

Appendix III. Occupational and Public Risks Associated with In-Situ Leaching

Introduction

EPA's Science Advisory Board (SAB) recommended that EPA present information on in situ leaching (ISL) mining operations and uranium mill operations to provide a more complete picture of uranium production. While this report focuses on the impacts associated with conventional surface and underground uranium mines, it provides limited background materials, in this and other appendices, on risks associated with uranium milling and ISL operations and wastes generated by those processes, even though they may not be considered TENORM by virtue of their regulation by the NRC and its Agreement States under the Atomic Energy Act and its amendments.

The NRC stated its intent in July 2007 (NRC 2007b) to develop a Generic Environmental Impact Statement (GEIS) on uranium milling which would provide more detailed information on the ISL process and impacts, and may include more recent information on the impacts. The reader is referred to that document when made available to the public in the future for additional background information and associated risk assessment.

This appendix summarizes information on environmental and health-related aspects of ISL operations. The primary sources used for this review are, *Technologically Enhanced Naturally Occurring Radioactive Materials from Uranium Mining. Volume 1: Mining and Reclamation Background* by U.S. EPA (2006), *An Environmental Overview of Unconventional Extraction of Uranium* by Marlowe (1984) and *A Baseline Risk-Informed Performance Approach for In Situ Leach Uranium Extraction Licensees* by Mackin et al. (2001).

Background

In situ leaching is an extraction process that is regulated by the NRC or its Agreement States; the waste materials and fluids are considered byproduct material (see Appendix VI of U.S. EPA 2006). However, these operations are subject to the requirements of UMTRCA implemented by EPA regulations 40 CFR Part 192 and subsequently by NRC's regulations in 10 CFR Part 40. While ISL operation wells are subject to permitting under EPA's Underground Injection Control (UIC) program (U.S. EPA 2006, Appendix VI), the restoration requirements are considered complementary to those of UMTRCA, which take precedence. ISL operations, also known within the uranium industry as "in situ recovery," or ISR, are discussed here to provide a more complete representation of the impacts from uranium production. The following several points assume that ISL is economic.

ISL is used when environmental conditions are favorable, such as the following:

- The ore body is located in a saturated, permeable formation that is confined hydrologically both above and below
- The ore body is chemically amenable to the ISL process
- The uranium is present in multiple-layered roll fronts that may be offset by faulting

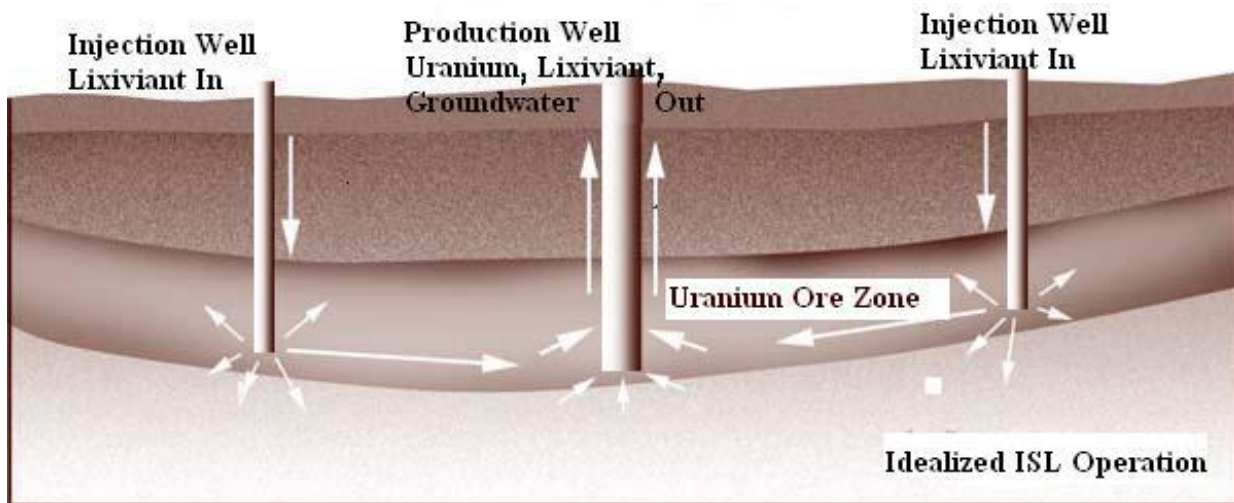
- The ore grade is low, and the ore body is too thin to mine by conventional means, however, if the ore body is too thin then even ISL may not be geologically possible

In this method of extraction, uranium ores are leached underground by the introduction of a solvent solution, called a lixiviant, through injection wells drilled into the ore body. The process does not require the physical extraction of ore from the ground, which makes it a much more economical option in many cases. Lixivants for uranium mining commonly consist of water containing added oxygen and carbon dioxide or sodium bicarbonate, which mobilize uranium. Other ISL facilities, especially in Eastern Europe, employ an acid-based lixiviant, though this method is rarely, if ever, utilized in the United States. The lixiviant is injected, passes through the ore body, and mobilizes the uranium. The uranium-bearing solution is pumped to the surface from production wells.

The pregnant leach solution is processed to extract the uranium, usually by ion exchange or by solvent extraction. The ion exchange process employs a resin that, once fully saturated with uranium, is flushed with a highly concentrated salt (e.g., sodium chloride) solution. This reverses the exchange process and releases uranium into the solution. The uranium solution is then sent to another process for concentration, precipitation, and drying as yellowcake. The solvent extraction process relies on unmixable properties between the pregnant leach solution and (uranium) solute. Normally, the solvents are organic compounds that can combine with either cationic or anionic solutes. For example, anionic solutions include amine chains and ammonium compounds, and cationic solutions are phosphoric acid-based. Figure AIII-1 shows a simplified version of the ISL process.

Figure AIII-1. Illustration of ISL Process

This figure shows a simplified version of how ISL solution mining works. Lixiviant is injected into the ground through wells on the left and far right, the fluid flows underground, dissolving uranium and carrying it in solution until it reaches a production well in the center. The fluid carrying dissolved uranium is returned to the surface from the production well, and piped to a production facility for refinement into yellowcake.



Source: Modified after ANAWA : <http://www.anawa.org.au/mining/isl-diagram.html>

When the ISL process is completed, the ore body and aquifer are placed in a restoration phase, as required by mine permits, NRC and Agreement State regulatory programs. Typically, the aquifer

must be restored to background or EPA drinking water maximum contaminant limit levels where possible or practical, or to Alternate Concentration Limits (ACLs) in terms of the presence of metals, organics, pH level, and radioactivity, approved by the NRC and its Agreement States. Therefore, in some cases, restoring it to the pre-operation level does not necessarily make it potable. EPA groundwater protection standards issues under authority of UMTRCA are required to be followed by ISL licensees of the NRC and its Agreement States. In addition to those requirements, ISL operators must apply for UIC permits from EPA. Through the UIC aquifer exemption process, EPA and its Delegated States determine if an aquifer or part of an aquifer is exempt from protection as an underground source of drinking water during the mining process. Approval of this exemption is necessary before a UIC permit may be issued for ISL mineral extraction wells. EPA requires, however, that non-exempted groundwater sources be protected from contamination. The UIC requirements are complementary to UMTRCA requirements and do not override the necessity for a licensee to take measures to restore well field groundwater.

According to Commission Order CLI-00-22, in situ leach mining (ISL) produces two categories of waste; (1) gaseous emissions and airborne particulates resulting from drying of yellowcake (which could take place at an off-site facility), and (2) liquid waste associated with operations including well field processing and aquifer restoration (Dicus et al. 1999). A variety of methods exist to address liquid waste disposal and storage at ISL facilities, including the use of evaporation ponds, deep-well injection, land application, and surface discharge under a National Pollution Elimination System (NPDES) permit.

Figure AIII-2. Picture of an in situ leach field

Unlike a conventional mine, ISL operations produce minimal solid waste. This picture from the Wyoming Association web site shows an ISL well field.



Source: <http://www.wma-minelife.com/uranium/insitu/insitufr.htm>

Potential Environmental and Health Issues

While the primary environmental concern from ISL operations may be related to groundwater, Mackin et al. (2001) identify four primary risks from ISL operations in three categories :

- (i) Surface environment chemical hazards
- (ii) Surface environment radiological hazards
- (iii) Groundwater chemical and radiological contamination hazards

The main risks to the worker are from the surface chemical and radiological hazards associated with various types of accidents at the site. Conversely, the risks to the general public pertain to the contamination of drinking water sources. Therefore, site-specific accidents would not affect the public unless a large prolonged release of hazardous chemicals and/or radionuclides were allowed to contaminate the local water supplies. In addition to hazards during ISL operation, site rehabilitation presents environmental and health concerns. Each of these issues is discussed in the sections to follow.

(i) Surface Environment Chemical Hazards (Mackin et al. 2001):

Twelve chemicals are commonly used in ISL operations and could pose hazards to ISL workers, but are unlikely to affect the general public. These chemicals, along with their intended purpose at ISL facilities, are summarized below in Table AIII-1. Potential hazardous situations involving each of these chemicals are discussed in the paragraphs following the table.

Table AIII-1: Typical Chemicals Found at ISL Operations

Chemical	Formula	Purpose at ISL Operations
Anhydrous Ammonia	NH ₃	pH adjustment during uranium precipitation phase
Sulfuric Acid	H ₂ SO ₄	Used to separate the uranium from the carbonate complex pumped from below the surface
Oxygen (gaseous and liquid)	O ₂	Oxidant added to lixiviant used for extraction of uranium forming UO ₃
Hydrogen Peroxide	H ₂ O ₂	Oxidant used during the precipitation phase of uranium
Sodium Hydroxide	NaOH	pH adjustment during radium removal phase
Barium Chloride	BaCl ₂	Used as a precipitant for radium during restoration and wastewater treatment
Carbon Dioxide	CO ₂	Carbonate used to keep oxidized uranium in solution, also used for pH adjustment of lixiviant
Hydrochloric Acid	HCl	pH adjustment during radium precipitation phase
Sodium Carbonate	Na ₂ CO ₃	Carbonate used to keep oxidized uranium in solution, also used in the regeneration/recycling resin
Sodium Chloride	NaCl	Used to regenerate/recycle the resin for further use in uranium extraction
Hydrogen Sulfide	H ₂ S	Used in groundwater restoration to decrease the solubility of various heavy metals
Sodium Sulfide	Na ₂ S	Used in groundwater restoration to decrease the solubility of various heavy metals

The main hazard posed by ammonia would be if a pipe were to break inside the processing plant. The liquid ammonia, assumed to be under high pressure, would likely have a significant spray in such an event and would pose a risk to the skin and eyes of any localized worker. In addition, as the ammonia quickly evaporates, an inhalation hazard would exist that would be exacerbated by poor ventilation. The possibility also exists for a leak in the primary holding tank or associated piping which transfers the ammonia from outside the plant to its application site.

Similar to ammonia, a break in the pipes used to transfer sulfuric acid, sodium hydroxide, and hydrochloric acid inside the plant would pose a hazard, as it is highly corrosive to the skin. Sulfuric acid and sodium hydroxide would not pose a significant inhalation hazard unless the ventilation systems in the plant were not in operation or if a worker encountered a “spray” caused by smaller leaks in the piping system. A hydrochloric acid leak could lead to a vapor inhalation hazard, especially in confined spaces. These chemicals are also highly reactive with one another and so multiple localized failures, as might be the case with fire or explosions, would cause an even greater hazard.

Oxygen gas poses a significant hazard because of its combustible properties. Similarly, hydrogen sulfide and sodium sulfide also pose risks because of their flammable properties, in addition to an inhalation as well as an eye/skin irritation hazard.

Hydrogen peroxide’s main risk pertains to the degradation of the chemical into hydrogen and oxygen gas which can be caused by mechanical shock, incompatible materials, light, ignition sources, excess heat, strong oxidants, rust, dust, or a pH greater than 4.0. Also, if the chemical is contained within an especially rigid tank, the casual degradation of the H_2O_2 into water and oxygen gas would cause expansion which could rupture the holding tank. A pipe failure event within the processing plant holds similar risks to that of ammonia and sulfuric acid.

Barium chloride is only considered a hazard if it is inhaled or ingested. Since the chemical is in solution form at an ISL plant, this would only become significant if the worker encountered a “spray” from a leaky piping system. Carbon dioxide from a leak can pose a risk of asphyxiation if it occurs in a sufficiently confined space. This can be avoided if a self-contained breathing apparatus were used when entering confined spaces where the displacement of oxygen with carbon dioxide is possible.

Sodium chloride and sodium carbonate both are very irritating to the eyes and the skin. In addition, sodium carbonate can pose an inhalation hazard when it is in its salt stage (dust inhalation) or from small leaks which form a spray of the sodium chloride/carbonate solution. Sodium carbonate also reacts readily with HCl and H_2SO_4 .

(ii) Radiological risks

Thickener Tank Failure

The thickener tank stores wet yellowcake slurry before it is sent to a precipitation operation and dried into U_3O_8 yellowcake. Thickener tank failure can pose an inhalation risk to workers if

spills are not cleaned up before the contaminants are allowed to dry. This accident scenario would not be a significant risk to off-site residents.

The thickener tank itself does not pose any external exposure risk, as most of the uranium progeny have been removed and the alpha component would be significantly attenuated by the slurry. Annual external exposures have been calculated to be 120 mrem for the limiting case of a worker standing directly next to the thickener tank for an entire 2,000 hour work year (Mackin et al. 2001).

If the yellowcake slurry is allowed to dry after a spill incident, it would pose a significant risk of uranium inhalation. Conservative treatments indicate that the dose to the public from a massive spill and subsequent airborne contamination event remain below the radiation dose limits established by 10 CFR 20 for members of the general public, however, the intake to an unprotected worker has the potential to exceed the 5 rem annual occupational limits (Mackin et al. 2001).

Yellowcake Dryer Accident

As discussed above, the dried yellowcake which consists of quantities of U_3O_8 , can pose a significant inhalation hazard to the onsite worker when it is allowed to dry. Failure of the dryer cake systems can stem from a number of accidents, including fire/explosion (worst case), spill over of dryer contents due to a faulty discharge valve, failure of offgas treatment systems causing the gases to release into the dryer area, and damage to the facility via natural disasters. It is important to note that the failure of the yellowcake dryer systems due to natural disasters is effectively bounded by the fire and explosion scenario. Exposures from a yellowcake dryer accident would presumably be of similar magnitude to that of the thickener tank scenario. (Mackin et al. 2001)

Exposure to Pregnant Lixiviant or Loaded Resin

Pregnant lixiviant and loaded uranium resin may pose a radiological hazard as an external exposure source, and present the possibility of inhaling elevated levels of radon-222. The most likely indoor exposure incident would occur if the pregnant lixiviant/resin were released due to a pipe or valve failure during the ion-exchange process, at which point the solution would drain from the ion-exchange column and the radon gas would be released to the air.

In addition to the inhalation hazard from radon, the pregnant lixiviant contains some other radioisotopes of interest that may also cause a significant exposure. These radioisotopes are shown in Table AIII-2, along with typical activity concentrations (Mackin et al. 2001).

Table AIII-2: Radionuclides with Typical Activity Concentrations* found in Pregnant Lixiviant/Loaded Resin

Radionuclide	Activity Concentration (pCi/L)
²²² Rn	8.0×10^5
²²⁶ Ra	3.4×10^3
Natural Uranium (²³⁴ U, ²³⁵ U, ²³⁸ U)	1.7×10^5
²¹⁸ Po	3.4×10^3
²¹⁴ Bi	3.4×10^3
²¹⁴ Po	3.4×10^3

*Progeny assumed to be in equilibrium

Conservative treatments of a possible spill incident have been modeled to show that a maximum annual exposure would be 27 mrem to a subject standing on a spill of infinite area and depth; with the consideration of loaded resin, this value becomes much lower. Since such a spilling event would likely be cleaned up expeditiously, such an exposure is not likely and is also well within the limits established in 20 CFR 20 for the general public, as well as the site worker (Mackin et al. 2001).

Exposures from the failure of near surface piping and subsequent runoff into containment ponds can also pose a possible hazard to workers. It is likely that the inhalation component in this scenario is negligible due to the dilution of the radon gas releases by ambient air; however, the external component would be similar to the indoor scenario previously described. See the next section for further discussion of outdoor near-surface releases.

(iii) Groundwater Contamination Risks

Due to the nature of the ISL process (specifically the low pH and oxidation mechanisms), other heavy metals and hazardous elements are also mobilized from the ore and can contaminate the groundwater. These elements include the radioisotopes and progeny of uranium, thorium, radium, and radon, as well as the non-radioactive elements such as arsenic, vanadium, zinc, selenium, and molybdenum (for a more complete list see Table AIII-3). Because these elements become mobilized in the target aquifer by the process of uranium extraction, it is possible for them to migrate out of the ore body into surrounding aquifers which might feed the local water supply. The underground propagation of this contamination into surrounding water is known as an excursion. Horizontal excursions refer to the lateral movement of the water, while vertical excursions indicate contamination of aquifers above and below the target ore body.

In order to detect and minimize this process, ISL facilities drill monitoring wells outside of the main well-field at a distance sufficient to detect any excursion events, while minimizing any erroneous indicators as a result of normal fluctuations. Horizontal excursions are more common than vertical excursions, but do not often become problematic to the outside water supply as long as they are detected and cleaned up within a reasonable time period. Vertical excursions are generally a result of well casing failure (ineffective cementing of well casing), improper sealing of abandoned exploration wells, or discontinuous or permeable natural confinement layers. Similar to horizontal excursions, vertical excursions do not pose a significant threat unless allowed to persist over significant periods of time—this is unlikely if geological properties of the

confinement layers are accurately characterized (to prevent downward vertical excursions), and the well shafts are effectively cased and proper monitoring well stations have been established. Along with well monitoring techniques, general practice at ISL facilities is to limit the injection of lixiviant so that it is always slightly less in volume than the product solution that is pumped out of the aquifer. This operating policy, known as “process bleed,” would effectively preclude excursions caused by overloading the aquifer, and the subsequent expansion and redistribution of the water.

In the United States, excursions have been frequently detected by the monitoring wells located around the well field. One of the more infamous and environmentally problematic ISL operations was located at Irigary, Wyoming. This facility was plagued by persistent environmental excursions which began in mid-March of 1979, and were not brought under control until early July of that same year. The Wyoming Department of Environmental Quality reported that these excursions were a result of the neglect of injection pressure monitoring as well as testing the integrity of the well casings (Mudd 1998). Another significant example is the Bruni mine in Texas, where there was a continued problem with both leachate spills and excursions. The Texas Department of Water Resources reported that at one point during the operational period the Bruni mine was cited for fourteen excursion incidents, while only five had originally been reported (Mudd 1998). Despite these scenarios, no significant contamination of local water supplies has been reported as a result of these excursions.

A separate technical analysis of excursions in early Texas and Wyoming ISL facilities (Staub et al. 1986) provides a more detailed review of the horizontal and vertical groundwater impacts from those facilities. In that study, concern was greatest in instances where a near surface aquifer had been polluted by vertical pollution from surface evaporation ponds, unplugged exploration boreholes, and improperly constructed injection/production wells. The associated impacts were greatest due to use of such water bodies by livestock or humans, and the difficulty and expense of aquifer restoration.

In addition to the below ground excursion incidents, the groundwater can become contaminated due to failure of the near-surface or surface piping systems which transfer the pregnant lixiviant from the well field area to the processing facility. Typical activity concentrations for the radionuclides present in lixiviant are given in Table AIII-2. In addition, Table AIII-3 displays the maximum measured concentrations of non-radioactive contaminants in pregnant lixiviant based on a survey of available licensing documents (Mackin et al. 2001). Once the pregnant lixiviant solution is released, there are three potential outcomes for the contamination; runoff into surface bodies of water, absorption into the soil and possible subsequent infiltrations of the groundwater, or runoff into a surface pond designed to prevent groundwater contamination. The first two scenarios show the possibility for contamination of drinking water sources and would have an obvious environmental impact if not dealt with in a timely fashion. The third scenario poses a possible radiological hazard for workers at the site and is discussed in Section ii of this appendix.

Table AIII-3. Maximum Measured Non-radioactive Contamination in Pregnant Lixiviant

Contaminant	Concentration (mg/L)	Contaminant	Concentration (mg/L)
Arsenic	0.3	Barium	0.6
Boron	0.2	Cadmium	0.01
Chloride	1,800	Chromium	0.03
Copper	0.04	Flouride	1
Iron	0.02	Lead	0.01
Manganese	6	Mercury	<0.0001
Molybdenum	62	Nickel	0.09
Nickel	0.09	Nitrate	1
Selenium	5	Silver	<0.01
Sulfate	1,200	Total dissolved solids	5,500

(iv) Post-Operation Site Restoration and Rehabilitation

There are two main methods employed to restore the contaminated aquifer back to its preoperational conditions. In general, the first method employed is termed “groundwater sweep,” and involves pumping out the equivalent volume of groundwater from the mined aquifer and replacing it with fresh uncontaminated water. The volume of water pumped out of the mined ore zone is known as the “pore volume.” The pore volume can then be moved to an evaporation pond to remove the water and then dispose of the residual wastes. An alternate disposal of the pore volume is to inject the water through wells permitted through EPA’s SDWA UIC program into much deeper aquifers designated for waste disposal. In this case, the increased levels of contaminant should not affect neighboring aquifers or potential drinking water sources. This method has proven to be useful at the beginning stages of the restoration process. However, because of the heterogeneous properties of the ore zone aquifer, complete restoration of the mining site by this technique alone is not economical. Furthermore, many site locations do not have the resources for the large amount of clean groundwater that is required for an extensive groundwater sweep operation.

The second technique that can be employed is treating the contaminated pore volume via reverse osmosis. Here, the water is pumped out of the ore zone and passed through a reverse osmosis membrane at high pressure. This process separates the aquifer water into a highly concentrated liquid contaminant and a clean water volume known as the reverse osmosis (RO) permeate. The RO permeate is then recirculated into the ore zone using alternating pumping wells to effectively flush the heterogeneously distributed lixiviant present in the aquifer. The benefits of reverse osmosis are that no outside source of groundwater is needed to replace the pumped pore volume, since the volume is being treated and re-injected into the depleted ore zone. In practice, this method can only be employed after groundwater sweeping, because the high concentrations of contaminants during the initial stages of the restoration process tend to disrupt the RO membranes (Davis and Curtiss 2005).

Chemicals such as hydrogen sulfide or sodium hydrosulfide may also be added to the re-injected water during the later stages of restoration to achieve a state of “chemically reducing conditions.” The effect of these chemicals is to decrease the solubility of several contaminating

metals that are of concern, including uranium, selenium, arsenic, and molybdenum. However, there are other contaminants, such as radium, which remain mobile under chemically reduced conditions. Barium chloride is often used to precipitate radium out of waste water and can also be used during aquifer restoration to mitigate the effect of radium contamination (Mackin et al. 2001).

Despite these efforts at returning the mining site to its original preoperational state, it has been very difficult to achieve complete site rehabilitation. Not all of the contamination can be removed because lixiviant will be present in sections of the aquifer that are in areas of lower porosity. The efforts to create a chemically reduced condition to render the heavy metals insoluble may not apply to all contaminants of interest. Furthermore, achieving complete rehabilitation of the site is very time consuming and costly. New technologies, including bioremediation, may potentially improve the restoration of groundwater and wastewater impoundments (Rio Tinto 2007). The surface facilities and groundwater must be reclaimed in accordance with EPA requirements for radiation and environmental protection in 40 CFR 192 (see Volume I).

Summary

In situ leaching for uranium poses several possible environmental and health-related concerns. Through the extraction and processing of uranium ore into yellowcake, many hazardous chemicals and radionuclides are utilized or concentrated which, coupled with certain accident scenarios, can pose significant risk to workers at these facilities. From a radiological standpoint, risks are mainly significant to on-site workers, and have been shown to be minimal for the public (Mackin et al. 2001). From a hazardous chemical standpoint, the immediate concern is for on-site workers; however, the risk to the public can become significant if a prolonged release of hazardous material is allowed to contaminate nearby drinking water sources.

The leaching process poses the risk of contaminating neighboring aquifers which, in turn, might affect significant water supply sources. This can happen through horizontal and vertical excursions below the surface, or from events such as pipe failure on or near the surface. The risk of excursions is mitigated by the inclusion of vertical and horizontal monitoring wells located around the perimeter of the ore zone, as well as the operational practice of “process bleeding.” The wells are designed to detect excursions in a short period of time, so that corrective actions and cleanup operations can take care of the problem before the water sources outside of the mining site are significantly degraded.

Finally, in situ leaching poses a problem from a restoration standpoint. Although there are multiple techniques to restore the mined aquifer to its preoperational state, in many cases the lixiviant can never be completely purged from the site. Attempts to bring the aquifer to a chemically reduced state cannot account for all types of contaminants, and the entire rehabilitation process is both expensive and time consuming.

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