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MANAGEMENT OF WASTES CONTAINING RADIOACTIVITY FROM MINING AND MILLING OF URANIUM ORES IN NORTHERN AUSTRALIA

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Abstract

The procedures and controls to achieve safe management of wastes containing radioactivity during the mining and processing of uranium ores are mainly site-specific depending on the nature, location and distribution of the ore and gangue material.

Waste roc'. and below-ore-grade material containing low levels of radioactivity require disposal at the mine site. In open cut mining the material is generally stockpiled above ground, with revegetation and collection of run-off water. Some material may be used to backfill open cuts. Management of these wastes requires a thorough investigation of ground water hydrology and surface soil characteristics to control dissipation of radioactive material.

Dust containing radon and radioactive particulates is produced during ore milling, and dusts of ore concentrate are generated during calcination and packaging of the yellowcake product. These dusts are managed by ventilation and filtration systems; working conditions and discharges to atmosphere will be according to the Australian Code of Practice on Radiation Protection during Mining and Milling of Uranium Ores.

The chemical waste stream from leaching and processing of the uranium ores contains the majority of the radioactivity resulting from radium and its decay products. Neutralized effluent is discharged into holding ponds for settling of solids.

This paper describes the nature of wastes containing radioactivity resulting from the mining and milling of uranium, and illustrates modern engineering practices and monitoring procedures to manage the wastes, as described in the Environmental Impact Statement produced by Ranger Uranium Proprietary Limited for public hearings.

1. INTRODUCTION

Australia's uranium resources have been assessed [1] at 279,000 tonnes of uranium recoverable at costs up to $US 30/lb U_3O_8 ($A 62/kg U). Over 80% of these resources are situated in the Alligator Rivers area, Northern Territory [Fig. 1], in an area with a monsoonal climate and annual rainfall of up to 1900 mm over a 5 month period [2].

Present environmental legislation to minimize the consequences of mining operations did not exist when uranium was mined and milled at Rum Jungle. Procedures accepted at that time have resulted in a degree of environmental degradation which is unacceptable by modern standards. The heavy rainfall resulted in breaching of embankments retaining tailings, erosion of ore and waste stockpiles, and water contaminated with heavy metals, uranium, and its decay products entering local rivers [2,3].

It has been calculated that, after cessation of mining, up to 142, 80 and 56 tonnes of copper, manganese and zinc respectively were released during 1973-74, and there has been bacterial leaching from abandoned stockpiles of sulphide bearing waste rock [3].

The techniques for treatment and disposal of wastes from uranium production [4,5] have depended historically on the timescale of operations, the country of origin, local environmental conditions at the mine and mill and national attitudes towards the environment. A cost benefit analysis of alternative treatment procedures to reduce pollution has been conducted for US conditions [6].

Water management and disposal of tailings are key design features for meeting environmental standards for uranium production in the Northern Territory [7]. New Codes of Practice for Waste Management and Radiation Protection in the uranium industry have been written [8,9]. This paper reviews proposals for uranium development conforming to the new environmental requirements.

2. GENERAL SOURCES OF WASTES IN URANIUM MINING AND MILLING

Wastes produced during extraction of uranium from its ores are mainly site-specific; they depend in quantity and nature on the composition of the ore body and its host rock, the method of mining employed and the chemical treatment to recover uranium, and have been reviewed [4,10] as follows :

2.1 Uranium mining

(i) Open-cut mining produces solid wastes from the overburden and the host rock; underground mining produces less waste rock. Some Northern Territory deposits are suited to both methods. Solution mining, where the ore is leached in situ has not been suggested for this region.

(ii) Water containing minerals leached by natural weathering of costeans is produced during exploration, and from leaching of stockpiles of uranium ore, below-ore-grade materials and minerals in waste rock and overburden; contaminated surface and underground waters enter the mine.

(iii) Radon and other radioactive decay products are liberated from stockpiles of uranium ore and from the mine; siliceous and radioactive dusts are produced from blasting operations and from stockpiles; petroleum combustion products arise from excavation equipment.

2.2 Milling of uranium ores

(i) Siliceous dust containing radon and other decay products is produced during ore crushing and grinding.

(ii) Solid products (tailings) containing radioactive and mineral constituents of the ore and solid reagents, e.g. oxidants or neutralizing agents, are discharged from the leaching stage.

(iii) Liquid effluents generated during purification of the uranium contain chemical wastes from ore leaching, uranium purification and precipitation as a diuranate, dissolved mineral constituents rejected during purification, together with traces of uranium and radioactive decay products, and dissolved or entrained solvent.

(iv) Particulate dusts are produced from calcination of diuranate and crushing and packaging of yellowcake.

3. NATURE OF WASTES

3.1 Radioactive constituents of uranium ore

The radioactive decay series for uranium-238 is shown in Table 1. The radiologically significant isotopes in the series are the long-lived alpha emitters ^{238}U, ^{230}Th and ^{226}Ra; ^{222}Rn and its short-lived daughter products; and ^{210}Pb and ^{214}Po, the long-lived daughter products of ^{222}Rn. About 0.3 Ci of each isotope is associated with ore containing 1 tonne of uranium if secular equilibrium exists [11].

Up to 99% ^{226}Ra and of ^{230}Th may be rejected as insoluble waste at the leaching stage; ^{210}Pb, ^{210}Po and ^{214}Po may appear in wastes or with the uranium concentrate product, depending on the treatment processes.

3.2 Mineral constituents of uranium ore

The acid used in the leaching process will dissolve a proportion of minerals with soluble sulphates such as iron, copper, vanadium, molybdenum, and arsenic in addition to uranium present in the feed ore; the bulk of others such as lead will remain unleached. These soluble ions are rejected into the waste stream at the solvent extraction or ion exchange purification stage. Carbonate leaching processes extract uranium more selectively, leaving solid impurities with the gangue material.

3.3 Chemical wastes

The chemicals introduced in uranium milling are specific to the particular flowsheet, but typically can include sulphuric acid and sulphates, carbonates, chlorides and nitrates, bases such as ammonia, lime, magnesia and sodium hydroxide, and small amounts of potassium permanganate, copper sulphate, manganese dioxide, cyanides and polyacrylamide flocculents. Except for ions consumed during precipitation of diuranate (sodium, magnesium, calcium, or ammonium), the residual chemicals will ultimately be found in the waste stream.

Chemical wastes from an acid leach process allied to either solvent extraction or ion exchange will contain the spent sulphuric acid leachant and possibly a neutralizing agent e.g., lime. Manganese ion will be present if manganese dioxide is employed as an oxidant during leaching.

Milling processes recovering uranium by solvent extraction may include alkyl phosphates, secondary and tertiary amines, certain alcohols, kerosene and fuel oil. A small proportion of the organic extractants is lost by solution and entrainment in the waste stream. Wastes may additionally contain sodium ion derived from sodium carbonate employed in solvent washing processes.

Processes where uranium is recovered by ion exchange employ beds consisting of anionic resin beads. Losses of the solid resin to the waste stream occur through attrition and suspension. Wastes from the processes will also contain nitrate or chloride ions used for elution of uranium.

4. PROPOSED OPERATIONS IN THE NORTHERN TERRITORY

An ore deposit discovered by Ranger Uranium Mines Pty Ltd (RUM), and proposed for early development, has recently been the subject of a national environmental inquiry [12]. The No. 1 ore body contains about 20×10^6 t of ore averaging 0.25% uranium oxide for which open-cut mining methods are proposed. The second ore body is smaller; its upper portion is suitable for open cut mining while the lower might require underground mining. Both ores are suited to acid leaching.

The Environmental Impact Statement [13] proposed an annual production of about 2500 t uranium in yellowcake from about 1.2×10^6 t ore and 4×10^6 t waste rock, with provision for doubling throughput. Mining operations will involve drilling, blasting, loading, bulldozing and radiometric sorting of ore and waste rock, followed by hauling and separate stockpiling of primary ore and oxidized ore (0.05 - 0.25% U_3O_8), below grade material (0.02 - 0.05% U_3O_8) and waste rock (<0.02% U_3O_8). Treatment plant operations for recovery of uranium are shown schematically in Fig. 2.

5. MANAGEMENT OF WASTES IN THE RANGER OPERATION

RUM will conduct mining and milling [13] in accordance with certain recommendations [9,14]. Fig. 3 is an artist's impression of the Ranger mine and mill highlighting the waste management features.

5.1 Airborne wastes

5.1.1 Siliceous and radioactive dusts and radon

Dusts of siliceous rock containing radionuclides from uranium ore produced during open-cut mining and ore hauling and stockpiling will be laid by water sprays. Blasting will be timed and sized to facilitate dispersal of dust and radon gas. Primary drilling equipment will be operated from closed cabins; secondary drilling may involve wetting agents or dry extraction. Dust concentrations and radon in the open cut will be monitored and personnel exposure controlled.

Similar dusts produced during crushing and grinding of ores will be extracted by fans feeding wet scrubbers and returned to the grinding circuit. Yellowcake dust from calcination, crushing and product packaging will be similarly extracted, scrubbed and returned to the yellowcake thickener. Automated packaging equipment will be located in a separate building operated slightly below atmospheric pressure. At a production rate of 2500 t U/y, estimates of daily quantities of uranium in dust released to atmosphere [13] are: open cut (100 g), crushing plant and rod mill (1050 g), yellowcake plant (2200 g), with up to 260 g from ore and waste dumps dependent on wind speed [2].

An upper limit to the total daily discharge to atmosphere of 17.7 Ci of radon has been similarly estimated [2] for the following sources during production: mining operations and pit walls (7.3 Ci), ore dumps and stock-piles (2.6 Ci), crushing plant (2.2 Ci), treatment plant (1.7 Ci) and tailings retention system (3.9 Ci).

5.1.2 Sulphur oxides

Manufacture of sulphuric acid will result in the discharge of about 1 t/d of sulphur as SO_x (x = 2 and 3) to atmosphere through a 40 m high stack to ensure acceptable ground concentrations.

5.2 Waste rock

Initial waste rock from the open cut will be used for construction of roads, retention ponds and foundations; the majority will be consolidated into a dump of area 150 ha, height 120 m and containing 160 x 10^6 t [13].

5.3 Water management

Contaminated water for management will arise from the following sources:

- neutralized effluent and tailings from the treatment plant,
- water contaminated with minerals and radioactivity entering and pumped from the open cut,
- contaminated surface run-off water from ore stock-piles, the waste rock heap and the disturbed mine and mill catchment areas.

The water management program is based on on-site containment with recycle to the mill and disposal by evaporation of all mill effluents, the bulk of mine seepage water and a proportion of surface catchment and run-off water, with controlled discharge of the latter during wet season flooding ensuring adequate dilution and dispersion. Water management is achieved by a tailings retention system and three retention ponds (Fig. 4).

5.3.1 Retention of tailings

The slurry of leached tailings and solvent extraction raffinate, which contains the majority of the radioactive and mineral effluent from the mill, will be neutralized to pH 7-8 with lime before discharge to tailings retention. Neutralization of tailings is effective in reducing concentrations of dissolved minerals, activity and amine solvent which could otherwise be present in seepage water from the system [15]. Tailings neutralization has been compulsory in Ontario, Canada since 1960, possibly because of the wet environment, but is not widely practised in the USA [16], where modern mills retain tailings at pH 2 and rely on sandstone to neutralize and precipitate contaminants subsequent to seepage [16,17].

The RUM tailings retention system is designed to retain all solids and minimize seepage through walls and base; it conforms to standards issued by the International Commission on Large Dams (ICOLD) [18]. It has a design capacity for 27 x 10^6 t of settled tailings over a storage area of about 100 ha and a perimeter embankment initially of height 16 m to be increased in stages to 30 m. Construction of a second retention system to contain a further 23 x 10^6 t of tailings is proposed. Embankment construction from earth and rockfill was selected on bases including low permeability and known seismic resistance. Tailings will be stored under a blanket of at least 2 m of water during the life of the plant to prevent drying out and dispersal by wind; on cessation of operations the retention system will be stabilized by revegetation.

5.3.2 Water management operations

The system in Fig. 4 consists of three sections [19,20]:

(a) A closed circuit with water recycle between the mill and tailings retention system; water losses from this circuit will only occur through evaporation and by seepage through the walls and floor of the dam.

(b) An open circuit collecting contaminated water from the open cut, to be used as a primary source of make-up water for the mill. Some discharge of this water to the environment, possibly with treatment, e.g. to retain radium, may be required in later operation [2].

(c) An open circuit collecting in two retention ponds the run-off from catchment areas where ore and waste are stockpiled. A controlled discharge of this water after analysis is proposed during floods. The proposal to maintain control of open catchment areas is unique in the mining industry [20]. Annual estimates of discharges are heavy metals - 200 kg; radioactivity - 0.05 Ci Ra and 0.07 Ci U [2] which are small by comparison with estimates [13] of normal annual flow in a local creek of about 4 t of heavy metals and 0.2 Ci Ra.

6. FURTHER ENVIRONMENTAL WORK

Inevitably, there are some uncertainties in quantifying releases of contaminants to the environment, for example [2],

- the particle size distribution and source strength in release of siliceous and radioactive dusts and radon during operations;

- the quality and quantity of water necessary to be discharged from the process;

- the long-term solubilities of solid contaminants in a retention system; and

- the optimum measures to rehabilitate the area after mining and milling have ceased.

The surveillance program to be carried out during operations is required to confirm the accuracy of the present estimates. Supporting work by the AAEC aims at improving management of wastes from uranium mining and milling, and obtaining information for the determination of realistic standards in waste management at proposed operations, and for assessment of the cost effectiveness of the treatment options under Australian conditions.

Items of particular interest include:

(a) The study of radium and heavy metal concentrations in mixtures of tailings and process waste solutions following neutralization with lime and treatment with barium chloride or barium sulphate.

(b) Determination of the factors and mechanism governing the natural leaching of radium from tailings.

(c) Determination of radon release from ores and tailings as a function of ore type and particle size and from submerged tailings retention systems, and assessment of the feasibility of removing radon from air streams.

(d) Studies of surface hydrology at prospective mines and mills to determine the destination of released contaminants.

(e) Bioassay studies of the effects of released contaminants including determination of bioaccumulation factors in ecosystems and investigation of transfer mechanisms in food chains.

7. CONCLUSION

The hazards of mining and milling uranium, if properly controlled, are not sufficient to justify a decision not to develop uranium mines in Australia [12]. The uranium mining industry has shown a capability and willingness to conform to environmental standards laid down by national and international bodies.

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TABLE 1

THE URANIUM RADIOACTIVE DECAY SERIES

Isotope	Symbol	Historical name	Half-life	Radiation	Alpha energy (MeV)†	Remarks
Uranium-238	U-238	Uranium I	4.5×10^9 y	α		β rays in uranium ore come from these
Thorium-234	Th-234	Uranium X_1	24.1 days	β γ		
Protactinium-234	Pa-234	Uranium X_2	1.18 min	β γ		
Uranium-234	U-234	Uranium II	2.50×10^5 y	α γ		
Thorium-230	Th-230	Ionium	7.6×10^4 y	α		end up in tailings
Radium-226	Ra-226	Radium	1620 y	α		
Radon-222	Rn-222	Radon	3.82 days	α	5.49	gas
Polonium-218	Po-218	Radium A*	3.05 min	α	6.00	collects on dust in mine
Lead-214	Pb-214	Radium B	26.8 min	β γ		β and γ rays in ore come from these
Bismuth-214	Bi-214	Radium C	19.7 min	β γ		
Polonium-214	Po-214	Radium C'*	2.7×10^{-6} min	α	7.69	
Lead-210	Pb-210	Radium D	22.0 y	β γ		May be used to monitor Rn-222 exposure
Bismuth-210	Bi-210	Radium E	5.0 days	β		
Polonium-210	Po-210	Radium F	138.4 days	α	5.30	
Lead-206	Pb-206	Radium G	Stable			

* *Ra-A and Ra-C' constitute the alpha hazard in exposure to Rn-222 and its daughters.*

† *MeV stands for million electron volts. It is a measure of the energy with which radiations are emitted from radioactive isotopes. A 5 MeV alpha-particle will penetrate some 40 microns of soft tissue.*

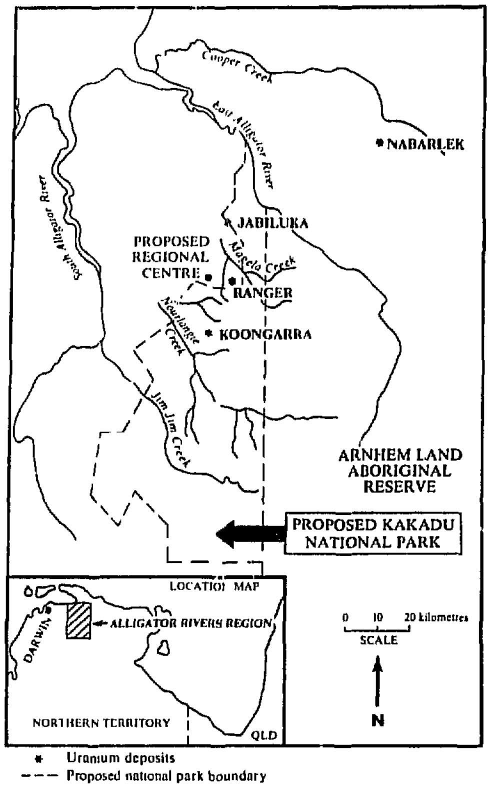


FIGURE 1. MAJOR URANIUM DEPOSITS IN THE NORTHERN TERRITORY
(From Reference 12)

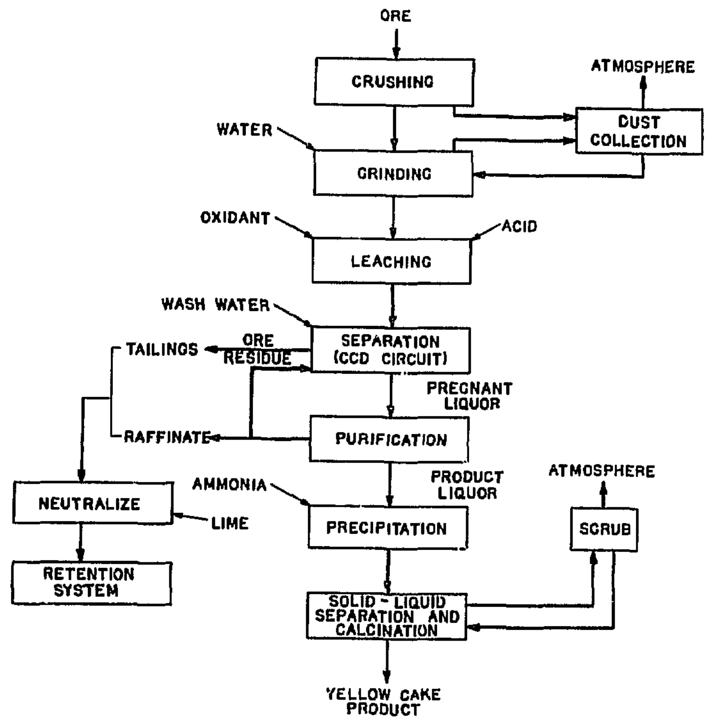
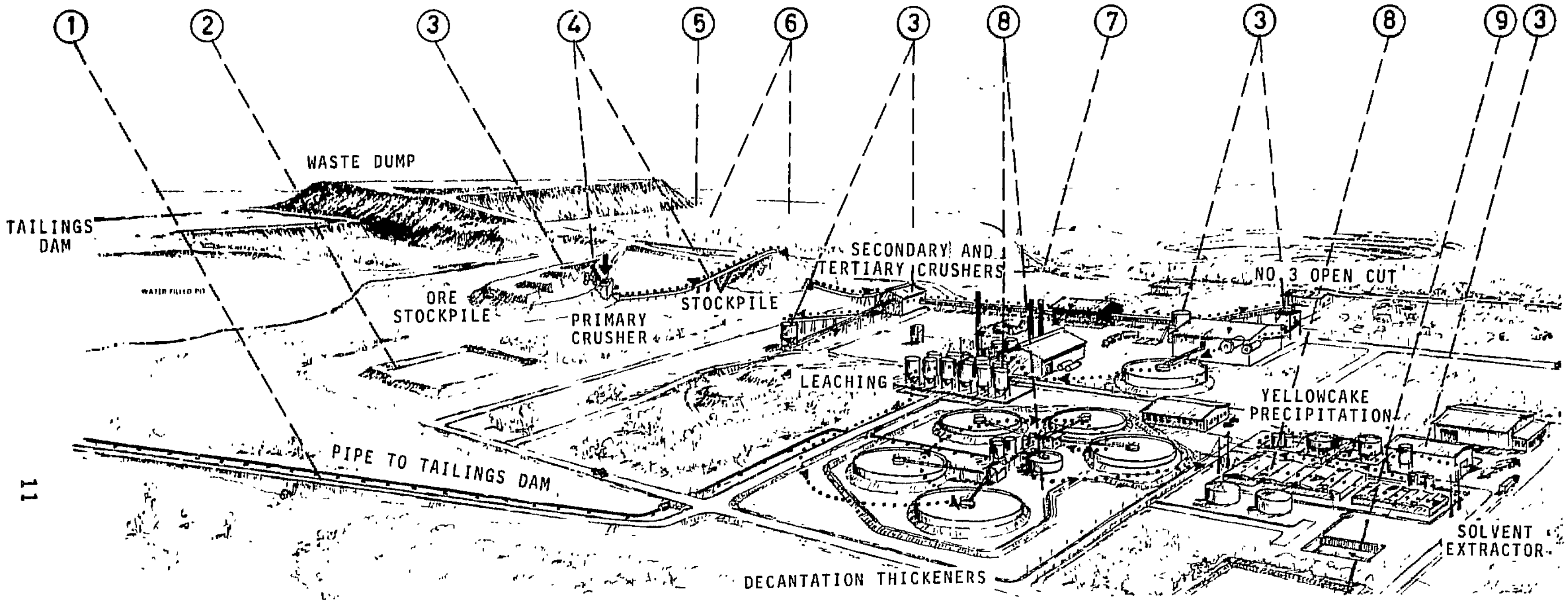


FIGURE 2. URANIUM MILL - ACID LEACH PROCESS SIMPLIFIED BLOCK FLOW DIAGRAM



(1) Road raised to act as dam for spills from tailing pipeline

(2) Mine water storage pond holding water till safe to release

(3) Dust collector/scrubber achieving 95% dust removal

(4) Water spray reducing dust liberation

(5) Dam collecting run off from waste heap

(6) Bores sunk to monitor contamination of groundwater

(7) Dam collecting run off from plant area

(8) Area bunded to contain spillage

(9) Sump allowing return of spillage to process

FIGURE 3. ARTIST'S IMPRESSION OF THE RANGER MINE & TREATMENT PLANT (From Reference 13)

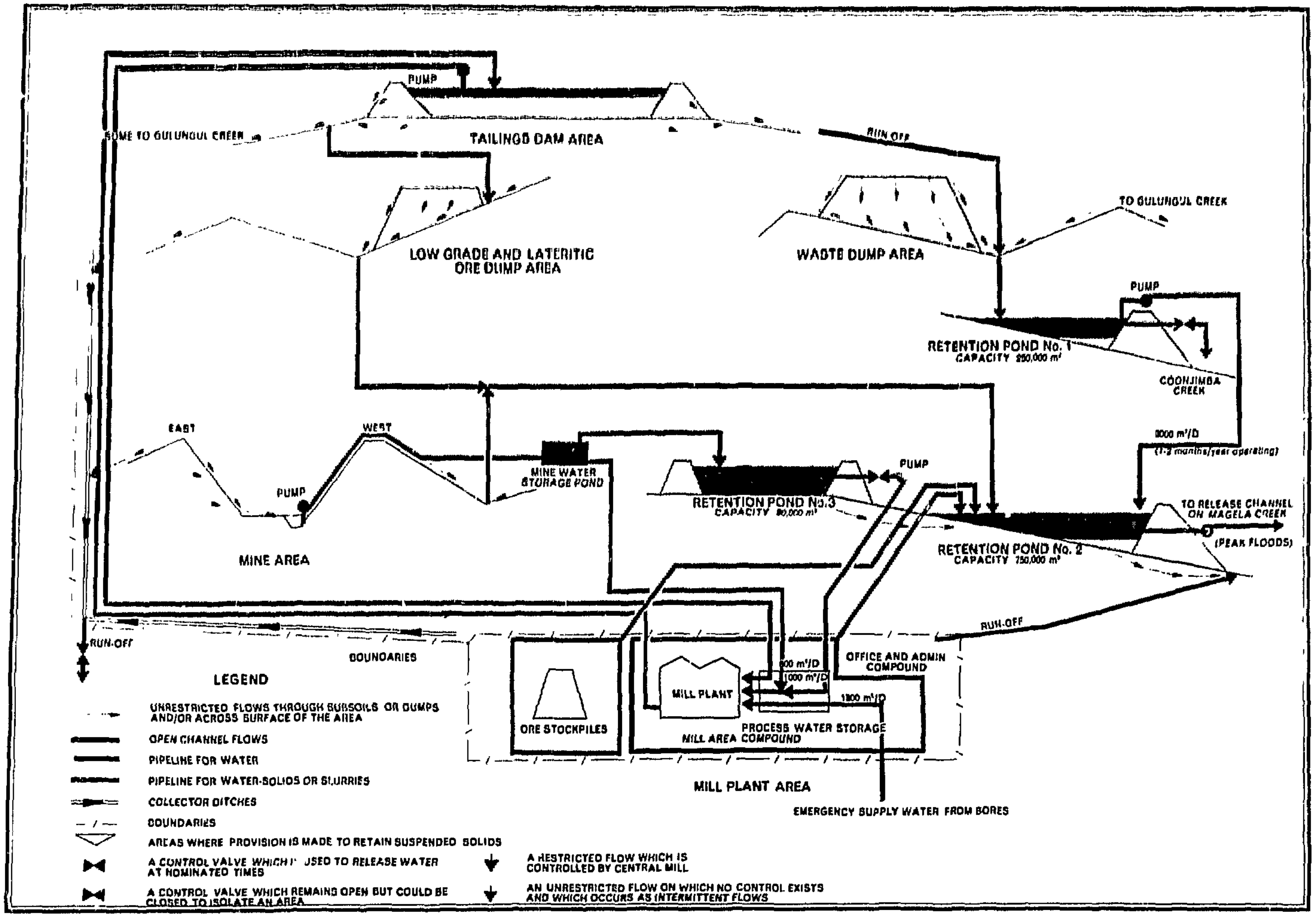


FIGURE 4. SITE WATER MANAGEMENT DIAGRAM (From Reference 13)