Mines Branch Information Circular IC 125

NOTES ON THE SAFE HANDLING OF URANIUM ALLOYS IN INDUSTRY

by

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SYNOPSIS

This Circular summarizes some of the physical and chemical properties of uranium metal which may have a bearing on the industrial production of uranium-containing alloys. The legal aspects of natural uranium use are reviewed and some experimental measurements on airborne uranium dust obtained during pilot plant tests are presented.

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Circulaire d'information IC 125

NOTES SUR LE MANIEMENT SANS DANGER DES ALLIAGES D'URANIUM DANS L'INDUSTRIE

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RÉSUMÉ

La présente Circulaire expose brièvement quelques-unes des propriétés physiques et chimiques de l'uranium métal qui pourraient influer sur la production industrielle d'alliages uranifères. L'auteur examine les aspects juridiques de l'emploi de l'uranium naturel et présente certaines mesures expérimentales des poussières d'uranium présentes dans l'air, prises au cours d'essais dans une installation pilote.

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INTRODUCTION

The recent metallurgical development of uranium steels (1) and of other uranium alloys of potential industrial interest makes it desirable to review the handling problems that may arise in large-scale use of uranium metal for such purposes. For this reason, a number of facts concerning uranium and its alloys have been compiled here for convenient reference. This report is not intended to be a complete listing of reports and literature concerning the safe handling of uranium and only those matters have been selected which may be relevant in dealing with uranium alloys in the range of compositions contemplated at present. It will be assumed throughout that the uranium involved is natural uranium, or uranium depleted in its normal content of the isotope U-235.

PHYSICAL AND CHEMICAL CHARACTERISTICS OF URANIUM METAL

In the production of uranium alloys there are three distinct stages of materials handling: 1) the transportation, storage and handling of pure uranium metal; 2) the heating and casting of the metal mixture; and 3) the further treatment and handling of the finished alloys. Before dealing with these processes the main characteristics of uranium will be summarized.

Uranium is a hard, heavy, grey-white metal of density 19 g/cm^3 at room temperature (3, 4). Its atomic weight is 238.1,

its melting point 1129°C (2064°F), its boiling point 3838°C (6940°F), its valence 3, 4 or 6. Uranium metal is highly reactive; it readily reacts with non-metallic elements and forms numerous intermetallic compounds with Group III and transition group elements (5, 6). Its vapour pressure is 5.2×10^{-6} mm at 1410°C (2570°F), 2.8×10^{-5} mm at 1500°C (2732°F) and 1.4×10^{-4} mm Hg at 1600°C (2912°F) (7).

Because of its reactive nature, uranium is highly pyrophoric when in finely divided form (6). This property seems to be associated with the formation of hydrides on exposure to moisture and is a factor of importance in the storage and handling of finely divided uranium metal, such as uranium chips and turnings (7). It is no problem for massive pieces, where the ratio of bulk volume to surface area is high.

RADIATION CHARACTERISTICS OF URANIUM

Uranium, as is well known, is a radioactive element, that is, it decays spontaneously, changing its physical and chemical nature in the process. The probability of such decay taking place is usually expressed in terms of the half-life of the material, which is the time in which, on the average, half the atoms involved will have decayed. In the case of uranium the decay products themselves are radioactive and a whole chain of radioactive elements is produced. On purification and refining, the uranium is freed from all the decay products, notably radium and its daughter products, which have

accumulated in the mineral over the years.

Pure uranium metal is made up essentially of three isotopes of the element; uranium-238, uranium-235, and uranium-234. Table 1 lists the main properties of these isotopes and their immediate decay products (10, 11). Only the more important radiations, contributing at least 10% of the total radiation for any nuclide, are listed.

As can be seen from Table 1, highly-purified uranium will emit mainly alpha particles when freshly prepared. After a few weeks, however, the beta- and gamma-ray emitters Th-234 and its short-lived daughter Pa-234, and to a lesser extent Th-231, will build up to equilibrium concentrations and emit feeble, but measurable beta and gamma radiations. Because of their low energy these radiations are emitted essentially from the surface of the metal only, the rest being absorbed internally, and an increase in bulk of the material will not greatly increase the total amount of radiation detected. The longer-lived decay products of uranium, listed in Table 1, will not build up again in measurable quantities once they have been separated out. This applies particularly to radium-226 which, through its decay products, is the principal source of the gamma radiations detected in natural uranium ores. As alpha particles are readily absorbed by a few inches of air, a thin film of moisture or any thin layer of paper, the beta rays of protactinium-234 constitute virtually the only radiation readily

TABLE 1

Properties of Principal Uranium Isotopes and Important Daughter Products

Nuclide	Abundance %	Half-life	Type of decay	Decay product	Energy of particles (MeV)	Energy of γ-rays (KeV)
U-234	0.0058	2.48×10^5 yr	α	Th-230	4.76, 4.71	---
U-235	0.71	7.13×10^8 yr	α, γ	Th-231	4.39, 4.56	180, 140
U-238	99.28	4.51×10^9 yr	α	Th-234	4.18	---
Th-230		8×10^4 yr	α, γ	Ra-226	4.68, 4.61	190, 68
Th-231		25.6 hr	β, γ	Pa-231	0.09, 0.30	22, 80
Th-234		24.1 d	β, γ	Pa-234	0.19, 0.10	93
Pa-231		3.4×10^4 yr	α, γ	Ac-227	5.00, 4.94	95, 320
Pa-234		1.18 min	β	U-234	2.32	---

detected from uranium alloys and have been utilized as a convenient means of checking the uniformity of composition of such metal samples.

As is well known, the nuclide uranium-235 is fissionable and represents the most common and convenient source material for nuclear energy development at this time. However, in the concentrations in which it is found in natural uranium it is impossible to start a nuclear chain reaction in air, no matter how great the quantity of uranium involved. In other words, natural uranium, without any enrichment in U-235 content, cannot by itself reach critical conditions (12, 13). In a reactor such critical conditions can be created by immersion in heavy water or other moderating material. In any industrial plant, however, solid natural uranium metal can be piled readily without any worry on this score (so long as the total tonnage is kept low and no lattice is formed with a moderator, such as graphite).

TOXICITY CONSIDERATIONS

From the foregoing, it is evident that the radiation emitted from uranium metal is neither penetrating nor intense. It does not, therefore, constitute a serious external radiation hazard. On the other hand, when uranium is ingested into the body its long half-life and the high energy of the alpha particles may constitute a hazard. It is important, therefore, to avoid ingestion, by handling the metal with gloves at all times, by not eating or smoking while handling pure uranium metal, and by reducing the possibility of inhaling

dust-borne uranium particles.

The chemical toxicity of uranium is a much more serious health factor than its radioactivity (14). Uranium in soluble form is concentrated in the kidney and some cases of acute uranium poisoning have been reported in the literature. The toxicity of uranium is comparable to that of lead or mercury, though the detailed physiological effects are different. This can be seen from a comparison of the maximum allowable concentrations in air (15), as given in Table 2.

TABLE 2

Maximum Allowable Concentrations (MAC)

Metal	Form	MAC in air
Mercury	metallic	0.1 mg/m^3
Mercury	organic compounds	0.01 mg/m^3
Lead	metal	0.15 mg/m^3
Uranium	soluble	0.05 mg/m^3
Uranium	insoluble	0.25 mg/m^3

In the nuclear energy industry it is customary to express maximum permissible concentrations in microcuries/cc of air for any radioactive isotope*. Table 3 lists the 1959 recommendations of the International Commission on Radiological Protection (I.C.R.P.) (16), which have been used as a guide by the Government of Canada (2). In columns 2 and 4, the microcurie per cubic centimetre (μc/cc) values have been converted to corresponding $\mu g/m^3$ values.

TABLE 3

I.C.R.P. Tolerances for Uranium

	Maximum Permissible Concentrations in Air			
	occupational exposure		general public	
	μc/cc	$\mu g/m^3$	μc/cc	$\mu g/m^3$
Uranium-natural (soluble)	7×10^{-11}	200	2×10^{-12}	5.7
Uranium-natural (insoluble)	6×10^{-11}	170		

The figures in Table 3 are seen to be slightly more stringent than those listed in Table 2, which are based on normal industrial practice (17); however, the I.C.R.P. values are the ones that must be met in any future operations involving uranium (18). Experience has shown that with proper ventilation no difficulties should be encountered in maintaining low uranium concentrations in air during uranium-alloy preparation. This also applies to uranium storage areas, which should be kept dry and well-ventilated.

* Units are defined in the Appendix.

PREPARATION OF URANIUM STEELS

In discussing the above rules and considerations as they apply to the production of uranium steels containing up to 0.5 per cent uranium by weight, two stages must be clearly distinguished. The first is the actual foundry operation, involving molten metal at a temperature well above the melting point of uranium. This operation is relatively brief and any liberation of uranium to the air will cause only short-time exposure of the operator. The second is the cold handling, forming and fabricating of the finished alloy, where the concentration of uranium is very low but long-term exposures are possible.

Tests on various uranium steels have been conducted at the Mines Branch laboratories of the Department of Mines and Technical Surveys over the past year. During several of these tests air samples have been collected to establish representative levels of uranium concentration in foundry air before, during and after typical casting and rolling operations. Table 4 lists the results of some typical runs. The two tests were not identical. In Test No. 1 a 500 lb heat of steel was poured into three ingots containing 0, 0.05 and 0.10% uranium respectively, with the uranium added to the ladle. In Test No. 2, 0.2% uranium was added to the furnace and allowed to diffuse before pouring; the metal was then cast in open troughs. It should also be borne in mind that the slag layer is

relatively thin in this type of batch operation. To determine the uranium content, the air filters were all run under an end-window Geiger counter. Allowance has to be made for the decay of bismuth-214 which arises from radon which is normally present in the atmosphere. The count rates listed constitute, therefore, the stable residue after Bi-214 decay. Several filter samples were obtained simultaneously in different locations and this accounts for the overlap in time and the variations in uranium picked up. It is seen that the count rates were quite low in most places, both before and after the casting operation. Filters 6 and 8 record a momentary increase in uranium concentration, which approaches the maximum value for non-occupational exposure, but this activity has been traced to the stirring up of dust during sweeping and clean-up operations and has been reduced in later tests. In any case, the exposure values listed in Table 3 represent average values for continual exposure during a 40-hour week and on that basis the figures in Table 4 show a comfortable margin of safety. Investigations are under way, even so, to find an explanation for the sudden increase in airborne uranium during pouring of the metal.

Regarding the finished alloy it will be realized that the uranium at this stage is well-dispersed and constitutes no chemical or radiation hazard. A rough calculation, using the absorption factors given in the literature, shows that steel containing 0.1% uranium will emit 360 betas/min/cm^2 of surface from a 1/8 in.

TABLE 4

Foundry Air Samples - January 11, 1961

Filter No.	Test No.	Location	Exposure Time	Volume sampled (cu ft)	Net count rate after three days (c/m)	Contained uranium ($\mu g/m^3$)
1		6 ft from moulds	9:35 to 10:29 am 54 min	2484	25.6	0.45 (calc)
2		10 ft from moulds	9:35 to 10:29 am 54 min	2376	11.9	0.22 (calc)
3	1	Close to ladle during pouring	11:04 to 11:10 am 6 min	288	Backgd.	--
4	1	6 ft from moulds	11:04 to 11:15 am 11 min	605	4.48	0.33 (calc)
5	1	10 ft from moulds	11:04 to 11:15 am 11 min	594	1.66	0.12 (calc)
6		6 ft from moulds	11:17 to 2:0 pm 163 min	7708	219.0	1.38 (chem)
7		10 ft from moulds	11:17 to 2:0 pm 163 min	8200	46.1	0.26 (calc)
8	2	6 ft from moulds during and after pouring	2:15 to 4:30 pm 135 min	6345	162.8	1.02 (chem)
9	2	10 ft from moulds during and after pouring	2:15 to 4:30 pm 135 min	6345	38.3	0.26 (calc)
10	2	8 ft from furnace during heating and slagging	2:23 to 2:52 pm 29 min	1537	21.9	0.62 (calc)
11	2	Close to ladle during pouring	3:12 to 3:20 pm 8 min	424	1.3	0.13 (calc)

thick plate; this activity will increase with thickness to a saturation value. A 1/4 in. thick plate will emit 450 betas/min/cm^2. To illustrate this point, Table 5 lists comparative count rates obtained with a portable Geiger counter for three uranium steel samples, about 4 sq in. in area and for a typical wrist watch with a luminous dial.

TABLE 5

Radiation Readings

Sample	Weight	U content	Gamma counts	Beta counts
Background	--	--	60 c/min	80 c/min
1 C U	129 g	0.063%	60	80
2 C U	122 g	2.11 %	80	2700
3 U	119 g	0.003%	60	80
Watch dial	--	--	180	2950

Larger quantities of the alloy samples will produce relatively small increases in count rates, because of self-absorption and self-shielding effects. To show this, the radiation from a pile of six uranium steel ingots, weighing about 1 ton and containing 0.05 % U, was measured. The results are shown in Table 6.

TABLE 6

Radiation Dose from 2100 lb Uranium Steel Ingots

Configuration	Position of dosemeter	Dose measured
Ingots lying flat, side by side	2 in. above middle of row	less than 0.1 mr/hr
	touching ends of middle ingots	"
	1 in. from end of row	"
Ingots lined up upright	1 in. from end of row	"
	along side of row	"
	2 in. above middle ingots	"
Ingots grouped in tight circle	inside circle	"
	2 in. above centre of circle	"
	along outside face of circle	"

It is evident that the dosage values are well below the maximum permissible dosage of 2.4 mr/hr for occupational exposure, or 0.2 mr/hr for non-occupational exposure for continuous work over a 40-hour week. It compares with a normal unavoidable daily dose of 2 mr for radiation from all natural causes (15).

LEGAL ASPECTS

In Canada uranium is a "prescribed substance" under the Atomic Energy Control Act. The Atomic Energy Control Regulations (2) lay down that an Authorization from the Atomic Energy Control Board is required to authorize dealings within Canada as regards uranium contained in any substance that contains not less than 0.05 per cent by weight of the element uranium, or for dealings which involve, during any calendar year, a total of at least 10 kilograms of contained uranium element.

In the Appendix to the Regulations the "scheduled quantity" of natural uranium is listed as 500 microcuries*. This is equivalent to 1420 grams of natural uranium. The Regulations also lay down the maximum permissible dose of ionizing radiation for whole-body exposure as 3 rem for any period of 13 consecutive weeks, and 5 rem for any period of 52 consecutive weeks. For exposures restricted to the limbs, higher permissible doses are allowed.

Any large-scale use of uranium in industry requires, therefore, the authorization and approval of the Atomic Energy Control

* Dealings in smaller amounts are not subject to regulation.

Board. In addition most provinces require concurrently the approval of the provincial health department, which should be consulted to ensure conformance with local regulations.

Shipping and handling of uranium steel material should present no hazards. Transportation is subject to the Board of Transport Commissioners' Regulations (19), but these concern primarily the rules for labelling and for maximum external radiation levels which have to be maintained. Bulk uranium steel should have less external radiation than bulk uranium ore which can be shipped in open cars at present.

CONCLUSIONS

The production of uranium-bearing steel by industry should present no special problems as long as certain elementary precautions are observed in the handling of the raw material and proper ventilation of all working areas is provided. This is reflected by recommendations in the literature advocating the use of uranium metal as ballast weights in aircraft and in portable isotope containers (20), as well as by discussions on disposal of uranium-contaminated scrap steel (21, 22).

Any company entering this field would be well advised to work in close contact with the relevant Federal and Provincial authorities who are conversant with this field and prepared to advise and assist in any preliminary operations.

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APPENDIX

Definition of Units (2)

1. Microcurie (μc) - means that quantity of a radioactive substance that is disintegrating at the rate of 37,000 disintegrations per second*.

   (1 millicurie (mc) = 1000 microcuries)

2. Roentgen (r) - means that quantity of X- or γ radiation such that the associated corpuscular emission produces in 1 cc of air, at normal temperature and pressure, ions carrying one electrostatic unit of quantity of electricity of either sign (16).

   (1 milliroentgen (mr) = 10^{-3} roentgens)

3. Rem (rem) - means, in relation to the body or any organ of the body, the dose of any ionizing radiation that has the same biological effectiveness as a dose of 200-250 thousand volt X-rays whose energy is absorbed by the body or such organ in the amount of 100 ergs per gram.

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* By long-established usage in internal dose calculations, one curie of recently extracted uranium is considered to correspond to the sum of one curie from U-238, one curie from U-234 and 0.0243 curie from U-235 (23).