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TECHNICAL ANALYSIS OF ON-SITE DISPOSAL OF SPACE GRADE PLUTONIUM WASTE

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ABSTRACT

The Savannah River Site is considering disposing of a relatively small volume of waste containing a relatively high radioactivity content of plutonium 238 from the production of power sources for the space program in on-site disposal units. A major part of the overall program is to demonstrate that on site disposal can be done while meeting all of the applicable regulatory criteria.

INTRODUCTION

The waste in question consists of the residual material used in the production of power sources for the US space program. It contains concentrations of Pu-238 that exceed the regulatory definition of transuranic waste, i.e., 3.7KBq/g (100 nCi/g), which means that in the normal course of events it should be sent to the Waste Isolation Pilot Plant in Carlsbad NM for disposal. However, in order to meet the transportation requirements for off site shipment, the waste in question must be characterized and repackaged. The nature of the Pu-238 material creates some very special concerns with regard to worker safety during handling which is the primary reason for considering on site disposal options that would eliminate the need to open existing waste packages.

In order to provide a defensible basis for a decision on the disposal of Space Grade plutonium waste, a technical analysis is being developed. This process uses the techniques developed at SRS to carry out performance assessments required for disposal of low-level radioactive waste. This paper describes the application of a simplified version of this methodology to provide confidence that this waste can be safely disposed at the Savannah River Site while meeting the various regulatory limits that might be imposed.

PERFORMANCE CRITERIA

A number of regulatory paths are being considered that might lead to on site disposal of the waste in question. A discussion of this topic is the subject of a separate paper [1]. Whatever course we decide to take, it will require a demonstration that the waste can be safely disposed. The specific performance criteria considered in this study are discussed below

DOE Order 435.1 Performance Objectives for Low Level Waste Disposal

At a minimum, any radioactive waste disposed at SRS will have to meet the performance criteria for low-level waste disposal as set forth in DOE Order 435.1 [2]:

Production and support facilities shall be closed so that a reasonable expectation exists that the following performance objectives will be met:

- Dose to representative members of the public shall not exceed 0.25 mSv/year (25 mrem/year) total effective dose equivalent (EDE) from all exposure pathways, excluding the dose from radon and its progeny in air.

- Dose to representative members of the public via the air pathway shall not exceed 0.10 mSv/year (10 mrem/year) total EDE, excluding the dose from radon and its progeny.

- Release of radon shall be less than an average flux of 0.74 $Bq/m^2/s$ (20 $pCi/m^2/s$) at the surface of the facility. Alternatively, a limit of 0.019 Bq/L (0.5 pCi/L) of air may be applied at the boundary of the facility.

In addition to the performance objectives, the Order requires, for purposes of establishing limits on the concentrations of radionuclides that may be disposed of near-surface, an assessment of impacts to water resources and to hypothetical persons assumed to inadvertently intrude into the low-level waste disposal facility. Table I lays out the performance measures and the associated points of compliance.

USDOE Order 435.1 states that "The performance assessment shall include calculations for a 1,000-y period after closure of potential doses to representative future members of the public and potential releases from the facility to provide a reasonable expectation that the performance objectives identified in this Chapter are not exceeded as a result of operation and closure of the facility."

Intruder Analysis

USDOE Order 435.1 provides a performance measure pertinent to impacts to hypothetical persons who are assumed to inadvertently intrude into a closed facility which specifies that calculated annual total EDE to such individuals not exceed 1 mSv (100 mrem) for chronic exposure scenarios. For acute exposure scenarios, calculated doses are not to exceed 5 mSv (500 mrem) total EDE. Institutional controls are assumed to be effective in deterring intrusion for at least 100 y following closure of the facility. Passive controls, in the form of engineered barriers or features of the site, can be claimed as further deterrents to intrusion.

In general, the chronic exposure scenarios address reasonable and credible pathways. However, consumption of groundwater and crop irrigation are exposure pathways that are excluded from the intruder analysis [3]; impacts of groundwater contamination are evaluated separately in this study.

Groundwater Analysis

USDOE Order 435.1 requires an analysis of groundwater concentrations of radionuclides leached from the facility in order to address both the all-pathways performance objective and the water resources impact assessment requirement. Protection of the public according to the stated performance objectives requires that calculated annual dose to a hypothetical future member of the public shall not exceed 0.25 mSv (25 mrem) total EDE from all exposure pathways, including potential ingestion of groundwater. The point of compliance is the point of highest calculated dose beyond a 100-meter buffer zone surrounding the facility.

For the water resources impact assessment requirement, USDOE Order 435.1 does not specify either dose or concentration limits for radionuclides in water. Therefore, there is some ambiguity in applying the requirement even though, as described previously, at SRS the performance measure is interpreted as requiring that concentrations of contaminants in groundwater should not exceed values specified in USEPA standards for public drinking water supplies as given in the Safe Drinking Water Act (40 CFR Part 141) [4].

The Primary Drinking Water Standards for radionuclides, promulgated on December 7, 2000, are used in this study [4]. The current 0.04 mSv/year (4 mrem/year) standard for beta and/or photon emitters in drinking water requires that MCLs be developed based on internal dosimetry data from National Bureau of Standards (NBS) Handbook 69 [5] and specified MCLs for ^{3}H and ^{90}Sr. A listing of the resulting MCLs is available in the Implementation Guidance for Radionuclides [6]. There are several radionuclides for which MCLs are not available in this listing. For these an MCL can be derived assuming a limit of 0.04 mSv/year (4 mrem/year) EDE and internal dosimetry based on ICRP Publication 30 [7]. Table I compares the MCL and the concentration equivalent to the 25 mrem/year all pathway dose.

Table I. Comparison of MCLs and allowable groundwater concentrations based on the 25 mrem per year performance objective for off-site individuals

Radionuclide	MCL, Bq/L (pCi/L)	Allowable concentration based on 25 mrem per year, Bq/L (pCi/L)
U-234	7,000 190,000	4.8 (130)
Np-237	0.56 (15)	0.33 (8.9)
Pu-238	0.56 (15)	0.33 (8.9)
Pu-239	0.56 (15)	3.0 (8.1)
Pu-240	0.56 (15)	3.0 (8.1)
Pu-241	11 (300)	15 (400)
Pu-242	0.56 (15)	0.31 (8.3)
Am-241	0.56 (15)	0.28 (7.6)

Air Analysis

The all-pathways performance objective of USDOE Order 435.1 includes all modes of exposure, including the air pathway, but excluding exposures to radon and short-lived progeny. In addition to this objective, calculated dose via the air pathway is not to exceed 0.10 mSv/year (10 mrem/year) total EDE, again excluding dose from radon and short-lived progeny. Again, the point of compliance is the point of highest calculated dose beyond a 100-meter buffer zone surrounding the closed facility.

Radon Emanation Analysis

Radon is addressed separately in a performance objective under USDOE Order 435.1, with separate applicable limits. In most cases, the limit for radon should be an average ground surface emanation rate of 0.74 Bq/m^2/s (20 pCi/m^2/s), which applies in this analysis.

Containment Requirements

A proposed on-site disposal system should be capable of meeting the performance standards for the disposal of transuranic waste set by the EPA [7]. The criteria states that for every 1 million curies of TRU waste disposed, only a total of one hundred curies is allowed to reach the accessible environment over a ten-thousand year period.

There objectives and requirements are summarized in Table II.

Table II. Performance Objectives, Assessment Requirements, Points of Compliance, Time of Compliance and Containment Requirements

Component	Performance Objective/Measure	Point of Compliance
All pathways	≤ 0.25 mSv (25 mrem) in a year, not including doses from radon and progeny	Point of highest projected dose or concentration beyond a 100-m buffer zone surrounding the closed facility
Air pathway	≤ 0.10 mSv (10 mrem) in a year, not including doses from radon and progeny	Point of highest projected dose or concentration beyond a 100-m buffer zone surrounding the closed facility
Radon	either (1) an average flux of ≤ 0.74 Bq/m2/s (20 pCi/m^2/s), or	Closed facility surface
	(2) an air concentration of ≤ 0.02 Bq/L(0.5 pCi/L)	Point of highest projected dose or concentration beyond a 100-m buffer zone surrounding the closed facility
Hypothetical inadvertent intruder	1 mSv (100 mrem) in a year from chronic exposure	Closed facility
	5 mSv (500 mrem) from a single event	Closed facility
Impact on water	The SRS interpretation is that	Point of highest projected dose

Table II. Performance Objectives, Assessment Requirements, Points of Compliance, Time of Compliance and Containment Requirements

Component	Performance Objective/Measure	Point of Compliance
resources	concentrations of radioactive contaminants should not exceed standards for public drinking water supplies established by the USEPA (40 CFR Part 141).	or concentration beyond a 100-m buffer zone surrounding the closed facility
Time of Assessment	10,000 years (40CFR191)	
Containment Requirement	Cumulative total of 3.7 TBq per 3.7E4 TBq (100 Ci per 1,000,000 Ci) of alpha-emitting transuranic isotopes with half-lives greater than 20 years over 10,000 years (40CFR191).	Accessible environment

KEY MODELING ASSUMPTIONS AND PARAMETERS

For protection of the public and the assessment of impacts to water resources, exposure pathways involving direct ingestion of groundwater and release of volatile radionuclides to the atmosphere are the pathways of dominant concern for this analysis. For the intruder analysis, there is no clear dominance of exposure pathways or scenarios, and doses vary greatly by radionuclide.

Assumptions of greatest importance to the projection of groundwater concentrations are those that affect the projection of release from the closed facility and subsequent transport to the point of compliance. Release from the waste forms is a strong function of the amount of water infiltrating the closed facility, the manner in which radionuclides are bound to the facility, physical/chemical sorption properties of individual radionuclides, solubility of the radionuclides, and the presence of engineered barriers to water flow. The amount of infiltrating water and hydraulic properties of the soil matrix are important to the estimation of the transport to the water table; however, over long periods of time, when steady-state conditions are approached, hydraulic properties become less important because the flow rate becomes controlled by the rate water infiltrates to the source zone. Ultimately, groundwater concentrations are a function of the rate radionuclides reach the water table, which are affected by the parameters listed above, and of the hydraulic properties of the aquifer matrix. Simulation of these important processes requires a number of generally simplifying assumptions. Those that most affect the projected groundwater concentrations are: 1) representation of the end state of the facility as concrete rubble; 2) no engineered cover system is assumed to be in place; 3) sorption is assumed to be adequately represented by non-site-specific sorption coefficients (K_ds) for many radionuclides and materials; and 4) all radionuclides are assumed to exist as surface contamination, and are available for transport.

Assumptions of greatest importance to the estimation of dose resulting from release of volatile radionuclides to air have to do with the rate at which volatile radionuclides are released to the atmosphere and the time at which the releases occur.

For estimation of dose to inadvertent intruders, exposure scenario definitions (assumptions) are perhaps most critical to the performance analysis. Probably the most important assumptions are: 1) the inadvertent intruder has no knowledge of prior activities at the site: 2) the intruder will build a home or drill a well at the location of the closed facility, rather than in uncontaminated areas; 3) the intruder excavates or drills at the earliest time possible relative to degradation estimates for the various materials; and 4) exhumed contaminated material is mixed with uncontaminated soil, and a garden is planted in the resulting mix. These important assumptions tend to maximize the calculated dose to the intruder, and thus provide a pessimistic evaluation of performance of the closed facility with respect to impacts on intruders.

MODELING WORK

The option that was analyzed in this study was to place all of the Space Grade waste to be left at SRS at the location known as Pad 1. Pad 1 currently contains waste received from Los Alamos and Mound in the 1970's (Figure 1) which presents particularly difficult challenges to characterize and repackage.

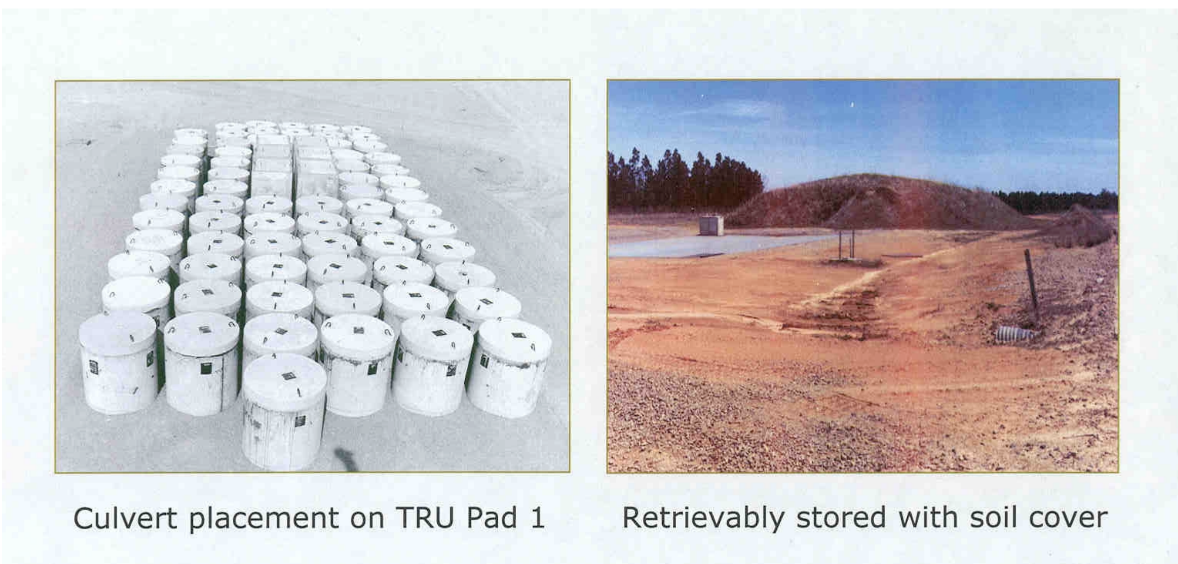

Figure 1. Views of TRU Pad 1 at the Savannah River Site

Pad 1 currently holds 266 m^3 of waste containing about 6.7E3 TBq (180,000 Ci). An additional 3.7E3 TBq (100,000 Ci) and 1178 m^3 is best suited to remain at SRS. The total waste volume is about 7 times greater than what is now on Pad 1, so the area in the model was taken to be 64 m x 64 ft, which is approximately 7 times the area of Pad 1, which measures 15 m x 38 m.

The PATHRAE performance assessment computer program [9] was used for this work to calculate groundwater and intruder results. Radionuclide inventories use were based on the isotopic distribution of Space Grade plutonium as shown in Table III. Am-241 was generated by the radioactive decay of Pu-241. The PATHRAE code does not treat the radioactive decay of Pu-238 into U-234. A hand calculation was done to show that 1.0E4 TBq (275,000 Ci) of Pu-238 will decay into slightly less than 3.7TBq (100 Ci) of U-234 over a one-thousand year period. An initial inventory of 3.7 TBq (100 Ci) of U-234 was therefore added to the PATHRAE input file. The peak groundwater concentration before 10,000 years, or the concentration at 10,000 if the peak concentration occurs after 10,000 years, was calculated.

Table III Isotopic Distribution and Radionuclide Inventory Used

Radionuclide	Half Life, years	Inventory, TBq (Ci)
U-234	244,000	3.7 (100)
Np-237	2,140,000	5.2E-3 (0.14)
Pu-238	87.7	1.0E04 (2.75E+05)
Pu-239	24,100	7.0 (1.92E+02)
Pu-240	6560	3.8 (1.03E+02)
Pu-241	14.3	2.3E+02 (6.13E+03)
Pu-242	373,000	5.8E-03 (1.56E-01)

The sorption coefficients (K_ds) assumed in the analyses of release from waste forms are listed in Table IV. Selection of K_ds was made according to the following rationale. Site-specific values of soil K_ds are considered most appropriate; when available, they were used. Next, the comprehensive listing of default values by Sheppard and Thibault [11] was consulted for K_ds in soil. The sandy soil K_d was selected for "soil" because this value tends to be lower than for other soil types, and thus is conservative (i.e., may overestimate radionuclide mobility) with respect to water resource impacts. For concrete, a listing of K_ds by Bradbury and Sarott [12] was consulted. For isotopes of Pu, a limit on solubility in a cementitious environment of concrete entombment, where the pH is expected to be well in excess of 7, is assumed to affect availability for transport. The solubility limit of 4.4×10^{-13} M was SRS Performance Assessments[10].

Table IV. Elemental Sorption Coefficients (K_ds) for Radionuclides of Interest

	K_d (ml/g)	
Nuclide	Soil[a]	Concrete[b]
Am	1900	5000
Np	5	5000
Pu	300[c]	5000
U	800[d]	2000

[a] Values are for sand from Sheppard and Thibault [11], unless otherwise noted.
[b] Values from Bradbury and Sarott [12].
[c] Site specific value from Cook [13].
[d] Site specific value from Serkiz[14].

Three intruder exposure pathways were analyzed, Food Grown on Site, Direct Gamma Exposure and Dust Inhalation. These are the components of the standard intruder scenarios (agricultural, resident and post-drilling) used in the SRS performance assessments [10]. The doses from each pathway were summed to give the total intruder dose per curie of residual inventory.

The key model inputs for the study are given in Table V.

Table V. Key Parameters for the PATHRAE Model

Property	Value	Source
Number of Isotopes	8	Goldston [1]
Length of Facility	64 m	See text
Width of Facility	64 m	See text
Density of Aquifer	1600 kg/m^3	EAV PA [10]
Longitudinal Dispersivity	1.0 m	Assumption
Transverse Dispersion	0	Assumption
Vertical Dispersion	0	Assumption
Residual Saturation	0.7	EAV PA [10]
Sat. Conductivity of Vertical Zone	100 m/y	EAV PA [10]
Cover Thickness	2.0 m	Closure Cap Report [15]
Waste Thickness	2.5 m	Culvert height
Waste Volume	1810 m^3	Goldston [1]
Distance to Well	100 m	DOE 435.1 [2]
Well Distance Off Centerline	0 m	Assumption
Density of Waste	1600 kg/m^3	EAV PA [11]
Waste Container Lifetime	0 yr	Assumption
Water Infiltration to Waste	0.4 m/yr	Site Average
Horizontal Velocity of Aquifer	50 m/yr	Collard and Hiergesell [16]
Porosity of Aquifer	0.44	EAV PA [11]
Distance from waste to Aquifer	17 m	Measurement 1/5/04
Well Screen Length	5 m	Standard SRS water table monitoring well
Surface Erosion Rate	3.0×10^{-6} m/yr	EAV PA [10]
Precipitation Runoff Rate	0.4 m/yr	EAV PA [10]
Porosity of Unsaturated Zone	0.44	EAV PA [10]
Bulk Density of Soil	1600 kg/m^3	EAV PA [10]
Leach Constant	Based on Concrete	Bradbury & Sarott [12]

RESULTS

The groundwater modeling results are given in Table VI. The only radionuclide in the inventory to reach to 100 meter well within 10,000 years is Np-237. The other radionuclides are below the cut-off value in the PATHRAE code of 3.7E-10 Bq/m^3 (10^{-20} Ci/m^3). These results are consistent with reported results from studies of disposal units with similar characteristics using two and three dimensional computer models [17]

Table VI. Groundwater Results Peak Concentration over 10,000 Years at 100 Meter Well

Radionuclide	Concentration Limit Bq/L (pCi/L)	Peak Concentration Bq/L (pCi/L)
U-234	7,000 190,000	--
Np-237	0.56 (15)	0.015 (0.4)
Pu-238	0.56 (15)	--
Pu-239	0.56 (15)	--
Pu-240	0.56 (15)	--
Pu-241	11 (300)	--
Pu-242	0.56 (15)	--
Am-241	0.56 (15)	--

The intruder results are shown in Table VII. The thickness of the SRS closure cover system precludes excavation into the actual waste and provides a great deal of shielding from direct gamma exposure.. Drilling is the only credible method to bring waste material to the surface. Since this involves a relatively small volume of contaminated material, the resulting doses are relatively low, and they decrease along with the decay of the major component in the waste, Pu-238.

Table VII. Intruder Results

Radionuclide	100 years Dose/year mSv (mrem)	300 years Dose/year mSv (mrem)	500 years Dose/year mSv (mrem)	1,000 years Dose/year mSv (mrem)	10,000 years Dose/year mSv (mrem)
U-234	2.3E-4 (2.3E-2)	2.8E-4 (2.8E-2)	2.8E-4 (2.8E-2)	2.8E-4 (2.8E-2)	1.8E-4 (1.8E-2)
Np-237	4.4E-6 (4.4E-4)	4.4E-6 (4.4E-4)	4.4E-6 (4.4E-4)	4.4E-6 (4.4E-4)	3.6E-6 (3.6E-4)
Pu-238	0.3 (30)	0.06 (6.0)	0.012 (1.2)	2.4E-4 (2.4E-2)	0 (0)
Pu-239	4.7E-4 (4.7E-2)	4.7E-4 (4.7E-2)	4.7E-4 (4.7E-2)	4.7E-4 (4.7E-2)	3.0E-4 (3.0E-2)
Pu-240	5.7E-5	5.6E-7 (5.6E-5)	5.5E-7 (5.5E-5)	5.1E-7 (5.1E-5)	1.6E-7 (1.6E-5)
Pu-241	3.5E-8 (3.5E-6)	0 (0)	0 (0)	0 (0)	0 (0)
Pu-242	3.7E-7 (3.7E-5)	3.7E-7 (3.7E-5)	3.7E-7 (3.7E-5)	3.7E-7 (3.7E-5)	3.6E-7 (3.6E-5)
Am-241	6.0E-5 (6.0E-3)	4.4E-5 (4.4E-3)	3.2E-5 (3.2E-3)	1.4E-5 (1.4E-3)	0 (0)
Total	0.3 (30)	0.062 (6.2)	0.013 (1.3)	1.0E-3 (0.1)	5E-4 (5E-2)

There are no radionuclides in the Space Grade waste that occur in the gaseous state, with the exception of radon produced by decay of Pu-238. Release and uptake of particulates can only occur as the result of intrusion, and have been dealt with in that analysis. Radon is discussed in the next paragraph.

The generation and release of radon is considered by using the results of a study on a disposal facility with characteristics similar to entombed waste on Pad 1[17]. Since the radon release limit is based on surface area, 0.74 Bq/m2/sec (20 pCi/m^2/sec), the result of the study is given in terms of a disposal limit expressed as curies per square meter of disposal unit. The result for Pu-238 is 140 TBq/m^2 (3,700 Ci/m^2). As shown in Table V, the area used for is 64 m by 64 m, or 4100 m^2. Therefore, a total 140 TBq/m^2 x 4,100 m^2, or 5.7E5 TBq (1.5E7 Ci) of Pu-238 could be disposed as still meet the radon emanation criteria. The estimated inventory of 1.0E4 TBq (275,000 Ci) is well below this limit so entombment of the Space Grade waste would not produce radon releases in excess of the limit.

To demonstrate the feasibility of meeting the containment requirements of 40 CFR 191, to results of the Intermediate Level Vault (ILV) will again be used. The regulation requires assurance that the disposal facility will release less than a cumulative total of 3.7 TBq (100 curies) of total plutonium for every 3.7E4 (1,000,000 curies) disposed. This is a cumulative fractional release rate of 1E2/1E6, or 1E-4. Table VIII gives the peak annual fractional release to the water table calculated for the ILV, this peak times 10,000 to calculate a cumulative release as if the peak rate were extended for 10,000 years. The cumulative release rate over 10,000 years is 1E-6, which is well within the requirements of 40CFR191[8].

Table VIII Cumulative Release over 10,000 Years

	TBq (Ci)	ILV peak fraction	TBq (Ci) over 10,000 yrs	
Np-237	5.2E-3 (0.14)	2.35E-6	1.2E-4 (3.22E-3)	
Pu-238	1.0E04 (2.75E+05)	5.61E-26	5.7E-18 (1.54E-16)	
Pu-239	7.0 (1.92E+02)	7.13E-8	5.1E-3 (1.37E-1)	
Pu-240	3.8 (1.03E+02)	1.44E-7	5.5E-3 (1.48E-1)	
Pu-242	5.8E-03 (1.56E-01)	5.90E-9	3.4E-7 (9.20E-6)	
Total	1.0E4 (2.80E+5)		1.1E-2 (2.88E-1)	
Cumulative Total / Initial Inventory				1.1E-06

CONCLUSIONS

Scoping calculations and the results of studies on existing disposal units at the Savannah River Site have been used to show that it should be possible to demonstrate that it is technically possible to dispose of a small volume of waste containing an appreciable quantity of Pu-238. This finding makes it possible to begin discussions with technical and regulatory bodies with the goal of safely disposing of this waste in a manner that is protective of both the SRS site workers, the economy and the environment.

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