Chemical Decontamination of Goiania Materials Containing ^{137}Cs

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In 1987-1988 Brazilian Atomic Energy Commission (CNEN) had to decontaminate Goiania, capital of State of Goias, in middle of Brazil, due to the radioactive accident, caused by a group of people, who robbed a ^{137}Cs source from a closed radiotherapy facility, and dismounted it, at work in a junkyard, intending to get money by selling its parts. Some people took the CsCl contaning material (1300 Ci ^{137}Cs) at home, fascinated by the “blue light” (Cs spectral emission, plus “Bremstrahlung”, “Cerenkov Radiation” and chemical fluorescence) thinking that they had a handful of valuable precious stones, that even produced light and brilliance. When the people get sick, they were brought to Hospital. A woman washed out the fluorescent material to the garden and took the contaminated Pb cylinder to Sanitary Head Office. CNEN was called, and we were responsible to decontaminate a large spectrum of materials contaminated with ^{137}Cs, heading a group of CNEN technical people, under emergency situation, under pressure, in hurry. We had to decontaminate water, urine and feces, bank notes, cement, granite, walls and roofs of houses, soils, asphalt, schools and hospital, the source device, and rooms, beds and bathrooms of Hospitals, as well as hands and feet of people. Some information about the chemical processes and decontamination work was published in the CNEN-IAEA Goiania Report (1) , based in our internal report, of 1988. In the present work we present and discuss the essentials of chemical decontamination of different materials contaminated by ^{137}Cs and the way we applied the processes, within the contour conditions : weather, availability of chemical products, always under pressure of time and public exposure, intending to publish it without limitation of that formal report. We realized that published reports proposing methods to decontaminate materials and a city (Harshaw Report) were too “soft”, time consuming and with low yields for that situation we found in Goiânia. The processes had to be much more effective and faster. It is not only necessary to substitute Cs^+ ions for NH_4^+ or K^+, using NH_4Cl, but it is necessary initially to attack chemically materials to liberate Cs^+ and to remove these ions by ion exchange in solid materials, to achieve high yields and decontamination factors. We used $FeKFe(CN)_6$, called “K Prussian Blue” (KPB) and not $K4Fe(CN)_6$ (PB). We compared KPB produced by Brazilian Navy Laboratory with PB paint pigment, USA and PB of Radiogardase capsules (Heyl Laboratory, Germany, approved by USA Public Health in 2003. Laboratory analysis of KPB showed chemical composition $Fe(III)K(Fe(CN)_6)$, quantitative substitution of Cs by K ions in aqueous suspension, Solubility Product 1.10^{-12} M^3 , compared wit 1.10^{-40} for PB, and absence of free cyanide ions. Experiments with 10 mL urine of people, contaminated with ^{137}Cs, mixed with 200 mg of PB or KPB, and slow agitation, until equilibrium was established, measuring with gamma / beta radiometry, showed > 99,8% Cs removal for KBP and < 28% removal for PB (USA paint pigment and Radiogardase). We produced KPB at the site, inside of cation exchange resins, (Amberlite IR 150, sulfonated PE-DVB), designated by RH, by reacting the RH resin directly with $FeCl_3$, followed by $K_4Fe(CN)_6$ solution, in 60 to 100 L plastic recipients. The RK resin containing KPB has very high specific surface and larger external dimensions, when compared with KPB powder. RK-KPB we used to decontaminate water and urine, in columns, and feces, in batch operations. We also produced cotton towels and “T-shirts”, and RK-KPB containing creams for use by contaminated people and polyurethane containing KPB, to decontaminate soils at the field. To remove Cs ions from cement, soils and clay or dust containing material, RK-KPB was combined with a strong supply of H^+, K^+ and Al^{3+} ions from a $KAl(SO_4)_2$ solution , acidified with aqueous HCl (propyleneglycol HCl solutions, for less polar materials, or wax or asphalt coated materials) HF for granite .We decontaminated the source structure, first removing the paint coating with solvents, before treatment with KPB/Al/K/acid. The structure weighed 304 Kg, without stainless steel and Pb collimator cylinder. Painted materials sometimes contain PB pigments, and it is better to use alkaline solutions to decompose PB, to liberate Cs. Soils with high exposure dose at surface layers were partially decontaminated to allow people to work at place, using polyurethane and cotton towels containing KPB, after soaking the place with $HCl-KAl(SO_4)_2$ solutions. KPB is much more effective than PB because of much higher distribution coefficient for KPB, due to Cs x K exchange, thermodynamically favorable, in comparison with Cs x Fe(III) / exchange. Resin/KPB is very effective to take Cs^+ directly from hands and feet, instead of ingesting Radiogardase capsules, waiting Cs to go in the blood stream, to be exchanged at the intestines. Al^{3+} ions coagulate colloidal materials, H^+ attack oxides and carbonates, K^+ substitute Cs^+ and KPB exchanges K^+ by Cs^+, removing them from solution. These processes are much more effective than chemical treatment with NH_4Cl, or even with PB alone and should be recommended for other cases of contamination of materials or people, ^{137}Cs, ^{87}Rb, or Tl , hoping that they would not be necessary. $^{137}CsCl$ radiotherapy sources are much more dangerous than ^{60}Co sources, due to the high water solubility of CsCl and to similarity of Cs and K, to incorporate ^{137}Cs by organisms.

[1] Medeiros, J.A., Chemical Analysis Report : Laboratorio de Análise Mineral, CNEN (1987)
[2] Goiania Accident : CNEN – IAEA Report (1988)