo III (in 1ml distilled water) to the polymer solution (all were laboratory reagents). Followed additional stirring for 45 min. The red – violet, viscous gel solution was formed. Sited the beaker for overnight at room temperature without stirring for defoamed. Then this mixture was transferred to a clean polyethylene bottle for storage. The potential decontamination agents are: EDTA; acid oxalic; sodium carbonate; ammonium hydro phosphate.

### 3. Surface decontamination in the laboratory.

In a typical decontamination procedure, uranium (uranium oxide in 0.1 M HNO<sub>3</sub>) contaminated surface were prepared by evaporating uranium solution on to a variety of coupon. Each coupon was analyzed via  $\alpha$ ,  $\beta$  counting by MPC- 200 – PC Desktop Alpha/Beta Counting before being treated with the strippable coating. All coatings were allowed to dry (at least 24 h) before removal. The our coating displayed a distinctly color change in the positions of U contamination. The coatings were stripped, and the coupons were again analyzed via  $\alpha$ ,  $\beta$  counting. Using the counting rates before and after decontamination, decontamination efficiency (K) were calculated using formula 1, where  $\alpha_0 \& \beta_0$  (A<sub>0</sub>) is the counting before decontamination and  $\alpha_f \& \beta_f$  (A<sub>f</sub>) is the counting after decontamination.

Decontamination efficiency of a material is given by the decontamination coefficient K, which is calculated by formula:

$$K = [(A_0 - A_f)/A_0] * 100$$
(1)

Where: K - decontamination efficiency

A<sub>0</sub> - radioactivity (cpm) of material surface before decontamination,

A<sub>f</sub> - radioactivity (cpm) of material surface after decontamination

The polymer coating was used to decontaminate several different types of contaminated surfaces with uranium from uranium oxide in 0.1 M HNO<sub>3</sub>. and uranium from Nong Son uranium ore processing (in Vietnam). Decontamination efficiency (K) for stainless steel, mild steel, nickel, and painted cement were measured.

# **Results and Discussion**

## 1. The abilities of several decontamination agents for uranium

The abilities of several decontamination agents to absorb uranium were investigated. To determine the effectiveness of a potential agent, the surface of stainless steel coupon was contaminated with U. The contaminated surface was pasted with the polymer coating containing a potential decontamination agent (0.5% by mass). Each experiment was replicated 3 time. The results were summarized in table 1. After these studies were completed, it was determined that EDTA was the most effective decontamination agent for this particular coating. This decision was based on both the effectiveness and the ability to detect of U. The coating with another potential agents have no colorimetric response to U as the indicator is arsennazo III. This decontamination agent will be used in the polymer coating for all our studies.

Table 4. The study decontamination efficiency of potential decontamination agents for uranium

Potential	Before decontamination			After decontamination			Decontamination
agents	$\alpha_0$	$\beta_0$	$A_0 = \alpha_0 + \beta_0$	$\alpha_{\mathrm{f}}$	$\beta_{\rm f}$	$A_f = \alpha_f + \beta_f$	efficiency K (%)
A1	356	937	1293	3.5	34	37.5	97,1
A2	347	948	1295	4.2	30	34.2	97,4
A3	312	921	1233	3.1	32	35.1	97,2
P1	265	791	1056	3.0	57	60.7	94,3
P2	410	740	1150	4.9	41	45.9	96,0
P3	311	927	1238	3.6	38	41.6	96,6
N1	316	1010	1326	8.2	82	90.2	93,2
N2	250	995	1245	6.5	85	91.5	92,7
N3	257	950	1207	5.8	90	95.8	92,1
E1	368	738	1106	1.7	13	14.7	98,7
E2	354	781	1135	1.4	10	11.8	99,0
E3	362	770	1132	1.3	11	12.3	98,9

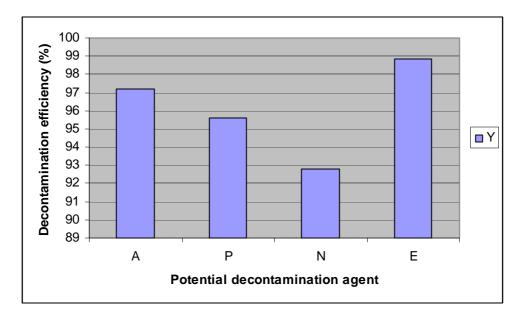
Where: A : Acid oxalic

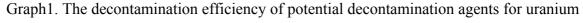
P: Ammonium hydro phosphate.

N: Sodium carbonate;

E: ethylenediaminetetraacetic acid disodium salt (EDTA),

The decontamination efficiency of potential decontamination agents for uranium were showed in graph 1





# 2. The visible detection limit for U

The uranium was detected by a visible color change from pink to green. Detection limits were measured as the smallest contaminant concentration on a specific surface that produced an observable colorimetric change. The visible detection limit for this coating is the smallest amount of U that will cause a *visible* color change. It is very important because this limit determines how small of an amount of U will actually be detected by a worker on-site. Surfaces of stainless steel were contaminated with specific amounts of uranium contaminant, as uranyl nitrate, and then pasted with the polymer coating. If a visible color change can observed, it was noted, and then a smaller amount of contaminant was tried. This process continued until the amount of uranium used resulted in no color change. The results were reported in table 2, the visible detection limit of polymer coating for uranium on stainless steel is around  $0.2 \ \mu g/cm^2$ , the result indicated that this coating is very sensitive to small amounts of uranium.

U concentration (µg/cm <sup>2</sup> )	visible color change?	U concentration (μg/cm <sup>2</sup> )	visible color change?
0.01	no	0.2	yes
0.02	no	0.3	yes
0.05	no	0.5	yes
0.1	no	1.0	yes
0.15	no	2.0	yes

Table 5. Determination of the visible detection limit for U on stainless steel using our decontaminating polymer coating

### 3. Surface decontamination.

To measure the decontamination effectiveness of the polymer coating on the different kinds of surface, the contaminated surface were prepared by evaporating uranium solution on to a variety of coupon. Each coupon was analyzed via  $\alpha$ ,  $\beta$  counting by MPC-200 – PC Desktop Alpha/Beta Counting before being pasted with the polymer coating. All

coatings were allowed to dry (at least 24 h) before removal. The our coating displayed a distinctly color change (from pink to green) in the positions of U contamination. The coatings were stripped, and the coupons were again analyzed via  $\alpha$ ,  $\beta$  counting. Using the counting rates before and after decontamination, decontamination efficiency (K) were calculated using formula 1, where  $\alpha_1 \& \beta_1$  (A<sub>0</sub>) is the counting before decontamination and  $\alpha_2 \& \beta_2$  (A<sub>f</sub>) is the counting after decontamination.

Decontamination efficiency of a material is given by the decontamination coefficient K, which is calculated by formula 1

The decontamination ability of our coating and 1101 decongel to several different types of surfaces contaminated for two kinds of uranium sample was studied. Uranium from uranium oxide in 0.1 M HNO<sub>3</sub> and uranium from Nong Son uranium ore processing (from uranium mill in Vietnam). They are studying in the laboratory of Institute for Technology of Radioactive and Rare Elements (ITTRE). The uranium solution contains 1mg/ml U; 4mg/ml Fe; 2mg/ml Al, and small amount of elements: Ca; Mg; Th; rare earth elements....

This study to indicate that uranium could still be high efficiency decontaminated and detected in the presence of the interfering elements.

The results are shown in table 2. The results showed that decontamination efficiency of our coating as high as 1101 decongel. But the our coating can detect uranium on the surface by color change of the coating (from pink to green), the 1101 decongel (blue) can not detect U. In addition our coating can decontaminate and detect thorium on the surface of difference material. The decontamination efficiency up to 99.5 % on the stainless steel surface

Surface	Coating	K (%)		
		U in uranyl nitrat	U in uranium ore	
		$(250 \ \mu g/cm^2)$	processing	
Stainless steel	our coating	98.7	97.5	
Stainless steel	1101 decongel	96.8	96.2	
Mild steel	our coating	93.2	90.6	
Mild steel	1101 decongel	95.4	94.2	
Nickel	our coating	93.7	92.4	
Nickel	1101 decongel	92.3	93.2	
Painted cement	our coating	75.4	72.3	
painted cement	1101 decongel	82.5	80.6	

Table 2. Decontamination efficiency of the coating on difference material

#### Conclusions

The polymer coating that we have developed actually meet the needs to surface decontamination in nuclear facilities. They are not only capable of decontaminating contaminated surfaces with high efficiency but also have behavior by indicating the positions of U, Th contamination. This is very important, because contaminated portions of the coating can be separated from uncontaminated areas and disposed of or treated accordingly. Furthermore, surface decontamination techniques by polymer coating can save costing for secondary waste processing that produced in decontaminating.

In Vietnam, radiation and radioisotopes have been applied in health care, agriculture, industry, geology, mining, meteorology, hydrology, education, research ... etc.

Especially in order to meet the energy demand in the future, the first nuclear power plant (NPP) will be put in operation in 2020 with capacity of 2000 MW. The application a surface decontamination technique with high efficiency and cost is very important, because the application and development of atomic energy in peace must ensure absolute safety for people and environment.

We hope that this study to developed the efficiency decontamination material that will able to apply in nuclear facilities of Vietnam in the future.

Reference:

1. S.V. Srao, K.B. Lai. *Surface decontamination studies using polyvinyl acetate based strippable polymer*. Journal of radioactive analysis chemistry. May, 2003.

2. Jen-chieh Chung, Che-Nan Chen Kon-MinLin Tsong-YangWei. Decontamination

*method of metal surface contaminated by radioactive element*. U.S patent application 5886.

3. H. Neil Gray, David E. Bergbreiter. *Application of Polymeric Smart Materials to Environmental Problems*. Environmental Health Perspective. Vol 105.supplement1. Feb 1997.

4. Jan Severa, Jaromir Bar. Handbook of radioactive contamination and decontamination