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Consideration of In-Growth of Radionuclides for Facility Hazard Categorization

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Abstract

This paper addresses issues associated with the effects of daughter product in-growth on the hazard categorization of facilities in accordance with DOE-STD-1027-92, "Hazard Categorization and Accident Analysis Techniques for Compliance with DOE Order 5480.23, Nuclear Safety Analysis Reports."[1] There is a list of issues that occur when performing facility hazard categorizations at DOE facilities. The first issue is when radionuclides are concentrated outside of their natural decay schemes, and the resulting daughter products exceed the hazard category three threshold quantity values (HC3 TQVs) while their parents do not. The second issue is if a parent nuclide is evaluated for the inhalation pathway, and the daughter product is evaluated using a different pathway and methodology. The third issue is when the parent and daughter are evaluated using the same pathway for exposure, but the daughter is significantly more radiotoxic than the parent. Lastly, when the TQVs were derived for hazard categorization, the methodology used involved a 24 hour exposure period during which, for the sake of simplicity, no consideration was given to decay and the subsequent in-growth of daughter products. Facility hazard categorization is a snapshot in time and does not provide an accurate inventory for long term operations and/or storage.

Introduction

The purpose of this discussion is to identify the issues associated with the in-growth of daughter products in the hazard categorization of facilities. Four issues arise when performing hazard categorizations involving radioactive sources. The first issue is when radionuclides are concentrated outside of their natural decay schemes, and the resulting daughter products exceed the hazard category three threshold quantity values (HC3 TQVs) while their parents do not. An example would be U-238 and its daughter Pb-210. The second issue is if a parent nuclide is evaluated for the inhalation pathway, and the daughter product is evaluated using a different pathway and methodology. The third issue is when the parent and daughter are evaluated using the same pathway for exposure, but the daughter is significantly more radiotoxic than the parent. Lastly, when the TQVs were derived for hazard categorization, the methodology used involved a

24 hour exposure period during which, for the sake of simplicity, no consideration was given to decay and the subsequent in-growth of daughter products.

Methodology

Facility hazard categorizations are required for Department of Energy (DOE) facilities that contain radioactive material. The categorizations are to be conducted in accordance with the guidance and limits contained in DOE-STD-1027-92. Attachment 1 of STD-1027-92 Table A-1 lists the hazard category two and three TQV limits for category 2 and 3 facilities. The radionuclides listed in Table A-1 are reported in curies and their corresponding mass in grams. The list of nuclides in Table A-1 is not a complete list. For additional hazard category 3 nuclides not listed in Table A-1, users are directed to LA-12981-MS, "Table of DOE-STD-1027-92 Hazard Category 3 Threshold Quantities for the ICRP-30 List of 757 Radionuclides,"[2] table of DOE-STD-1027-92 Hazard Category 3 TQVs for the ICRP-30 List of 757 Radionuclides. The hazard category 2 limits may be found in LA-12846-MS, "Specific Activities and DOE-STD-1027-92 Hazard Category 2 Thresholds."[3]

The HC3 threshold limits are established based on the Environmental Protection Agency's (EPA) assumptions[4] that result in the maximally exposed individual receiving a limit of 10 rem total effective dose equivalent (TEDE) at 30 meters over a 24 hour period. EPA 400-R-92-001[5] establishes the 10 rem dose limit; however, it should be noted that the 10 rem limit is established for external and inhalation exposure pathways. The ingestion pathway is not included. Manual EPA 400-R-92-001 was developed to assist public officials in planning for emergency response to nuclear incidences. The 10 rem dose limit is based on the early phases of accidents for the purposes of protecting people and property. No consideration was given for long-term storage of radioactive material and hazard classifications. The dose to an individual is evaluated for the parent nuclide only for the period of exposure with no consideration given for radioactive decay and the resulting daughter products.

The TQVs listed in Table A-1 of DOE-STD-1027-92, are established based on four exposure pathways and release values that EPA considered. The pathways include water (w), food (f), direct exposure (d) and inhalation (i). The HC3 TQVs are calculated on the amount of activity of a nuclide that would produce 10 rem at 30 meters.

A review of EPA 400-R-92-001 is required in order to fully understand the rational for the established exposure limits. Typically the period of exposure in the early phase of a release is considered short (approximately four days as defined in EPA 400-R-92-001); however, because this time period for exposure is considered quite short, it is not technically reasonable to include daughter products, which may take years to in-grow. This addresses the immediate doses and has no bearing on long term storage of nuclides with decay chains.

In the guidance provided in DOE-STD-1027-92, the models used assume that persons are exposed based on a 24 hour exposure for inhalation and direct exposure, but that persons are exposed for longer periods through the ingestion pathway. To address the ingestion pathway, the mitigating actions of relocation and the establishment of Protective Action Guidelines (PAGs) were used. It was assumed that exposed individuals would be protected from subsequent doses

based on relocation and radionuclide depositions that were lower in activity than the established PAGs.

Radioactive Decay

Radioactivity is the spontaneous emission of radiation, generally alpha, or beta particles, often accompanied by gamma rays from the nucleus of an unstable atom. This can also include the emissions of neutrons and spontaneous fission. Radioactive decay has three types of equilibrium. The types of equilibrium have an effect on how radioactive materials accumulate or transform into different elements.

Types of Equilibrium

There are three types of equilibrium. The first is Secular Equilibrium. This type of relation between parent and daughter activity occurs when the half-life of the parent nuclide is infinitely longer than that of the daughter nuclide. The following equation will produce the amount of activity of the daughter at time (t).

For $\lambda_1 << \lambda_2$

$$A_2 = A_1^0\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)$$

The next is Transient Equilibrium. In this case, the half-life of the parent nuclide is still larger than that of the daughter nuclide, but not infinitely.

For $\lambda_1 < \lambda_2$

$$A_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1^0 e^{-\lambda_1 t}$$

Finally, there is the case of No Equilibrium. Here the half-life of the daughter nuclide is larger than that of the parent. As a result and after a sufficiently long time, only the daughter activity will be left because the parent is decaying at a faster rate.

For $\lambda_1 > \lambda_2$

$$A_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1^0 e^{-\lambda_2 t}$$

Below are two examples of the types of decays associated with operations and long-term storage. The examples will show secular equilibrium and the case of no equilibrium.

In-growth of Pb-210 from a 500 mCi Ra-226 source (Secular Eq.) for 41.5 years:

$$A_2 = 500 \text{ mCi}(e^{-(.693/1599y*41.5y)} - e^{-(.693/21y*41.5y)})$$
$$A_2 = 500 \text{ mCi}(9.82E\text{-}1 - 2.54E\text{-}1)$$
$$A_2 = 364 \text{ mCi}$$

The in-growth of Po-210 was shown for informational purposes only. The radiotoxicity of Po-210 is extreme and, therefore, should be of concern and interest. Figure 1 shows the Pb-210 in-growth from Ra-226, as shown in the equation above.

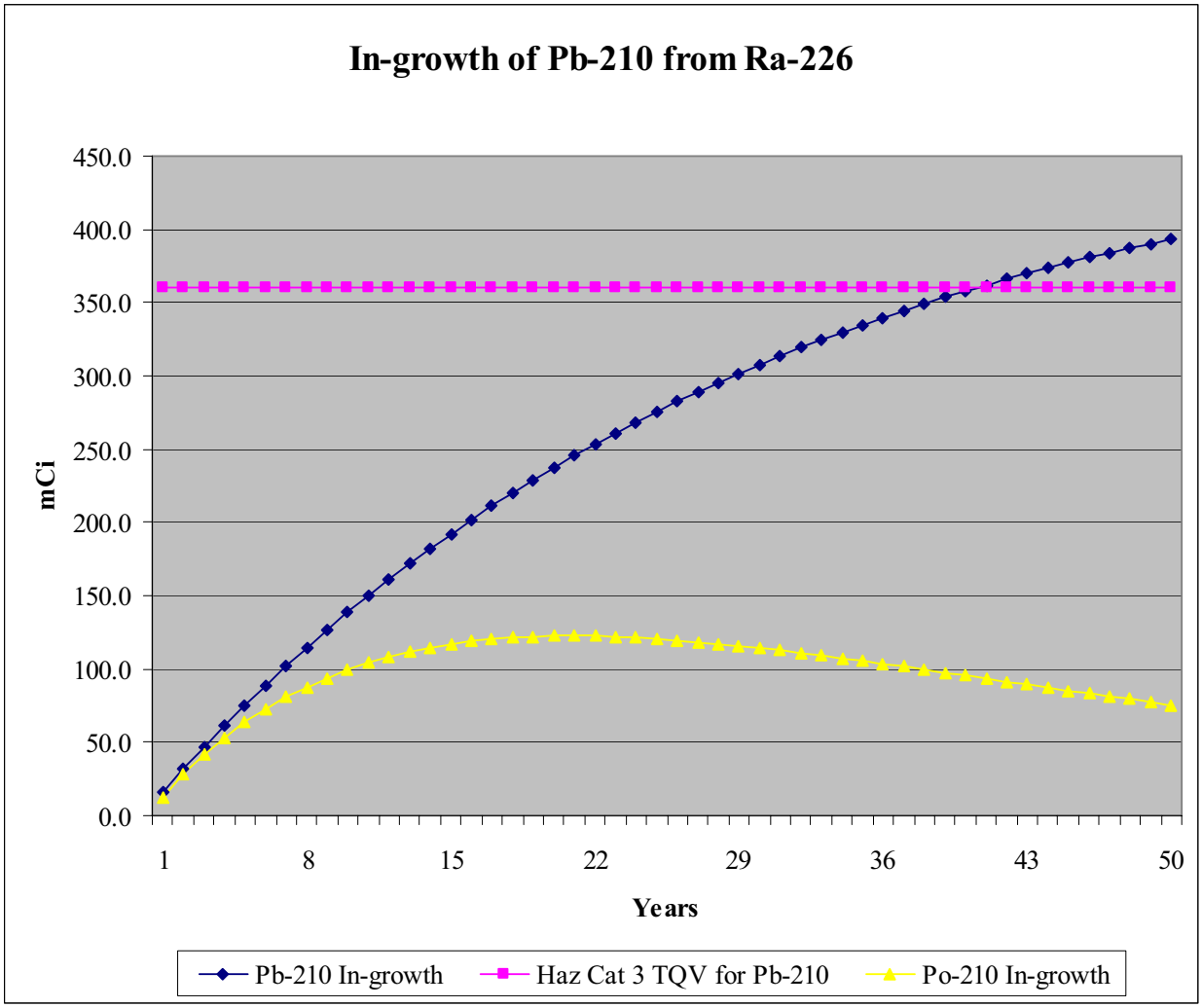

Figure 1. Pb-210 In-growth from Ra-226.

In-growth of Am-241 from Pu-241 (No Equilibrium Eq.) for 14.4 years:

$$A_2 = [(.693/432)/(.693/432)-(.693/14.4)](32\ Ci)\ (e^{-(.693/14.4y*14.4y)})$$

$$A_2 = [1.604E\text{-}3/\text{-}4.652E\text{-}2](32\ Ci)(5.0E\text{-}7 - 9.77E\text{-}1)$$

$$A_2 = (-3.448E\text{-}2)(32\ Ci)(-4.7704E\text{-}1)$$

$$A_2 = 0.526\ Ci \text{ or } 526\ mCi$$

The graph in Figure 2 is a log normal depiction of no equilibrium decay and in-growth over a 50 year period. There are many legacy sources of material that have had nearly 50 years of decay. The decay time in the equation above was 14.4 years to show the time at which the Am-241 grows into a HC3 limit from Pu-241.

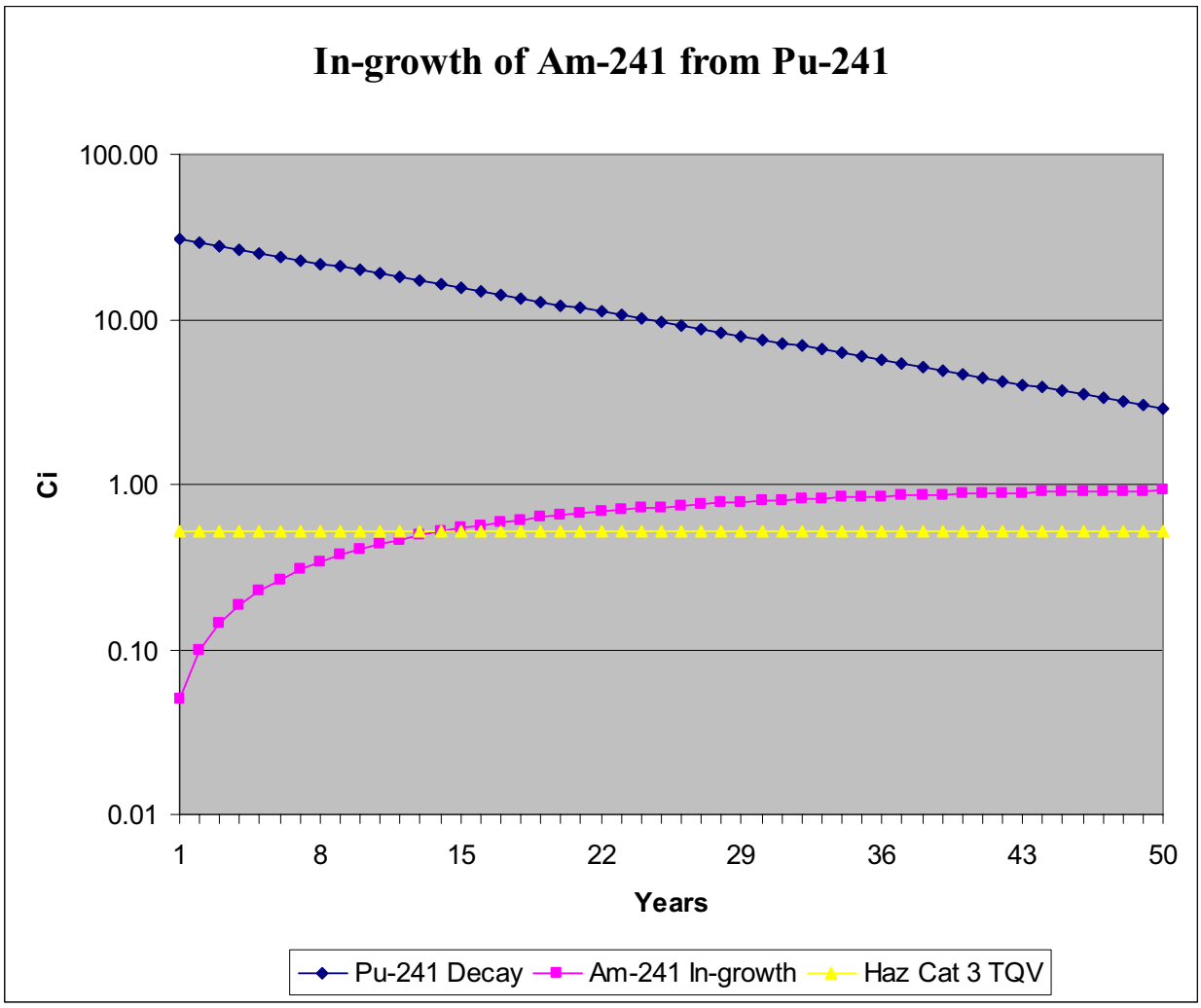

Figure 2. Am-241 In-growth from Pu-241.

The following sections will address the four issues associated with the current methodology of facility hazard categorizations.

Daughter Concentration Outside of Natural Decay Chains

The first issue is when radionuclides are concentrated outside of their natural decay schemes, and the resulting daughter products exceed the HC3 TQVs while their parents do not.

This occurs when the natural decay chains of Thorium, Neptunium, Uranium, and Actinium are altered and particular daughters, Ra-226 for example, are artificially concentrated. The time required to naturally in-grow Pb-210 to a HC3 TQV from the HC3 TQV for U-238 is approximately 100,000 years. Since this time frame is completely unrealistic for hazard categorizations, the issue of Pb-210 decaying beyond its HC3 TQV is not credible under natural decay circumstances. However, if the decay chain is artificially enhanced by chemically concentrating Ra-226, then in approximately 41 years the in-growth of Pb-210 will exceed its HC3 TQV, and the source will have become a HC3 "facility." In essence, the artificial concentration has effectively increased the initial amount of material present. What would normally require tons of U-238 to produce the HC3 TQV of Pb-210 now only requires a few hundred milligrams of Ra-226.

Not all in-growth issues deal with altering the natural decay chains. In the case of fission products, where the parent nuclide decays producing a radioactive daughter product, a condition of no-equilibrium could exist in which the daughter product has a significantly longer half life than its parent. The decay of Mo-99 to Tc-99 or Xe-135 to Cs-135 are some examples.

Different Exposure Pathways

The second issue is if a parent nuclide is evaluated for the inhalation pathway, and the daughter product is evaluated using a different pathway and methodology. This occurs when in a given decay chain, the parent nuclide (U-228) is evaluated as an inhalation risk while the daughter product of concern (Pb-210) is evaluated as an ingestion risk. The TQVs for the parent and daughter are not consistent as they pertain to the exposure pathways. The problem is further compounded when the natural decay chains of radionuclides are artificially concentrated as previously discussed. If the parent nuclide is in secular equilibrium with its daughters, and the exposure pathway is consistent between the two, then it can be assumed that the TQV values would be correctly based on the parent nuclide value. However, this is not the case with DOE-STD-1027-92, LA-12981-MS, or EPA's "Technical Background Document to Support Final Rulemaking Pursuant to Section 102 of the Comprehensive Environmental Response, Compensation, and Liability Act: Radionuclides."

The problem caused here, as a result of inconsistent pathways between the parent and daughter nuclides, is that it is not clear in the methodology presented in DOE-STD-1027-94 that separate biokinetics for systemic activity have been applied to the parent radionuclide and its daughter products. In *ICRP 72*[6] the usual assumption is that daughter products produced in vivo adopts the absorption parameters of their parent, if they are produced in the respiratory or gastrointestinal tract, and the biokinetics of their parent, if they are produced after absorption to blood. In all cases, the dose coefficients corresponding to the intakes of the parent radionuclide include contributions from the parents and their daughters. However, for inventory purposes, these factors should not influence how radioactive material should be accounted for in facilities. In

short, the issue is that physical decay schemes of various radionuclides and their associated intake and systemic pathways are not logically tied to the TQVs used in the guidance documents. DOE-STD-1027-94 does not take into consideration that Pb-210 decayed from U-238 must have the same pathway of exposure. Instead, the standard uses the EPA pathway that produces the lowest TQV value irrespective of the parent radionuclide intake pathway. Under DOE-STD-3009-94,[7] the ingestion pathway scenario is excluded as a pathway requiring analysis for exposure, yet some TQV limits listed in DOE-STD-1027-94 are based on ingestion.

Same Exposure Pathways – Different Radiotoxicity

The third issue is when the parent and daughter are evaluated using the same pathway for exposure, but the daughter is significantly more radiotoxic than the parent. An example case would be Pu-241 decaying into Am-241. Both Pu-241 and Am-241 are evaluated as an inhalation pathway exposure. Also in this scenario, we have what is called "no equilibrium." In this case the daughter has a significantly longer half life than the parent, and the daughter grows at essentially the rate of decay of the parent until complete decay occurs. The issue here is that over a relatively short amount of time, given that you start with slightly less than HC3 TQV of Pu-241 (approximately 32 Ci) and store it for 14.5 years (one half life of Pu-241), the source will have grown into a HC3 facility. The result is a potentially and significantly higher dose to the receptor following an accident or inadvertent release. The rem/Ci for Pu-241 is 1.0E7 while Am-241 is 5.2E8[3]. The increased dose consequence from Am-241 as a result of its higher radiotoxicity is nearly 52 times that of Pu-241. The difference is significant enough to warrant further evaluation of the current DOE-STD-1027-92 methodology.

The issue of radiotoxicity not being considered is again confirmed by the HC3 TQVs listed in DOE-STD-1027-94 for Pu-238, Pu-239, and Am-241. Radiotoxicity is the potential of an isotope to cause damage to living tissue by absorption of energy from the disintegration of the radioactive material introduced into the body. The HC3 TQV limit for Pu-238 is higher than that of Pu-239, and the TQV for Pu-239 is equal to the TQV for Am-241. Both Pu-238 and Am-241 are significantly more radiotoxic than Pu-239. This is because of the higher specific activities and modes of decay.

Methodology Used in EPA Study

Lastly, when the TQVs were derived for hazard categorization, a key portion of the methodology the EPA study used involved a 24 hour exposure period during which, for the sake of simplicity, no consideration was given to decay and the subsequent in-growth of daughter products.

The "Technical Background Document to Support Final Rulemaking Pursuant to Section 102 of the Comprehensive Environmental Response, Compensation, and Liability Act: Radionuclides," study was based on a short term accident release scenario that considered four pathways for exposure previously mentioned: water (w), food (f), direct exposure (d) and inhalation (i). The pathway methodology was not consistent between parent-daughter decay schemes. Part of this could be attributed to the fact that in each pathway of exposure, the EPA study clearly stated that decay was not accounted for due to the short release and exposure time to the dose receptor. In each case long term decay and in-growth were not considered. While this methodology is

sufficient for accident scenarios and emergency events, it is not adequate for stable long term operations and storage of radioactive material.

Many DOE facilities have radioactive material that is either stored without a current decayed inventory, or they have legacy waste or artifacts, such as source vials or radium needles, that have decayed into HC3 facilities. Often the current facility operations management and staff are unaware that the material has changed their categorization. In the case where a facility or operations does address legacy material or has concerns of daughter product in-growth, the current methodology allows for inventory decay; however, this is only a "snap-shot" in time of the current inventory. Within a few months or years, this inventory is no longer an accurate measure of material present.

Additionally, no consideration was given for artificial concentration of radionuclides outside of their natural decay chains. As stated previously, this makes a significant impact on the time and amount of material required to push a non-nuclear facility over the HC3 threshold limits simply by storing legacy material or waste.

Finally, the methodology does not address decay daughters as being more radiotoxic than the parent nuclides. This is the case for a good number of decay chains. In some instances, it's the first daughter product that rapidly in-grows and becomes the nuclide of concern (Pu-241 to Am-241); others may take decades before the daughter product becomes a radiotoxicity concern (Ra-226 to Pb-210). In both scenarios, time is the controlling factor. Since the EPA study was only concerned with accidental releases up to four days, there is no formal mechanism to deal with in-growth that lasts years and decades.

Conclusion

The four presented issues associated with facility hazard categorizations make it nearly impossible to perform an accurate hazard categorization utilizing the current DOE-STD-1027-92 methodology. At some point, the affected facilities will need to perform inventory decays to ensure that they remain below the HC3 TQVs. A forward looking future decay evaluation would also be of benefit to help the facility project what their inventory will be and to make proactive arrangements to either dispose of legacy sources and waste or to prepare for re-categorization.

Under the current system, to accurately categorize a facility, a current decay of the facility will need to be performed. Additionally, the ICRP 30 Dose Conversion Factors (DCFs) should be updated to the current ICRP 68/72 in order to consider the changes in the DCFs and the affects that individual nuclide radiotoxicity has on them. Also, since the ingestion pathway has been left out of DOE-STD-3009 guidance and methodology, the direct exposure and inhalation pathways would become uniform for both the parent and daughter nuclides regardless of the decay mechanism.

There are commercially available software packages that track radionuclide inventories that have built-in decay functions based upon the receipt or assay dates of the material inventoried. These provide a real time running inventory of the material stored in the facility. This approach may be

more effective at addressing this issue than a time consuming and costly re-evaluation of the EPA's and DOE's hazard categorization methodology.

References

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2. LA-12981-MS, "Table of DOE-STD-1027-92 Hazard Category 3 Threshold quantities for the ICRP-30 List of 757 Radionuclides," August 1995.

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