بسم الله الرحمن الرحيم

SUDAN UNIVERSITY OF SCIENCE & TECHNOLOGY

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A Study of Uranium Enrichment Techniques

دراسة تقنيات تخصيب اليورانيوم

THESIS SUBMITTED AS REQUIREMENTS OF THEDEGREE OF M.Sc.IN PHYSICS

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بسم الله الرحمن الرحيم

الآيــــــــــة

قال تعالى :

﴿شَهِدَ اللَّهُ أَنَّهُ لا إِلَهَ إِلَّا هُوَ وَالْمَلائِكَةُ وَ أُولُو الْعِلْمِ قَائِماً بِالْقِسْطِ ﴾

صدق الله العظيم

سورة آل عمران الآية (18)

I

DEDICATION

To:

My

Family

Acknowledgment

First of all praises and glary be to Allah almighty Lord of the world the most beneficent, and the most merciful. May His blessing be upon His prophet almighty and all those prophets before him.

 Second I thank Dr. Rawia AbdElgani Elobaid Mohammed , supervisor of this thesis, who gave her time and the benefit of her experience and knowledge.

Without her help, this would not have reached a successful conclusion, owe her much more than words can describe. Special thank goes to my parents

Finally, and above all thank god, who helped me to bring this work to completion.

Abstract

This research is a study of the techniques of uranium enrichment and physical separation of isotopes, which often resort to when it is difficult to separate them by the usual chemical methods, including many of the mixtures that have already been used, such as uranium enrichment by laser, more common centrifuges, diffraction, thermal spread, magnetic separation and plasma.

The enrichment process is important to take advantage of the uranium element as a fuel for various nuclear reactors, in the military industry (nuclear bomb), the production of electric power, some medical and agricultural industries and the monitoring of other industrial products.

In all various enrichment processes, were produced is depleted uranium, which has become an important and effective role in some reactors (covert generation reactors), heavy industries, armor-piercing super-armor and radiation protection.

All enrichment techniques are technically very difficult and complex, and need to develop special programs with a relatively long time, patience, accurate and continuous monitoring to avoid problems and the periodic maintenance of the equipment used.

It is very difficult to enter the world of nuclear industries because all scientific information is highly clandestine and represents the national wealth of the major industrial countries that monopolize it as a safeguard against the spread of nuclear weapons.

The enrichment of uranium in its capacity is linked to economy and politics as well as scientific and technical cooperation between countries and educational institutions such as universities can enter the field of enrichment, such as research and development projects and companies as enriched uranium to extract capital.

المستخلص

هذا البحث هو دراسة علمية عن تقنيات تخصيب اليورانيوم وعمليات فصل النظائر بالطرق الفيزيائية والتي غالبا ما نلجأ اليها عندما يصعب فصلها بالطرق الكيميائية المعتادة ،وتتضمن العديد من النماذج التي استخدمت بالفعل مثل تخصيب اليورانيوم بالليزر وأجهزة الطرد المركزي الاكثر شيوعا والانتشار الغازي والانتشار الحراري والفصل المغنطيسي والبلازما.

وتاتي اهمية عمليات التخصيب للاستفادة من عنصر اليورانيوم كوقود للمفاعلات النووية المختلفة وفي الصناعات العسكرية (القنبلة النووية) وانتاج الطاقة الكهربائية وبعض الصناعات الطبية والزراعية وعمليات مراقبة المنتاجات الصناعية الاخرى .

في جميع عمليات التخصيب المختلفة ينتج مايعرف باليورانيوم المنضب الذي اصبح له دور مهم وفعال في بعض المفاعلات (المفاعلات المولدة السرية) والصناعات الثقيلة وصناعات رووس القذائف الخارقة للدروع والوقاية من الاشعات .

ان جميع تقنيات التخصيب هي في الواقع صعبة جدا من الناحية التقنية ومعقدة ، وتحتاج الي وضع برامج خاصة بمعدل زمني طويل نسبيا وصبرومتابعة دقيقة ومستمرة للتغلب علي المشاكل الانية وصيانات دورية للمعدات المستخدمة .

ان الدخول في عالم الصناعات النووية صعب جدا لان جميع المعلومات العلمية تعتبر سرية للغاية وتمثل ثروة وطنية للدول الصناعية الكبرى التي تحتكرها كوقاية للحد من انتشار السلاح النووي .

ان تخصيب اليورانيوم في حد ذاته مرتبط بالاقتصاد والسياسة بالاضافة الي التعاون العلمي والتقني بيين الدول ويمكن للمؤسسات التعليمية مثل الجامعات ان تدخل مجال التخصيب كمشاريع بحث وتطوير،والشركات باعتبار اليورانيوم المخصب مصدرجيد لجلب رووس الاموال .

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Table Abbreviations and Parameters Name

Token	Title	Unit
C_n	Specific Heat of Uranium	gol/kg.k
T	temperature	K OR °C
P	Vapor pressure	mm Hg
l	Length	m
G	Gravity Wheel	m/sec^2
σ_B	Tensile strength	kg/mm^2
δ	Elongation	non
M_{238}	Mass of $U^{223}8F_6$	a.m.u
M_{235}	**Mass of $U^{235}F_6$**	a.m.u
R_P	Ratio of numbers emerging on the product	-
Q		kcal/mol
KE	Kinetic energy	gol or ev
m	Mass	kg
v	Velocity	m/sec
Λ	Wavelength	m
σ	The absorption cross-section	Cm^2
Φ	The quantum flux	-
W	Energy of the pulse per unit area	μJ cm^{-2}
Ni	The atom at the ith level	
$h\nu_i$	Refers to the radiation in a transfer from the (i + 1)st to the ith level.	-
c	The speed of light	m/sec^2
Gi	The statistical weight of the ith level	-
A_i+1	The Einstein factor for the transition considered	-
e_B	Barrier efficiency	non
g	Actual separation gain	-
G_{ideal}	Ideal value	-
t_i	Total radiation lifetime of the level	second
n_{ph}	The photon density at the corresponding transition.	-
Z	The coordinate along the laser beam.	-
F	Feed stream	-
P	Product stream which is enriched in the desired isotope	-
T	The tails stream which is depleted in the desired isotope.	-
δU	Is the separative work and other symbols have the same Meaning as before	-
dM	Molecular weight difference	-
V	Peripheral velocity	-

R	Universal gas constant	-
T	Absolute temperature	-
α_0	Centrifuge separation facto	-
ℓ	Rotor length	m
D	Rotor diameter	
PE	Potential energy	
ω	Angular velocity	rad/s
ρ	Gas density	
D	Coefficient of self diffusion	
ΔM	Difference of molecular weights of isotopes being Separated	-
q	Chemical separation factor	-

Chapter One

1.1 Introduction

The enrichment industry flourished during the years following the Second World War for various purposes, including the production of a nuclear bomb and the operation of nuclear reactors to produce industrial fissile materials, as well as meeting the needs of the developed countries of electric power, which led to the development of technologies used for this purpose significantly, in order not to spread nuclear technology and use it is wrong.

The enrichment of uranium is not a danger to the world, but the goals for which are designed to enrich. There are many countries that seek to acquire nuclear technology for purposes that may be peaceful.

1.2 Aims

1. Provide basic process of uranium enrichment scientific information.
2. Put the general idea of each technique enriched with detail techniques.
3. Statement of uncertainty that accompanied the uranium enrichment technology as much as possible.
4. Benefit from research and previous experiences in enrichment.
5. Encourage students and researchers to go deeper in the field enrichment and a strong basis for the entry of nuclear industries.
6. Clarify the lack of seriousness of the uranium enrichment process.
7. Demonstrate the possibility of the country's uranium enrichment even limited potentials by providing the necessary information and make it a place of research and scientific debate.
8. Assist in providing the necessary information for the enrichment process for the purposes of research and development.

1.3 Research problem

1. Research discussed different ways to enrich uranium and analyze comprehensive and accurate analysis.
2. Showing some secret physics concepts that did not discuss the process of uranium enrichment by linking the information available with each other.

1.4 Importance of research

1. State measured the strength and progress of scientific and technological ability to produce enriched uranium and benefit from it in different applications.
2. Showing enrichment techniques and discussion lead to the development and easy access to nuclear and other industries

1.5 Previous studies

1. **Laser and Centrifuge Enrichment,Belfer Center for Science and International Affairs,Harvard Kennedy School, 3 November, 2013**

This research discussed the uranium enrichment technology, the laser generated from carbon as a technology monopoly of some countries, in addition to some of the basics in Laser Physics.

1. **Centrifuges: A new era for nuclear proliferation ,R. Scott Kemp2004**

Showing the general idea of uranium enrichment centrifuges and marketed with some theoretical fundamentals and areas of deployment and limit the spread of nuclear weapons.

2. **Uranium enrichment technologies Siv Gunderso Aalbergsjø2008**

This study dealt with uranium enrichment technology with some objectivity and focused on the comparison between the methods used between States and the Models of each country in this area.

3. **Introduction to Gas Centrifuges** , **Houston Wood, University of Virginia, Presentation for ISIS, February 13, 2003**

General information on the enrichment of uranium by centrifuges in addition to some of the basics of centrifuges.

4. **Isotopic separation and enrichment,M.ragheb 4/15/2015**

Studying isotope separation and applied to uranium in different spaces to (Gaseous, liquid, solid).

1.6 Thesis logout

Chapter One: Introduction, Aims, Research Problems, Importance of Research, Previous Studies, Thesis logout.

Chapter Two: Uranium Nature.

Chapter Three: Physical Properties of Uranium

Chapter Four: magnetic susceptibility and mechanical properties of uranium.

Chapter Five: Uranium Alloys.

Chapter Six: Uranium Enrichment technology.

Chapter Two

URANIUM NATURE

2.1 Introduction

This chapter deals with data on the physical, mechanical and chemical Properties of uranium, thorium and plutonium, their alloys and compounds, which are used or can be used as nuclear fuel, and gives concise information about the minerals and raw material sources of nuclear fuel, the principles of the technology of the raw material processing, and the production of the principal compounds, and of the pure metals and alloys. This chapter consider the basic requirements of nuclear fuel and give an account of methods of physic-mechanical and thermal treatment of fuel to obtain the maximum radiation stability and viability. They give comparative characteristics of different forms of nuclear fuel and evaluate their possibilities.

2.2 Discovery of Uranium and Its Position in the Periodic System

Uranium was discovered in 1789 by the German chemist Klaproth and named in honor of the planet Uranus, discovered five years earlier.

Klaproth attempted without success to obtain metallic uranium by reducing its oxides with carbon, prepared from sugar. Therefore he characterized it as a "submetallic" or "pseudo metallic" element, not having characteristic metallic properties.

It was not until 1841, half a century after the discovery of uranium that the French chemist Peligot succeeded in obtaining this element in a pure state by reducing uranium tetrachloride with metallic potassium. At the same time Peligot showed that in the reduction of uranium mixed oxide with carbon, as done by Klaproth, there is obtained only the lower oxide of uranium, i.e. its dioxide. In the reduction of uranium tetrachloride with potassium, however, a substance with characteristic metallic properties is obtained.

Nevertheless, complete proof of the metallic nature of uranium was not obtained until 1936, when the crystalline structure of uranium was determined scientifically by the X-ray method [1].

Peligot studied the compounds of uranium, assuming its atomic weight to be 120. He also discovered a method for purifying uranium compounds, by making use of the solubility of urinal nitrate in ethyl ether. Towards the end of the 1860's D. I. Mendeleev became interested in the study of uranium. He discovered Peligot's error in the determination of the atomic weight of uranium and showed that its atomic weight was 240.

D. I. Mendeleev placed uranium at the end of his periodic table in the group of chromium, molybdenum and tungsten. Proceeding from the fact that uranium was the last, heaviest element in the periodic table, the great Russian scientist later wrote: "I am certain that the investigation of uranium, starting from its natural resources, will lead to many new discoveries' confidently recommend anyone who is looking for new study topics to give particular consideration to the uranium compounds.

In our time this prediction of D. I. Mendeleev appears to be prophetic, if we take into account the significance which uranium has acquired. Owing to up-to-date knowledge of the composition of the periodic system of the elements the true position of uranium in it has now been determined.

As a result of investigations of the atomic spectrum of uranium vapors it has been shown that in the basic state it has the electronic structure According to

the number and constitution of the electron shells the uranium atom belongs not to group VI, but to the group of heavy actinide elements, similar to the lanthanide group [1].

However, there is no doubt of the chemical similarity of uranium to the elements of group 6a of the periodic system. Such a duplicity in the nature of uranium is explained by the close energies of the 5/ and *6d* electrons, which causes the possibility of passage of the 5/electrons to the *6d* subshell in processes of chemical interaction of the elements.

2.3 Use of Uranium

Three periods can be noted in the history of the use of uranium. In the first period, from the time of discovery to 1896, uranium was of interest only to a small circle of chemists and found almost no practical use, apart from a small-scale production of a yellow uranium pigment (sodium urinate),used in painting and in the manufacture of glass, enamels and varnishes.

The second period extended from 1894 to 1941. It started with the discovery by the French physicist Henri Becquerel of the radioactivity of uranium minerals, obtained from mines near Yakhimov (Czechoslovakia). The phenomenon of the radioactivity of uranium attracted the attention of physicists all over the world, and in 1898 in the analysis of Yakhimov Uranium ores and their treatment, Pierre and Marie Curie discovered the radioactive elements radium (Ra) and polonium (Po) accompanying natural uranium.

Radium, which has a high radioactivity, has been used medically for the treatment of some diseases. To produce radium, plants for processing uranium ores have been built in Czechoslovakia, U.S.A., Belgium (using Congo ores) and Canada. In the U.S.S.R. the production of radium from national uranium ores was organized in 1923.

In the period from 1906 to 1939 the total world extraction of radium amounted to some 1000 g and that of uranium to around 40001. Thus the second period in the history of the use of uranium is characterized by the extraction of radium from uranium ores. The uranium salts in radium extraction were by-products and did not find practical use.

Attempts to use uranium for alloying steels, made during World War 1, did not give positive results.

The third and present period in the use of uranium began in 1941, when investigations into utilizing the atomic energy of uranium for military purposes were commenced [3]. It is characterized by the great strategic importance which uranium and uranium deposits attained. The search for new uranium ores and deposits throughout the world in this period can even be compared with the "gold rush" era. A struggle for the possession and exploitation of uranium deposits in colonial and dependent countries developed between the chief capitalist countries.

After an extremely short historical period, lasting some ten years, a large uranium extraction and uranium processing industry had been built up in many countries. According to data in 1960 [4], the annual production of uranium (on conversion to U308) in the U.S.A. amounted to 15,0001, in Canada to 13,0001, in the Republic of South Africa to more than 60001, in the Congo Republic (in the capital, Leopoldville) to 10001, in Australia to 10001 and in France to 7501.

Table (2-1) Electronic Structure of the Uranium [2]

Shell	Subshell	Number of Electrons in shells and subshells		shell	Subshell	Number of electrons in shells and subshells
K, or 1	1s	2} 2		O, or 5	5s	2
L, or 2	2s	2 6 } 8			5p	6
	2p				5d	10 } 20 or 21
					5f	2 or 3
M, or 3	3s	2 6 10 } 18		P, or 6	6s	2
	3p				6p	6 } 9 or 10
	3d				6d	2 or 1
N, or 4	4s	2 6 10 14 } 32		Q, or 7	7s	2} 2
	4p					
	4d					
	4f					

In the period from 1943 to 1960 the U.S.A. invested more than 2 million dollars in building and reconstructing gaseous diffusion plants and extracted over 150,0001 U^{235} containing over 8001 U^{235} .

Scientists have devoted a great deal of research [5] to uranium. They have begun to use the new, powerful intrinsic force (atomic energy) for peaceful, as well as military purposes. Now atomic power is produced and nuclear engines developed for locomotive power and power plants; radioactive isotopes have become widely used.

Nuclear energy has now found practical use in naval ships. In 1960.

In many countries radioactive isotopes and nuclear radiation are widely used for peaceful purposes. The numerous radioactive isotopes obtained in nuclear reactors place in the hands of man new, extremely accurate instruments, permitting deeper study of processes taking place in living organisms, and of inorganic nature. Tagged atoms are used in science, industry, agriculture, farming, medicine and biology. Methods based on the penetrability of ionizing radiations, their damaging effect, release of energy, activation by radiation of other materials, etc., and have also been widely developed.

 Isotopes used in many research and medical establishments and by some industrial concerns.

However, isotopes will doubtless be used more widely in the near future.

2.4 Uranium as a Nuclear Fuel

Unlike other metals uranium is not used as a construction material but as

an extremely effective fuel or explosive substance. Some isotopes of uranium and plutonium are substances which are capable of fission or disintegration by the action of neutrons: $_{92}U^{235}$, $_{92}U^{233}$ and $_{94}Pu^{239}$. Of these three isotopes only $_{92}U^{235}$ occurs in nature, the other two are obtained in nuclear reactors by neutron bombardment of natural uranium or thorium. $_{9\,2}U^{238}$ is thereby partly converted to $_{94}Pu^{239}$ according to the reaction Therefore the isotope $_{9\,2}U^{235}$ is called a primary nuclear fuel and $_{94}Pu^{239}$ and $_{92}U^{233}$ are secondary nuclear fuels.

In the process of "combustion" or fission of the isotopes $_{92}U^{235}$, $_{92}U^{233}$ and $_{94}Pu^{239}$ a vast amount of energy is released. The calorific power of 1 kg U^{235} as fuel is approximately 50 million times greater than the calorific power of 1 kg coal.

The probability of nuclear fission reactions is usually expressed by the size of the effective cross-section σ, in area dimensions, since the probability of reaction between the fuel and a bombarding particle is proportional to the area of the transverse cross-section of the target nucleus.

The unit of cross-section is taken as $\sigma = 10^{-24}$ cm^2, which is called a barn. This value is approximately the same as the geometric cross-section of an atomic

nucleus, the radius of which is equal to 10^{-12}-10^{-13} cm. The value of a basically depends upon the nature of the bombarding particles and their energy [1].

2.5 Properties of Fissile Isotopes

For the majority of isotopes, the thermal neutron absorption cross-section varies approximately inversely proportional to the neutron velocity v.

In Table 2 there are shown the capture cross-section, absorption cross section and fission cross-section for three isotopes, capable of fission by thermal neutrons [6], In the fission process two groups of neutrons are formed: prompt neutrons and delayed neutrons [7]. The prompt neutrons amount to approximately 99% of the total quantity of fission neutrons and are emitted in a very short time interval (of the order of 10^{-14} sec). The delayed neutrons are emitted by fission fragments over a relatively large time interval after fission (on average from one to 55 sec).

Table (2.2). Thermal Neutron Properties of Fissile Fuels (v=2200m/sec)

Characteristic	$_{92}U^{235}$	$_{94}Pu^{239}$	$_{92}U^{233}$
Effective cross section, barn:			
fission σf	508	750	533
capture σ_c	107	315	52
absorption σ_a	687	1065	585
$\sigma = \sigma_c / \sigma_a$	0.184	0.420	0.098
Neutron yield:			
by one act of fission (v)	2.46	2.88	2.54
by one absorbed neutron (η)	2.08	2.03	2.31

The existence of around 1 % delayed neutrons gives the possibility of controlling the chain reaction in nuclear reactors. The energy of the fission neutrons lies in a wide interval; from 10 MeV (fast neutrons) to 0.025 eV (thermal neutrons). The speed of the fission neutrons varies accordingly from 1.955 x 107 to 2200 m/sec. U^{235} undergoes fission on capture both of slow (thermal) neutrons and of fast neutrons. U^{238} undergoes fission only on capture of fast neutrons with energy not less than 1.1 MeV. On capture of slow neutrons $_{92}U^{238}$ is converted to $_{94}Pu^{239}$, which according to its nuclear properties is close to $_{92}U^{235}$ and undergoes fission on capture of both fast and slow neutrons.

2.6 Critical Mass and Enrichment

The change in the amount of uranium, required for adjusting a chain reaction is a complicated function of different parameters. These parameters are determined by the following values:

1- enrichment (concentration) of fissile substances and the atoms surrounding them;
2- density and geometric form of the fuel;
3- the amount and nature of the additives contained in it[8].

Table (2-3). Minimum Critical Mass (Total) of Uranium for Different Degrees of Enrichment

U235 content %	Total critical mass of $U^{235} + U^{238}$, kg		
	Aqueous solution t(1)	**Unshielded metal** (2)	**Metal, shielded by water(3)**
90	.9	53	24.5
20	5.7	720	375
5	38.0	Infinite	Infinite
3	114	Infinite	Infinite
1.8	708	Infinite	Infinite

(1)**Reflector and moderator—water.**
(2)**Spherical form.**
(3)**Metallic spheres, shielded by water.**

2.7 Radioactivity and Isotopic Composition of Uranium

The U^{238} and U^{235} isotopes of uranium are parent radioactive elements.

The forerunners of two radioactive series, they are uranium and uranium- actinium. The mass numbers of the members of the uranium series are divisible by four with a remainder of 2, (An + 2 series) and the mass numbers of the members of the uranium-actinium series are divisible by four with a remainder of 3 (4/i + 3 series). Table 4 shows the relationship between the decomposition products in the uranium and uranium- actinium series in radioactive equilibrium [11].

The uranium family contains the stable element RaG, which represents the stable isotope of lead $_{82}Pb^{206}$. The most important members of this family are radium $_{88}Ra^{226}$ with a half-life period of 1590 years, radon Uranium as it occurs in nature

comprises two main isotopes: $_{92}U^{235}$ and $92U^{238}$ In the proportions 0.711% and 99.283%, respectively. The minor constituent $92U^{234}$ at a concentration of 0.006% is conventionally ignored in the enrichment business. Only $_{92}U^{235}$ is fissionable. Two commercial reactor systems, MAGNOX and CANDU, use natural uranium but more than 90% of the world's reactors require the concentration of the fissionable isotope $_{92}U^{235}$ to be increased to a level of 3 to 4%. This process is called uranium enrichment. Most of the reactors are of the light water cooled and moderated type: PWRs and BWRs [1,2].

The Uranium-238 Decay Chain

Atomic Number

| 82 | 83 | 84 | 85 | 86 | 87 | 88 | 89 | 90 | 91 | 92 |

Only main decays are shown
Gamma emitters are not indicated

| Th-234 24.1 d | α | U-238 4.5×10^9 a |

β | Pa-234 1.17 m

β

| Pb-214 26.8 m | α | Po-218 3.05 m | α | Rn-222 3.82 d | α | Ra-226 1600 a | α | Th-230 7.7×10^4 a | α | U-234 2.4×10^5 a |

β | Bi-214 19.9 m

β

| Pb-210 22.3 a | α | Po - 214 1.64×10^{-4} s |

β | Bi-210 5.0 d

β

| Pb-206 Stable | α | Po-210 138.4 d |

Element Names
U - uranium
Th - thorium
Ra - radium
Pa - protactinium
Rn - radon
Po - polonium
Bi - bismuth
Pb - lead

Half-life units
a - years
d - days
h - hours
m - minutes
s - seconds

Figure (2.1) the uranium-238 decay (a)
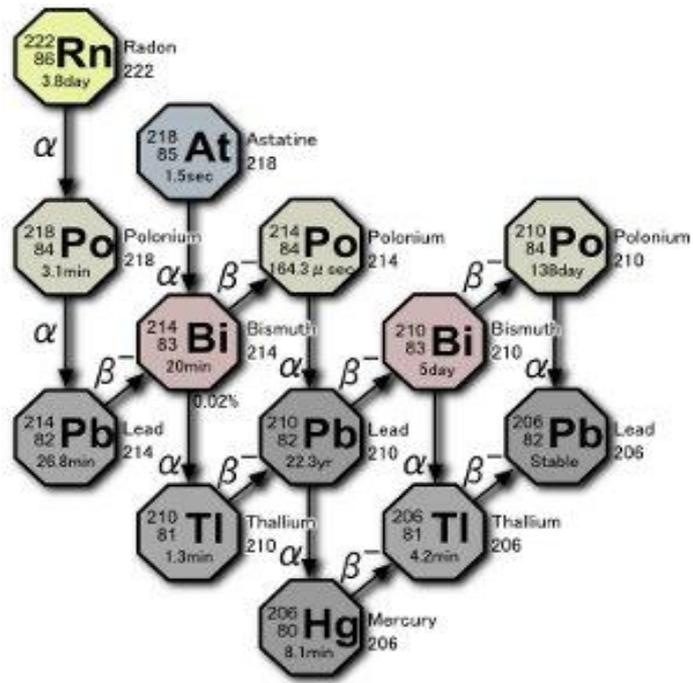
Figure (2.2) radon decay
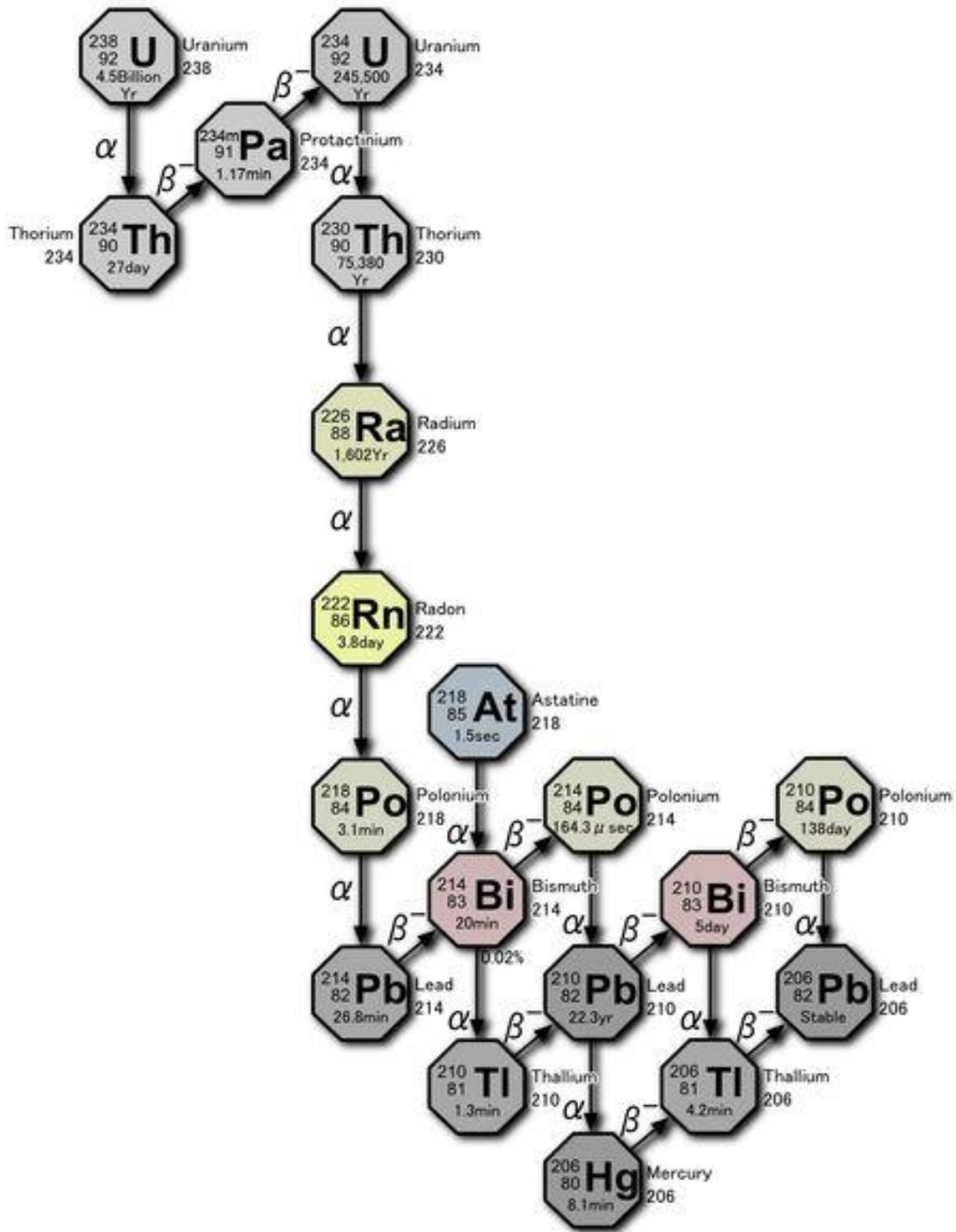
Figure (2.3) the uranium-238 decay (b).

Chapter Three

PHYSICAL PROPERTIES OF URANIUM

3.1 Introduction

There are many characteristics enjoyed by the element of uranium in nature and these characteristics apply to all natural and artificial uranium isotopes. The holiday of these characteristics can be discussed in this chapter from the point of view of the process of separation of isotopes by physical methods.

3.2 Physical Properties of Uranium

Uranium is a very heavy metal with a density of 19.05 g/cm^3. The freshly polished surface of metallic uranium is a silvery color, but after a few hours in air it grows dull and it first becomes golden with bluish tints, and then turns dark. For a long time uranium was thought to be an extremely high-melting metal, with a melting point of some 1850°C [9]. Later it was determined that the temperature shown related to an oxide film on the metal and that pure uranium (with an impurity content not exceeding 1.10^{-2} wt. %) has a melting point of 1130 ± 1°C [10].

As with other metals, some physical properties of uranium are almost independent of its purity, microstructure and method of manufacture.
Among these properties are: density, specific heat and thermal conductivity, latent heat of fusion and of vaporization, enthalpy and entropy, vapour pressure, phase transformation temperature and magnetic susceptibility.
Other physical properties of uranium, such as the coefficient of thermal expansion, the modulus of elasticity, mechanical and electrical properties, depend to a great extent on the texture of the uranium and on its composition.

3.3 Crystalline Structure of Uranium

In phase-transformation investigations it was determined that uranium has three allotropic modifications, the crystalline models of which are shown in Figure (3.1) in the [010] direction.
The covalent nature of the bonds in α-U influences many of its physical properties. Owing to its structure α-U has a strongly anisotropic nature.
The tetragonal modification, β-U, stable from 662 to 769°C has a complex lattice with 30 atoms per unit cell. The structure of β-uranium consists of layers which are perpendicular in the 001 direction. The projection of the layers into the (001) plane.
7bThe layers are at distances of$\frac{1}{2}$, $\frac{1}{2}$, and $\frac{3}{4}$, °C.

Table (3.1). Crystalline Data of Uranium Modifications

Phases	Elementary nucleus			Theoretical density g/cm2	Remark
	Type of lattice	Parameters A	No. of atoms in nucleus		
α-U, stable up to 662°C	Orthorhombic	*a* = 2-852 *b* = 5-865 *c* = 4-915	4	19.12	
β-U, stable from 662 to 769°C	Tetragonal	*a* = 10-759 *c* = 5-656 *a* = 3-524	30	18.11	at 720°C
γ-U, stable from 769to1130°C	Body-centered cubic		2	18.06	at 805°C

The β-U modification is anisotropic, although not to the same extent as α U.

The cubic body-center γ-U modification, which is stable from 769°C to melting point, differs from the two previous crystalline forms which are typical of metals, and has isotropic properties [1, 8].
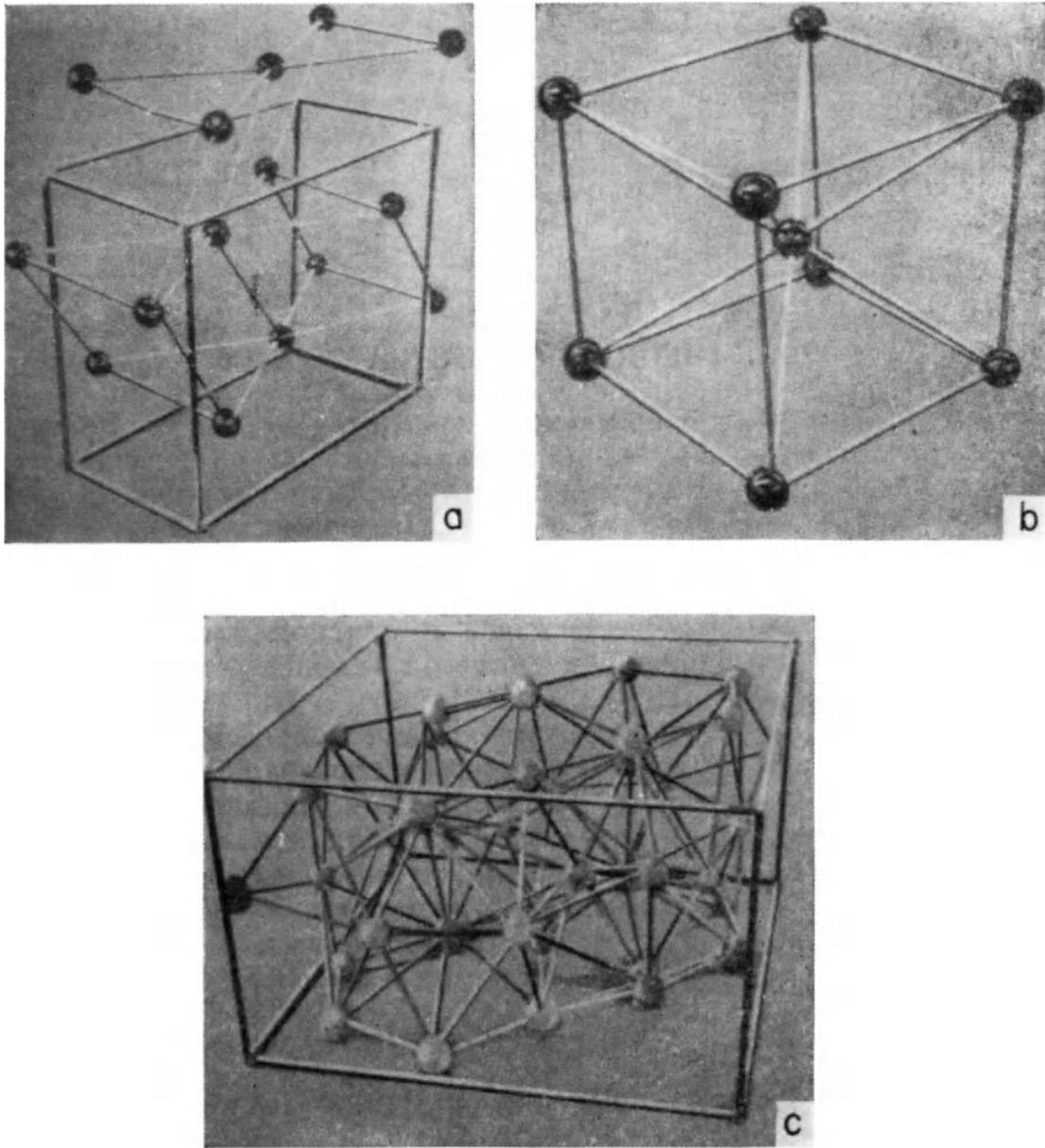
Figure (3.1). Models of the crystalline modifications of uranium.(a) α- phase; (b) γ-phase; (c) β-phase

The crystalline data of these modifications are given in Table 6.

The orthorhombic structure of α-U, similar to the structure of arsenic and other non-typical metals, is stable up to 662°C (Fig. 7a). It can be represented in the form of "corrugated" atomic layers in parallel (010) planes. The distance between the atoms of the inner layer is less than the distance between the different layers (2·8 and 3·2 Å respectively).

Since the bond strength increases with decrease in the distance between the atoms, the bond strength of the atoms in the corrugated planes is greater than between the planes. Projection of the atoms into the (001) plane

(a)

(b)
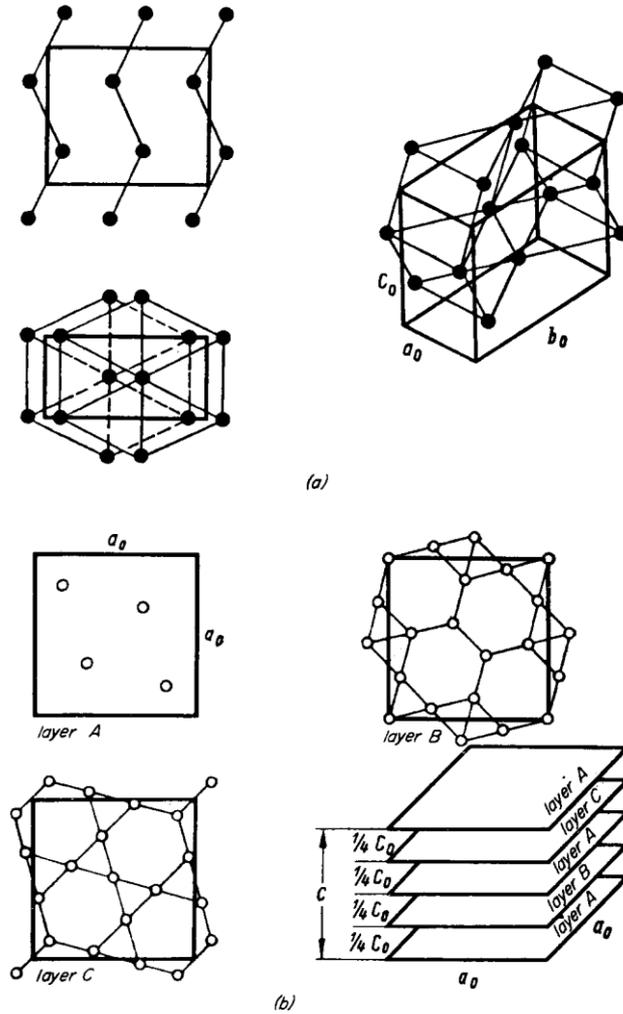
FIG. 7. Diagrams of the crystal lattice of the α-phase (a) and β-phase (b) of uranium.

suggests a hexagonal arrangement, with the sole difference that the layers of atoms, parallel to the basic plane are displaced forwards and backwards

3.4 Density of Uranium

The density of uranium depends on temperature and the crystalline modification in which it occurs. The calculated density and atomic volume of uranium in the different modifications according to the radiographic parameters, determined at normal temperatures.

The experimental values of the density of α-uranium can differ substantially from the density calculated according to the X-ray data. These values depend on the admixtures and porosity of the uranium and usually vary from 18.7 to 19.0 g/cm^3. The density of liquid uranium at its melting point is taken to be equal to 16.63 g/cm^3.

3.5 Thermodynamic Properties of Uranium
3.5.1 Specific Heat of Uranium

At 18°C the specific heat of uranium (0.028 cal/g-atom/degree) is about a third of that of copper (0.092 cal/g-atom/degree) and a quarter of that of iron (0·112 cal/g-atom/degree). The values of the specific heat of uranium at constant pressure (C_p) and at constant volume (C_v) at higher temperatures for determination of the specific heat of α-U at different temperatures [4,10,12]the following equation was used:

$$C_n = 3.15 + 8.44 \text{ x } 10^{-3} \, T + 0.80 \text{ x } 10^{-5} T^2 \tag{3.1}$$

3.6 Boiling Point and Vapour Pressure of Uranium

The boiling point of uranium has not been determined experimentally. Its value (3813°C) was found by extrapolation of the vapour pressure curve the vapour pressure of uranium at temperatures below 1500°C is practically insignificant and its determination is extremely difficult. The experimental data were obtained for the most part in the temperature interval from 1500 to 2000°C. At temperatures above 2000°C the determination of the vapour pressure of uranium with sufficient accuracy is extremely complicated.

In the following equation (for the temperature interval 1630 to 1970°K) is given for calculating the vapour pressure of uranium

$$lgp = -(\frac{23300}{T}) + 8.583 \tag{3.2}$$

Where p = vapor pressure, mm Hg; T = temperature.

3.7 Thermal conductivity of uranium

Thermal conductivity is one of most important physical properties of the uranium, since a nuclear fuel during work in reactor must transmit considerable amount of heat to the cooling medium. The specific power in cylindical fuel elements is limited to some extent by the high thermal stresses, which develop owing to the drop in temperature between the wall and the center of the core.

The drop in temperature in the core in its turn decreases with increase in the coefficient of thermal conductivity. Therefore the lower the thermal conductivity of the material, the greater are the internal stresses

Table (3.2) shows experimental values of the thermal conductivity measured over a temperature interval from -271.2 to +900°C, according to data in the same table and over the same temperature interval there are shown the results of experimental measurements of the electrical resistance and thermo-e.m.f. relative to platinum and values of the magnetic susceptibility.

The thermal conductivity of y-quenched cast uranium and that of uranium, sintered from powder of density 18.7 g/cm^3, are practically the same.

However, uranium cermet of density 18.0 and 18.3 g/cm^3 has a thermal conductivity which is comparatively lower than the other samples. This is explained by its porosity and by the oxidation of the uranium particles up to sintering [7].

Table (3.2) Thermal Conductivity, Electrical Resistance, Thermo-e.m.f. (relative to platinum) and Paramagnetic Susceptibility as a Function of temperature

U phase	Temperature		Electrical resistance Micro hms/cm	Thermal conductivity Mw/ cm² · sec · degree	Thermo-e.m.f. Relative to Pt. Mv	Paramagnetic suspectibility x 10^6 e.m.v./g
	c°	k				
α	< -271. 8	< 1. 3	Superconductor	—	—	–
	-271.2	2	Superconductor	30	—	-
	-173	100	13	220	—	—
	-73	200	23	260	—	—
	0	273	30	270	0-0	—
	27	300	39.5	270	—	1.66
	100	373	45	290	1.2	1.68
	200	473	50	310	2.8	1.71
β	300	573	55	330	5.0	1.74
	400	673	57-5	350	7.4	1.77
γ	500	773	59	380	10.5	1.81
	600	873	-	390	13.8	1.85
	650	923	59+	391	—	1.88
	666	939	55	391	15.6	1.92
	666	939	55.5	400	15.6	2.05
	700	973	56	410	17.5	2.05
	750	1023	53.5	416	19.4	2.05
	776	1049	54	416	19.7	2.05
	776	1049		423	19.7	2.10
	800	1073		434	21.4	2.10
	850	1123		446	23	2.11
	900	1173				2.12

3.8 Heats of Transformation of Uranium

The most reliable data on the heats of transformation of uranium are shown in Table (3.3).

Table (3.3). *Heats of Transformation of Uranium* [2]

Transformation	Heat of transformation	
	cal/g	kcal/mol
A $\longrightarrow$ β	2.86-2.99	0.68-0.71
β $\longrightarrow$ γ	4.80-4.89	1.14-1.17
Fusion	19.8-20.0	4.72-4.76
Vaporization	450	107
Sublimation	490	117

3.9 Elastic Properties of Uranium

Polycrystalline uranium differs from monocrystalline uranium inasmuch as it does not give a clear region of elastic deformation in the stress-strain curve. This is explained by the fact that polycrystalline uranium has considerable internal stresses, produced in the process of cooling the metal in the α-phase to the temperature of the ambient medium. If these stresses exceed the elastic limit of the metal, then for polycrystalline samples the elastic limit will generally not appear on the stress-strain curve. For these reasons the modulus of elasticity of uranium cannot be determined with sufficient accuracy from the results of tensile tests. It is usually determined by measuring the speed of sound in this metal [13].

3.10 Electrical Resistance of Uranium

Uranium is a poor conductor of electricity. Its electrical resistance is 17 times greater than that of copper and 11 times greater than that of aluminum.

It depends to a great extent on the crystallographic direction of the uranium grains, and hence on the texture produced during working of the metal. Uranium rolled at 300°C and annealed for 2 hours at 575°C has a specific electrical resistance of 26.4 microhms/cm. If its structure textured in such a way is broken by thermal treatment, then the specific electrical resistance of uranium increases to 30.0 microhms/cm . The specific electrical resistance of uranium in the principal crystallographic directions at temperatures of 273 and 4.2 K [13, 14].

Chapter Four
MAGNETIC SUSCEPTIBILITY AND MECHANICAL PROPERTIES OF URANIUM

4.1 Introduction

In nuclear reactors uranium elements are subject to complex stress conditions for a long time at high temperatures. Therefore the mechanical properties of uranium, such as tensile strength, hardness, brittleness, impact strength and creep resistance, under prolonged load are also very important characteristics for the construction and manufacture of metallic fuel elements.

4.2 Magnetic of Uranium

Uranium is a slightly paramagnetic metal. shown the magnetic susceptibility of uranium as a function of temperatur the magnetic susceptibility changes intermittently during the phase transformations .It was found that the magnetic susceptibility of uranium with a content of 0.07% iron and 0.006% nickel) at 20°C is equal to 1.74x 10^6 e.m.v./g. It was determined that uranium forms non-ferromagnetic alloys with many ferromagnetic elements. Its alloys with iron (5 and 10% show no signs of ferromagnetism) [15].

4.3 Mechanical Properties of Uranium

4.3.1 General Characteristics

The change in the mechanical strength of fuel elements under the action of corrosion, thermal stresses and radiation determines the working safety and economy of a reactor. Three main problems arise, which have not yet been completely solved and always occur with the use of metallic fuel elements.

These problems are:

1. The unusually large increase in creep of irradiated uranium even under negligible load.
2. Embrittlement of irradiated uranium, not relieved practically by annealing and.
3. the irradiation growth of uranium, leading to change in shape of the fuel element in the working process of the reactor and governing the above-mentioned creep and embrittleme In comparison with construction metals uranium has poor mechanical properties. It is comparatively difficult to machine and the texture, which in its turn is determined by the manufacturing technology of the elements and by the thermal treatment system, has a considerable influence on its mechanical properties. As with other metals, slight variations in the content of non-metallic admixtures in uranium can also affect its mechanical properties.

The chief characteristic property of uranium is its pronounced tendency to cold-hardening, which makes machine cutting difficult. In this respect uranium resembles austenitic steel with 13wt.% manganese, although at low temperatures it is considerably less ductile.

The second characteristic of uranium is the great dependence of the majority of its mechanical properties, particularly its ductility, on temperature.

In water-cooled reactors, uranium works at temperatures from 0 to 250°C.

There a knowledge of how the mechanical properties of uranium change in this temperature interval is very important, although it is also necessary to know them at higher temperatures.

The mechanical properties of annealed, coarse-grained uranium can be improved by heat treatment. The principal form of heat treatment is quenching, which is carried out by: heating the. Metal to the *irs-* and y-phase temperatures, holding it at these temperatures and then rapidly cooling it in water or oil.

Quenching of uranium of normal purity does not lead to fixation of the

γ - or β-phase, but causes a more or less sharp change in the macro and microstructure [16] and break-up of the grain. The chemical composition of the metal and its admixture content, primarily of iron, silicon, nickel and aluminum, have an effect on the extent of break-up of the grain. As a result of quenching uranium at the β- and γ -phase temperatures both its long-term and short-term characteristics in the temperature interval from 0 to 400°C are improved. In this respect the rate of cooling has a considerable influence on the properties of quenched uranium. Thus uranium with ultimate tensile strength $\sigma_B = 40$ kg/mm2, after quenching in water at 900°C acquires a strength (σ_B) of more than 60 kg/mm^2, one of approximately 55 kg/mm^2 after quenching in oil, and after quenching in argon its ultimate tensile strength remains the same as before quenching The mechanical properties of uranium can also be improved as a result of plastic deformation with subsequent recrystallization annealing at the α- phase temperature. Fine-grained uranium, prepared by a deformation and recrystallization method has a considerably higher tensile strength, with the same ductility, than the hot-rolled metal. Thus, the hot-rolled metal has an ultimate tensile strength of $\sigma_B \approx 40$ kg/mm2, a yield point of $\sigma To.2 \approx 25$ kg/mm2 and an elongation of $\delta \approx 5\%$, whilst deformed and recrystallized uranium has an ultimate tensile strength and yield point which are twice as high$(\sigma_B \approx 80$ kg/mm2 and $\sigma_{To.2} \approx 60$ kg/mm^2) with the same elongation

$(\delta \approx 5\%)$[12].

4.3.2 Tensile Properties of Uranium

the tensile properties of uranium depend to a certain extent on temperature in the interval from negative temperatures to + 850°C. With increase in temperature the

ultimate tensile strength (σB) of uranium drops from 60 kg/mm2 at 20°C to 8 kg/mm2 at 662°C [16].

4.3.3 Tensile Strength of Uranium in Endurance Tests

In uranium endurance tests it was found that its creep resistance, i.e. deformation capacity under the action of stress, does not exceed the yield strength at the test temperature. The creep resistance of uranium, as in short-term tests, depends on the texture of the metal and on its admixture content. in the creep of cast and hot-rolled uranium was investigated in the temperature interval from 20 to 600°C with different loads. The test period was 700-1000 hr at high temperatures and 6000 hr at room temperature.

The investigation of creep at high temperatures was carried out in an atmosphere of pure helium

4.3.4 Hardness of Uranium

The hardness of uranium depends only slightly on its method of treatment, but is reduced extremely strongly with increase in temperature [14].

4.3.5 Impact Strength of uranium

The impact strength of uranium is an extremely important mechanical property. The impact strength, i.e. sensitivity to concentrated stress, is given by the notch impact bend test of the sample. In other words, it determines the capacity of the metal for rapid deformation, particularly with the presence of notches, the capacity of sharp transitions of cross-section from thin to thick [17].

4.4 Effect of Mechanical and Thermal Treatment of on the Physico-Mechanical Properties of Uranium

THE properties of uranium during deformation have been intensively studied for some time past in connection with the necessity of increasing its stability during irradiation. The problem of increasing the stability of uranium and the life of fuel elements at high temperatures under irradiation in a reactor also has great practical significance. It will determine to a considerable extent the cost of the electrical energy of an atomic power station and the cost of producing plutonium. As already indicated, as a result of investigations it has been found that the properties of uranium can be considerably improved by suitable treatment. By the same means it is possible to increase the stability of uranium and

the life of the fuel elements.

4.4.1 Reaction of Uranium with Gases (O_2, N_2 AND H_2)

It has been found that at low partial pressures of oxygen (less than 10^4 atm) a film of the monoxide UO is formed on uranium metal; in air at temperatures below 100°C a film of the dioxide UO^2 is formed and in the temperature interval from

100-700°C there is formed on uranium a film of mixed composition consisting of UO_2 and U_3Os. At a temperature of around 1500°C, UO_2 slowly reacts with liquid uranium, forming the monoxide UO. Inclusions of UO_2 and UO are readily detected metallographic ally in commercial-grade uranium in very small contents.

$$3U + 4O_2 \xrightarrow[77]{>700°C} U_3O_3 + Q \tag{4.1}$$

$$U + \frac{3}{2}H_2 \xrightarrow{>225°C} UH_3 + Q_1 \tag{4.2}$$

$$U + \frac{3}{2}D_2 \xrightarrow{>250°C} UD_3 + Q_2 \tag{4.3}$$

$$U + \frac{3}{2}T_2 \xrightarrow{>225°C} UT_3 + Q_3 \tag{4.4}$$
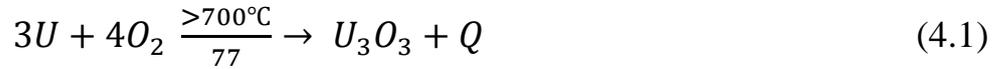
Where $Q_1 \approx Q_2 \approx Q_3 \approx$ 30.3 kcal/mol

4.4.2 Reaction of Uranium with Water and steam

In a damp atmosphere the oxidation of uranium metal takes place more rapidly than in air, and in water uranium has a very low corrosion resistance. The logarithm of the rate of corrosion of unalloyed uranium in hydrogen saturated water varies linearly with the inverse value of the temperature in the range from 50-225°C, whereby in boiling water the rate of corrosion amounts to 2-7 mg/cm²/hr [13]. The reaction of uranium with boiling water takes place according to the equation:

$$U + 2H_2O \xrightarrow{>100°C} UO_2 + 4H \tag{4.5}$$

Whereby the atomic hydrogen given off accelerates the corrosion of the uranium owing to the formation of hydride. In aerated water the rate of corrosion of uranium is at   first lower than in hydrogen-saturated water.   protective oxide film. However, the rate of reaction   then   increases and approaches the value obtained in hydrogen-saturated water, as can be seen from the curves . Steam reacts with uranium at a temperature of 150-250°C. It is assumed   that here the following reaction takes place:

$$7U + 6H_2O_{steam} \xrightarrow{150-250°C} 3UO_2 + 4UH_3 \tag{4.6}$$
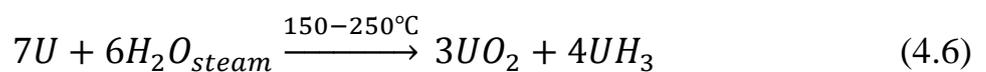
Chapter Five

URANIUM ALLOYS

5.1 Introduction

The possibility of using pure uranium for manufacturing metallic fuel elements is limited to a great extent by the properties of this metal described previously. Heat-treatment or thermo-mechanical treatment of uranium, permitting a more stable, fine-grained and quasi-isotropic structure to be obtained, does not completely solve the problem of the stability of metallic nuclear fuel. The use of alloys of uranium with different elements appears to be a better solution to the problem of increasing the stability of the fuel in metallic fuel elements.

5.2 General Characteristics and Requirements of Uranium Alloys

To these alloys there should appertain in the first place the binary alloys of uranium with molybdenum, aluminum, zirconium, niobium, chromium, silicon and a few other metals. Alloys of the systems uranium-zirconium-molybdenum, uranium-zirconium-niobium, uranium niobium- molybdenum and others appertain to the ternary alloys.

The uranium alloys capable of being used at present for nuclear fuel are divided into two groups: alloys with α-phase structure, in which a small amount of alloying admixture contributes to obtaining an untextured fine-grained metal, and alloys with a γ -phase structure, in which the alloying component is introduced in a quantity sufficient for partial or complete stabilization of the cubic γ -phase.

Alloys in the first group based on natural or slightly enriched uranium are used chiefly in thermal neutron reactors. As examples of these alloys there can serve alloys of uranium with 0.4 wt. % aluminum, 0.5-2.0 wt. % molybdenum, 2.0 wt. % zirconium, or 0.1 wt. % chromium. Alloys of the second group can be more generally used, but because of the high content of alloying admixture they require considerable enrichment. As examples of alloys of this type there can serve alloys of the systems uranium-molybdenum, uranium-zirconium, uranium-niobium and ternary alloys of the system uranium-zirconium-niobium with a considerable quantity of alloying components.

Uranium-thorium alloys are of great importance for constructing reactors with reproduction of nuclear fuel. The rational use of these alloys in many respects determines the prospective development of nuclear power.

Extremely varied requirements are made of uranium alloys. In the first place they must ensure minimum parasitic neutron capture, which permits the U^{235} charge in the reactor to be reduced. To satisfy this requirement it is necessary to use alloying elements with minimum neutron capture cross-section. The preservation of the

shape and dimensions of the fuel element cores during work in the reactor is a no less important requirement.

Change in the shape and dimensions of uranium cores during work leads to disturbance of the spatial arrangement of the fuel elements, which has a marked effect on the thermal system of the reactor and on the removal of heat. These changes can cause non-uniform distribution of the coolant flow and, as a result of this, local overheating and rupture of the protective jacket.

Uranium alloys must not only remain stable, but must also have high mechanical strength and ductility. High thermal currents lead to the formation of steep temperature gradients in the nuclear fuel, which cause internal stresses. As shown by calculation, the value of the maximum thermal stresses is determined not only by the geometry and magnitude of the heat flow per unit fuel surface, but also by the characteristics of the fuel such as the modulus of elasticity, Poisson's coefficient, the coefficient of thermal expansion and the thermal conductivity. The thermal stresses can be so high that with poor mechanical properties of the fuel either considerable plastic deformation occurs, or, in the case of low ductility of the material, cracks are formed. In this case and in other cases the normal working of the fuel elements becomes disturbed.

 Among the most important properties of a fuel there should be considered the ultimate tensile strength, the yield strength and the ductility, hence the relative elongation, the reduction in area of the transverse cross-section, as also the impact strength and the creep resistance. Together with these, an alloy used as nuclear fuel must have a high corrosion and erosion resistance with respect to the coolant. Under conditions of continuous working of the reactor damage is possible which can lead to rupture of the protective jackets of the fuel elements. In this case, with poor corrosion resistance the fuel and the fission products can be very rapidly washed out by the coolant, which leads to accumulation of these on the surface, in the spaces and communication channels of the primary contours and causes considerable difficulties in operation of the reactor [18].

In choosing an alloy as a nuclear fuel special attention should also be paid to its compatibility with the protective jacket material at the working temperatures. Here the possibility of a diffusion interaction between them must be excluded, otherwise damage, leading to break-up of the fuel elements, can occur. Finally one of the principal requirements made of alloys as to nuclear fuel is high workability.

The technological properties of alloys must ensure the possibility of an economically favorable mass production of fuel elements of the requisite shape and dimensions. Phase diagrams of uranium with almost all elements have now been published [10, 1]. On the basis of these diagrams the influence of the chemical

series of the alloying additives on the structure and properties of uranium alloys has been predicted and studied.

However, of a great number of alloys investigated only a few, and then only partially, satisfy the above-mentioned requirements. Among such alloys, as mentioned earlier, are the binary alloys of uranium with molybdenum, aluminum, zirconium, niobium and a few other elements.

5.3 Methods of Purifying Uranium ore Concentrates

5.3.1 Choice of the Method of Purification

The high requirements made of the purity of uranium metal cause great difficulties in the production technology of this metal. This technology is therefore linked with numerous difficulties and differs considerably from the production technologies of other metals. In addition to the stages of ore concentration, obtaining technical uranium compounds and reducing these to the metal, it includes a stage of fine purification of uranium compounds before their reduction. On the whole, as a result of preliminary purification, uranium is separated from more than 40 of the elements which accompany it in the ores. In the reduction process, however, not only is purification of the metal not achieved but it becomes contaminated as a result of the occurrence of admixtures from the crucibles and materials used as reduction agents and linings. Another feature of the production technology of uranium depends on its high chemical activity, leading to contamination of the metal by admixtures from the ambient medium, which causes the necessity of carrying out the individual operations in vacuum or in an inert atmosphere, of using chemically pure reagents, etc.

Thus the quality of uranium metal is almost completely determined by the efficiency of the chemical purification of the starting materials from which the metal is obtained.

5.3.2 Precipitation Methods:

1. Purification of Uranium by Crystallization of Uranyl Nitrate
2. Method of Carbonate Purification
3. Oxalate Purification Method
4. Peroxide Method of Purification
5. Extraction of Uranium by Tribute Phosphate

5.4 Industrial Methods of Obtaining UO_2, UF_4 AND UF_6

URANIUM dioxide, tetra fluoride and hexafluoride are important intermediate products in basic uranium production. They are also of independent value as nuclear fuel for certain types of reactors. The chemistry of the processes of obtaining uranium dioxide and uranium fluorides. Here we will discuss briefly the fundamentals of the industrial processes for obtaining the said products and the typical equipment used for these purposes

5.4.1 Production of UO₂

The chief intermediate product for the production of UO_2 is uranium trioxide which, can be obtained by different methods. In uranium works the most widespread production of UO_2 is from an aqueous solution of urinal nitrate by evaporation with subsequent calcimine of the nitrate and reduction of the UO_3 to UO_2 by hydrogen. Here the process of producing UO_2 consists of three basic stages:

1. Evaporation of the uranic nitrate solution to a concentration corresponding to the composition UO_2 $(NO_3)2 \cdot 6H20$.
2. Claiming of this salt to UO_3.
3. Reduction of the UO_3 to UO_2 by hydrogen.

As a rule, the uranic nitrate solution under industrial conditions contains from 90 to 280 g/1. uranium The content of free nitric acid in it varies from 0.01to0.05N.

The solution is evaporated at 120-140°C in vertical tubular condensers or in steam-heated boilers. The condensers are made of stainless steel.

Thermal decomposition of the uranyl nitrate takes place in accordance with the $UO_2(NO_3)2$—H20 phase diagram . Uranyl nitrate hexahydrate and trihydrate have incongruent melting points of 60 and 113°C respectively. The melting point of the dihydrate is 184°C. Above this temperature, decomposition of uranyl nitrate to UO_2 and oxides of nitrogen takes place. Thermal decomposition of uranyl nitrate in vacuo takes place considerably more rapidly than at atmospheric pressure, but vacuum denitration in the absence of a liquid phase gives a product in the form of amorphous UO_3.

Denitration is carried out in batch and continuous units. Batch units are stainless steel boilers of capacity from 250 to 12001, inside which mixers rotate at a speed of 36 rpm. The temperature conditions are such that the product obtained will satisfy the technical requirements.

In continuous denitration processes a stainless steel apparatus in the form of a tube heated in an electric furnace is used. A horizontal mixer-worm, which continuously mixes and transports the material, passes through inside the apparatus.

The continuous denitration process can also be accomplished in a fluidised bed in a vertical stainless steel tube [20]. The uranyl nitrate solution is fed into the apparatus at the side under pressure and is atomised by a jet in the centre part of the tube. Hot air is supplied under pressure through a porous diaphragm placed at the base of the apparatus. The drops of atomised solution are converted to rapidly

falling UO_3 particles, which settle at the bottom of the apparatus and are then discharged.

The gaseous reaction products are directed to the top of the apparatus through a series of porous metallic filters.

At the Mallinckrodt uranium works in the U.S.A. a semi-industrial apparatus for denitration on the fluidised bed principle with an output of 750 kg UO_3 with a surface of 1 m2 per hour was started up in 1957 The quality of the UO_3 powder obtained in different The reduction of UO_3 to UO_2 by hydrogen is also accomplished in batch or continuous units. The most widespread construction of apparatus for batch reduction is a horizontal stainless tube [20]. In the reactor there are racks on which small boats filled with UO_3 powder are placed. A detachable square flange, which has two nozzles, one for supplying hydrogen, the other for leading-out excess hydrogen and for its combustion, is hermetically fitted to the front end of the reactor. The assembled reactor is transferred to an electric furnace, which automatically keeps the temperature of the reduction reaction at 820°C. The reduction lasts some 5 hr, after which the reactor is withdrawn from the furnace and cooled at first by compressed air and then by water. Cooling of the UO_2 takes place under a current of hydrogen. The consumption of hydrogen is higher than that required theoretically by a factor of about 1-5.

Reduction of the UO_3 can be carried out in horizontal tubular furnaces, as in tungsten and molybdenum metallurgy.

Three types of reactors have been developed for the continuous U03reduction process:

1. Moving layer reactor.
2. Fluidized bed reactor.
3. Reactor with material transport by vibration.
4. In the moving layer reactor transport of the U03 is accomplished by a helical worm through a heated tube to meet the flow of hydrogen. The reactor tube is made of cast stainless steel. The working life of these materials in normal running amounts to 6 years.

The electric furnace heating the tube has four heating zones, the temperatures of which are regulated by thermocouples. Hydrogen or dissociated ammonia, fed in at the discharge end of the tube, is used as reducing agent.

The excess hydrogen is burnt at the charging end of the tube [20].

The uranium trioxide for reduction is in drums or in movable bins up to 51 in weight. The drums automatically move towards the bin for unloading. The UO_3 enters the worm feeder through a conical crusher from the bin.

If the UO_3 is in a movable bin, the latter is hoisted by a crane onto a platform and connected with the reaction tube feeder. In UO_3 reduction in a moving layer the

temperature of reduction, rate of feed of the starting product and rate of hydrogen throughput are of great significance.

The reaction temperature of UO_3 reduction by hydrogen has an effect on the chemical activity of the UO_2 obtained, which shows in its subsequent fluorination. The chemical activity of the original UO_3 in turn has an effect on the choice of reduction temperature. With sufficient UO_3 activity the rate of reduction is increased, as a result of which a considerable amount of heat is given off in a short time owing to the heat of reaction.

In this case it is necessary to set a reduction reaction temperature such that the overall temperature in the reactor would not be higher than the optimum temperature. At a temperature below the optimum temperature the rate of the reduction reaction drops and incomplete UO_3 reduction becomes possible. An increased reduction reaction temperature causes sintering of the product and grain coarsening. The optimum UO_3 reduction temperature to obtain uranium dioxide intended for fluorination is calculated as 650°C in all zones of the reactor. The rate of supply of UO_3 by the worm feeder amounts to some 250 kg/hr at 650°C, which corresponds to a time of stay of the product in the reaction tube of up to 1-5 hr. The rate of mixing of the UO_3 in the reactor does not have a substantial effect on the time of reduction. It is determined by the rate of rotation of the helical worm conveyer, which is equal to some 4 rpm.

The consumption of hydrogen in the reduction in practice is higher than that necessary for stoichiometry by a factor of 1-5. The apparatus for reducing UO_3 on the fluidized bed principle [20] consists of two vertical stainless steel tubes of diameter 350 mm set up in series.

The partly reduced product from the first tube passes into the second tube where it is reduced to UO_2 With a tube height of some 2000 mm the depth of the fluidized bed is kept at 1350 mm. Reduction in this case is by dissociated ammonia, which enters the reactor under pressure from a gas conduit through porous diaphragms, fitted in each of the two tubes.

The excess gas together with the product passes through a discharge pipe into a UO_2 receiving bin and is then burnt off. At the side of the first tube a worm feeder, which in turn is connected to a movable bin containing the initial UO_3, is fitted. The rate of supply of UO_3 for the reduction varies from 275 to 600 kg/hr depending upon the activity of the powder. The rate of flow of the gas correspondingly varies from 0-24 to 0-45 m/sec with a fluidized bed temperature of 540 to 620°C. Each reactor tube has a heating and cooling system. Heating of the tubes is produced by electrical resistance furnaces, cooling by a coil wound on the outside of each tube. The total uranium content in the dioxide obtained is 87·9 %. The unit for UO_3

reduction according to the principle of vibration transport of the reduction product is a muffle pipe of rectangular cross-section 150 x 600 mm, length 4500 mm.

The unit is made of 12 millimeter cast Inconel or 347 bar stainless steel.

The reduction reaction temperature depends on the physical properties of the UO_3 and on the reduction conditions: rate of feed of UO_3, amplitude and frequency of vibration, on which the rate of transport of the product depends.

In reduction of commercial-grade UO_3 the walls of the reactor are kept at a temperature of 590-650°C. The degree of reduction of the UO_3 amounts to 97 %. Incompletely reduced UO_3 on fluorination forms uranyl fluoride, which is an undesirable admixture in uranium tetra fluoride intended for the preparation of uranium metal.

The vibration reactor for UO_3 reduction, of the construction described, has a UO_2 output of around 150 kg/hr. New types of vibration reactors permit up to 250 kg UO_2 to be obtained per hour.

Uranium dioxide with low chemical activity for ceramic fuel elements is obtained by reducing UO_3 at 800-900°C. The UO_2 obtained under these conditions is more resistant to oxidation [1,4,8].

5.5.2 Production of UF$_4$

Uranium tetra fluoride can be prepared by different methods. One of the basic industrial methods of obtaining UF_4 is the fluorination of uranium dioxide by gaseous hydrogen fluoride, which is called hydro fluorination [20]. This method ensures the greatest yield of uranium from the dioxide in the dry fluoride, since it eliminates uranium losses in mother liquors and insoluble residues as also mechanical losses on drying and calcimine. According to its physical properties, the UF_4 obtained by the method of gaseous fluorination is most suitable for reduction to the metal. It contains the least H_{20}, UO_2 and UO_2F_2 in comparison with the products obtained by other methods.

5.4.3 Production of UF$_6$

The production of UF_6 at first took place in batch units [30]. then a continuous process was developed which is now used in the "PA duke", "Union Carbide Nuclear Co" and "DuPont Carbide Corporation" plants [21]. The continuous process of producing UF_6 consists of three stages:

1. Reduction of UO_3 by hydrogen to UO_2 in fluidized bed units.
2. Fluorination of $U02$ to UF4 by dry hydrogen fluoride in reactors with worm mixing.
3. Further fluorination of UF_4 to UF_6 by elementary fluorine in units, one of which.

The apparatus for fluorination is a monel metal cylinder of diameter 200 mm and height 3600 mm. It is equipped on the exterior with a spiral water cooler. Fluorine,

carefully heated to 360-400°C, is fed into the reactor by four jets. A stirrer with a gear, revolving at a rate of 1800 rpm, creates good contact between the solid particles of UF_4, entering at the top, and the fluorine. The temperature of the walls of the apparatus is kept between 330-540qC by means of the cooler.

The gaseous fluorination product, containing around 75% UF_6, is passed through water-cooled tubes and two cyclones, in which solid particles are collected. The gas is then fed into a cooling trap and a condenser of 1350 mm diameter and 4500 mm length with 48 thin tubes (diameter 12-5 mm).

Chapter Six
URANIUM ENRICHMENT TECHNOLOGY

6.1 Introduction

Due to its fissile properties, the light uranium isotope U^{235} is an important ingredient of the fuel used in nuclear reactors for power production. Uranium fuel enriched in U^{235} is used in most power reactors. Concentrations of U^{235} up to a few percent are required for such applications, while highly enriched U^{235} is used especially in nuclear weapons.

Uranium enrichment technology has obviously military implications even if current developments are aimed at civilian goals. This is why information in this area is largely classified and only some aspects of enrichment technology can be treated in open literature.

Separation processes can be more or less selective. High selectivity does not lead necessarily to the best plant economy. The combination of investment and power costs may well favor less selective processes. When selectivity is low, the process material must be submitted to the enrichment operation in a large number of subsequent steps (stages) to reach the wanted product concentration. The smallness of the single-stage separation effect and the rarity of the U^{235} isotope in the feed material lead to very large plant dimensions.

6.2 Enrichment process

Like all elements, uranium consists of different isotopes; the two that are the most abundant and relevant, are known as U^{235} and U^{238}. Natural uranium consists of approximately 99.3 % U^{238} and 0.7 % U^{235}. When uranium is to be used as fuel for a power plant or in a bomb, it is the less abundant isotope U^{235} that is needed because it is this isotope that is most likely to fission when it absorbs one neutron and releases several neutrons in each fission process. U^{235} is able to sustain a chain reaction if enough of the isotope is present. The challenge is to find a way to extract this isotope from the natural uranium, this process is called enrichment [22].

There are several ways of doing this, and some of the most important techniques are discussed in this search. It is customary to speak of three different levels of enrichment. Low-enriched uranium (LEU) contains less than 20 % U^{235}; the fuel used in power plants is usually enriched to about 3-5 %. Highly enriched uranium (HEU) contains more than 20 % U^{235}; uranium enriched to this level is used in some specialized reactors. If the uranium is enriched beyond about 90 %, it may also be referred to as weapons grade uranium.

The different methods for uranium enrichment all have in common that they make use of the different properties of the isotopes (either physical or chemical) to

physically separate $U^{235}$from U^{238}. No enrichment technology is perfect so the products will never be 100 % U^{238} in one place and 100 % U^{235}in another; there will always be two new mixtures, one which is enriched in U^{235}.

The physical property that most enrichment technologies make use of, either directly or indirectly, is the mass difference between the isotopes. Heavier particles move differently from lighter particles when experiencing the same external influence, and even though the mass difference between the two isotopes of uranium is very small, it is possible to exploit this to separate them from each other. In the gas centrifuge method, gas molecules containing U^{238} will concentrate closer to the centrifuge wall than the molecules containing U^{235}. The gaseous diffusion method exploits the fact that heavier molecules with a certain energy move slower than lighter molecules with the same energy and therefore will diffuse more slowly through a semi-permeable membrane. The mass spectrograph separates the isotopes because particles with a different charge-to-mass ratio will move differently in a magnetic field. Even chemical properties of a molecule may be affected by the mass of the atoms it consists of.

The separation elements in an enrichment facility and also the full facility may be described in a general way. In the following discussion some common factors of the various enrichment facilities will be described, and the elements may be considered as black boxes (it does not matter which enrichment technique is used). The feed, the form of natural uranium or uranium enriched to a lower degree than what is desired, is entering at one end, and the product (the enriched uranium) and the tails or waste (the depleted uranium) emerge at the other end. This terminology is also used for the entire facility. The separation factor of an element is the ratio of the relative amount of U^{235}in the product to the relative amount in the tails, and is therefore a measure of the efficiency. Separation factors are just above 1 for each step in the most common enrichment methods such as gaseous diffusion and gas centrifuges [22, 23].

6.3 Type of Uranium enrichment technology

❖ **Diffusion techniques**
1. Gaseous diffusion
2. Thermal diffusion

❖ **Gas centrifuge techniques**

❖ **Laser techniques**
1. Atomic vapor laser isotope separation *(AVLIS)*
2. Molecular laser isotope separation *(MLIS)*
3. Separation of Isotopes by Laser Excitation *(SILEX)*

❖ **Other techniques**
1. Aerodynamic processes

2. Electromagnetic isotope separation
3. Chemical methods
4. Plasma separation

6.4 Diffusion Techniques

6.4.1 Gaseous Diffusion:

Main Principle

This method indirectly uses the different masses of the isotopes. If a container is filled with gas and the walls are semi-permeable membranes, then the molecules in the gas will have a small probability of penetrating the walls. These walls will be referred to as barriers throughout this chapter. Molecules with a comparatively high speed will encounter the membranes more frequently than the slower ones and therefore have a larger probability of penetration. And since the lighter molecules on average move faster than the heavier ones, the gas on the other side of the wall will be enriched in the lighter molecules. In principle this process is very simple, but it is not very efficient because the mass difference between the molecules is so small. To make the diffusion go faster, a pressure difference is set across the membrane.
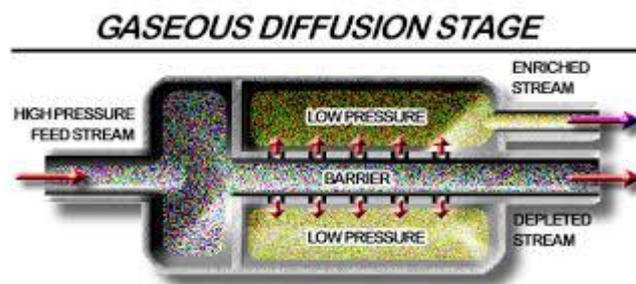
Figure (6.1) gaseous diffusion stage

The gaseous diffusion technology is relatively simple, but not very effective. The quality that can make gaseous diffusion interesting to a new participant in the enrichment industry, is that the technology has been proven to work on an industrial scale. The gaseous diffusion method employs uranium in the form of uranium Hexafluoride gas.

The basic physical principle underlying the gaseous diffusion method is the so-called 'equipartition principle' of statistical mechanics. This principle states that in a gas consisting of several types of molecules each type will have the same average energy of motion (kinetic energy). This equality of average energies is attained and preserved by the enormous number of collisions between molecules which are taking place at all times in the gas.

These collisions ensure that any excess energy which may have been associated with one component will rapidly be shared equally with all the others. This equal sharing is called thermal equilibrium. The kinetic energy [22].

KE of a molecule of mass m is related to its velocity v by the formula

$$KE = \frac{1}{2} mv^2 \tag{6.1}$$

Therefore, molecules which have the same average kinetic energy will have average velocities which differ in inverse proportion to the square roots of their masses. Using the symbol $<>$ to denote the average, the relationship can be written

$$< KE_1 > = < KE_2 > \tag{6.2}$$

$$\frac{1}{2} m_1 < v_1{}^2 > = \frac{1}{2} m_2 < v_2{}^2 > \tag{6.3}$$

Or

$$\frac{< v_1 >}{< v_2 >} = \sqrt{m_1/m_2} \tag{6.4}$$

For uranium hexafluoride gas made up of $^{235}UF_6$ and $^{238}UF_6$ th6e respective molecular masses are 349 and 352, so the ratio of the velocities is 1.0043, with the lighter $^{235}UF_6$mocules moving very slightly faster on the average. This last phrase is important, because the molecules move with a wide range of velocities, and the equipartition principle applies only to averages over large numbers of molecules [24].

The gaseous diffusion method of separation exploits this slight difference in average velocities by forcing the gas mixture to diffuse through a porous barrier under a pressure difference. The barrier is a thin wall of solid material containing many very small holes or passageways.

The faster molecules will encounter the holes more often than the slower ones and will therefore be slightly more likely to pass through, causing the gas which emerges on the other side of the barrier to be enriched in the lighter isotope. So the ratio of numbers emerging on the product side is

$$R_P = N_{235} \frac{< v_{235} >}{N_{238}} < V_{238} > = R_F \sqrt{(M_{238}/M_{235})} \tag{6.5}$$

Therefore, the ratio of the average velocities is the ideal enrichment factor

its The actual enrichment factor is less than this for two reasons. First, the concentration of the desired isotope on the feed side is not constant; it gradually decreases as the enriched mixture diffuses through the barrier. If the cut is assumed to be one-half, then this effect reduces the ideal enrichment gain α-1 by a factor of 0.69. An ideal cascade containing ideal separation elements would therefore have a = 1.00297, corresponding to a stage separation factor q = 1.00595 . This is further reduced by the separation 'efficiency', to be discussed below.
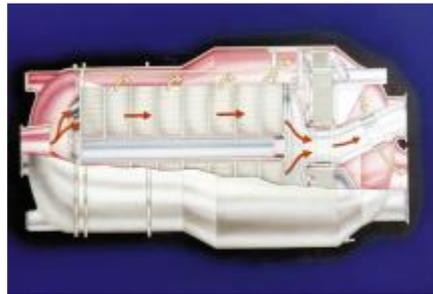
Figure (6.2) here, only the diffused gas is cooled at the outlet end of the diffuser housing, while the undiffused gas passes directly to the inlet of the compressor of the next lower stage.

6.4.2 Enrichment plant

Each stage in the gaseous diffusion process increases the fraction of U^{235} by very little (the separation factor is about 1.004), but contains a large amount of gas. This makes the process less flexible. Batch recycling is not a good option for increasing the level of enrichment, but stretching the cascade may be an alternative. If stretching is to be used in a gaseous diffusion plant, then precautions to prevent the possibility of a criticality accident are necessary to take. This also goes for batch recycling. More than 3500 stages is needed to produce weapons-grade uranium from natural feed, and there is an equilibrium time of more than a year

6.4.3 Separation stage

The main components of a single gaseous diffusion stage are :

1. A large cylindrical vessel called a diffuser or a converter that   contains the barrier;
2. A compressor used to compress the gas to the pressures needed for it to flow   through the barrier.
3. An electric motor to power the compressor;
4. A heat exchanger to remove the heat of compression;
5. Piping and valves for stage and inter-stage connections and process control [25].

The key parts in the gaseous diffusion method are the barriers and the compressors. This technology seems to be relatively simple to develop. It was done by the United

States in the 1940s, and reportedly every country that has tried since has been able to do it [26]. When the gaseous diffusion method was first developed, the corrosive nature of uranium hexafluoride was one of the major obstacles [23].

There must not be any leakage in the system letting uranium hexafluoride gas out or air in. Metals such as aluminum and nickel are used for the surfaces that are exposed to the gas, for example piping and compressors, because they are corrosion resistant. Since there is little enrichment in just one stage of a gaseous diffusion plant, the cascades are long. Also the equipment tends to be rather large, so a gaseous diffusions plant takes up a lot of space. When the gaseous diffusion plant at Oak Ridge was built in the early 1940s, it was the largest industrial building in the world. Support facilities for a gaseous diffusion plant would include a large electrical power distribution system, cooling towers, a fluorination facility, a steam plant, a barrier production plant, and a plant to produce dry air and nitrogen [25].

Figure (6.3) the gaseous diffusion plant built at Oak Ridge
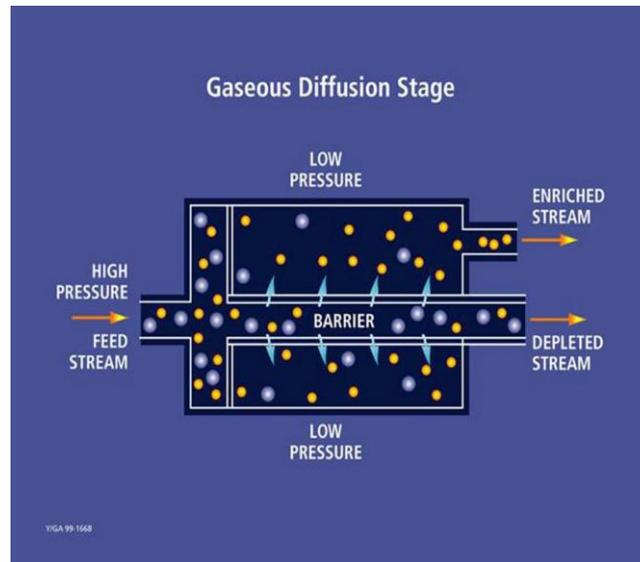
Figure (6.4) gaseous diffusion stage

In a typical stage the feed enters at a pressure of one-third or one-half atmosphere. Only half of the gas is allowed to diffuse through the barriers; this gas will have a lower pressure, and it is sent through a heat exchanger to be cooled. Then the gas is compressed and mixed with depleted gas from a higher stage. The cooling is necessary because the compression will substantially increase the temperature of the gas. This energy-demanding process is the main source of the high power consumption of the gaseous diffusion process. Modern compressors are similar to those used in jet aircraft engines. The compressor rotor rotates at high speed and must handle the corrosive uranium hexafluoride gas at high temperatures in continuous operation for many years [24].

6.4.4 The diffusion barrier

The central problem in the use of gaseous diffusion is the manufacture of a suitable barrier material. The difficulty in making the barrier can be appreciated if one lists the properties it must possess.

1. The average diameter of the holes (pores) in the barrier must be much less than the average distance travelled between collisions (the 'mean free path') of a molecule. If this is not satisfied then a molecule is likely to suffer one or more collisions near the entrance to the pore and inside the diffusion channel. This would cause exchanges of energy with other molecules and tend to cancel out the slight velocity difference between light and heavy molecules. At the same time the holes must be large enough to allow the gas to pass through at a reasonable rate.

  2. The barrier must be very thin so as to have an adequate permeability at reasonable pressure, but it cannot be so thin as to break under the necessary

pressure difference across it. It is desirable to have this pressure difference as large as possible to increase the rate of flow through the barrier.

3. The barrier material must be highly resistant to corrosion by the very corrosive gas UF_6. Any corrosion which occurs will cause plugging of the tiny holes in the barrier. Requirements 1 and 2 can be made more quantitative by considering the actual properties of UF_6 gas. A UF_6 molecule has a diameter of about 0.7 nanometers (nm), and at a pressure of about one-half atmosphere and a temperature of 80Â° the average separation between molecules is about 5 nm. The mean free path under these conditions is about 85 nm, so the average pore opening must be somewhat less than this, say about 25 nm. Using a rough geometrical argument it can be shown that such pore dimensions can be obtained by packing together spheres whose diameters are about 100 nm (0.1 pm) [24,25,26].

6.4.5 Gaseous Diffusion Stage

In the design of a separation stage all of these factors must be balanced in an optimum way. The operating pressures and temperatures, the properties of UF_6 gas, and the structure of the barrier all combine to produce barrier efficiency e_B, which is a number somewhat less than 1. The actualseparation gain is related to the c by:

$$g = e_B G_{ideal} \qquad\qquad (6.6)$$

The structure of the barrier also determines its 'permeability7, or the rate of flow of gas through a given barrier area for a given pressure difference across it. Using data obtained from experiments with argon it is possible to estimate the dependence of permeability on pore size . Knowing the permeability of the barrier and its efficiency, it is then possible to compute how much separative power can be produced by a given area of barrier material. As an illustration assume that the barrier efficiency is about 0.7. Then the actual separation gain will be:

$$g = 0.7 \times 0.00595 = 0.0042$$

Which can be shown to lead to about 1 kg SWU per square meter of barrier per year. Therefore a plant such as the one at Tricastin, with a total capacity of 10.8 million SWUI yr, might have a total barrier area of 10^7 m², or about 10 square kilometers. A single large stage with a capacity of about 12 000 SWUI yr will have a barrier area of roughly 1.2 hectares. The individual tubes which make up the barrier must be small enough to provide a large surface area for diffusion but large enough to permit easy flow of the process gas. Again, no information is available on the size of the tubes, but if it is assumed that the tubes are about 2 m long and 1 cm in diameter, then about 160 000 of them would be used in such a stage. This can be compared with some of the early US stages which contained several thousand tubes each.

Using the above value of g , it can be shown that an ideal gaseous diffusion cascade for the production of 3 per cent enriched product with a 0.2 per cent tails assay would have about 1 290 stages.

Table 5.1 can also be used to calculate the total cascade flow and the flow per stage. For a 10^7 SWUI yr gaseous diffusion plant the product flow P would be 2.4 x 10^6 kgl yr, and the total cascade flow would be between 4.6 x 10^{12} and 6.0 X 10^{12} kgl yr. If the cascade were squared off, a typical large unit might carry a flow of

5.5 x 10^9 kgl yr or 174 kg Ul s. This implies a flow through a given stage of 250 kg UF6/s, a value comparable to the rated capacities of the largest compressors at Recasting (190 kg/s).

It should be emphasized once more that the numbers used here are only estimates designed to produce approximate values for stage and plant parameters. The precise values of numbers such as barrier efficiency are well-guarded secrets.

6.4.6 Energy Consumption

The gaseous diffusion method consumes a lot of energy, about 2500 kWh/SWU. The energy consumption was more than ten times this level when the process was first developed, but it decreased by a factor of ten in about ten years of further development. One method for dealing with the expenses arising from the high energy consumption, which is inevitable in the gaseous diffusion method, is to use the plant only in periods of lower electricity prices. This has been done in France [23].

6.4.7 Countries Using these Technologies

The gaseous diffusion method was first used in the United States during World War II, but has since been used in the United Kingdom, France, China, Russia and Argentina as well. Since gaseous diffusion is an out-dated and highly power-consuming process, it is not in wide use anymore. Today only the United States and France use gaseous diffusion on any significant scale [27].

6.5 Laser techniques

6.5.1 Atomic Vapors Laser Isotope Separation (AVLIS)

6.5.1.1 Brief Description of the (AVLIS) Process as Applied to Uranium

In the beginning, LIS ideas in physical and industrial aspects were developed in the framework of national AVLIS programs on uranium enrichment. Some key steps and stages of this process.

Let us briefly describe the AVLIS process as applied to uranium. It is known that uranium conversion to appropriate fission fuel necessitates the concentration of U^{235} from 0.7% (the natural contents of isotope in a mixture with U^{238}) to 3 %. In

AVLIS technology, atomic vapor that is a natural uranium isotope mixture is obtained by electron–vacuum evaporation in a special high vacuum technological unit. Then vapor passes from the evaporation unit to flow former, where it is shaped to a required, for example, leaf-like form. Then it comes to an interaction zone with light beams produced by a dye laser system. The dye laser system is pumped by another high-power system of copper-vapor lasers. The ionization potential for uranium is U ion = 6 eV; the isotopic shift of uranium levels amounts to 0.08 cm^{-1}. Then uranium atomic vapor is photo ionized in the interaction zone via a three-step excitation process. The spectral width of radiation, the operating wavelengths $\lambda 1$, $\lambda 2$, $\lambda 3$, and the average power of dye laser beams are matched in such a way that after absorbing three sequential quanta of light at specified uranium atom transitions, only the required isotopes would be excited in cascade and photoionization processes. The operating levels of uranium atom are schematically shown in Fig. (6.5). Quanta of energy for each transition are near 2 eV. The choice of particular levels is a rather complicated spectroscopic problem [25, 28].

Uranium possesses a branched system of atomic levels and has many valence electrons. The wavelengths of possible laser sources are also shown in Fig(6.6) When a copper-vapor laser pumps dye lasers, all three wavelengths of the dye lasers are chosen in the red spectral range because a uranium atom has allowed transitions between levels suitable for efficient cascade excitation in this range (590–600 nm). In the three-step scheme, an atom at the upper auto ionization.

The atomic vapour laser isotope separation (AVLIS) process consists of a laser system and a separation system. Uranium in its pure metallic form is melted and heated in a crucible by an electron beam to the point where enough of the atoms evaporate. The evaporated atoms move radially away from the ingot. Warm liquid uranium is very corrosive and must not come into contact with the support structure. It is also important that the heat does not disappear, and that the atoms can move relatively freely; therefore an extensive mechanism is needed to create vacuum and contain the heat. The liquid uranium is in one end of an evacuated chamber, and laser beams are cutting through the other end. As the uranium gas moves away from its source, the atoms collide and most of their internal energy is converted to kinetic energy. When the atoms reach the radiation zone (the laser beams), about 40 % of them are in their lowest energy state (the ground state) [28]. When the atoms reach the radiation zone, the U^{235}atoms which are in the ground state will be excited two times and then ionized (see Figure (6.5)). The ionization energy for the uranium atom is about 6.2 eV.12 In theory, just one laser with the exact energy could be used to ionize the atom, but in practice it is necessary to do the ionization in at least two steps. Three steps of about equal size have been used to take advantage of the readily accessible and well-established technology of rhodamine dye lasers. The ions are then collected on charged plates oriented

parallel to the radial direction, while most of the neutral atoms continue outwards and are collected on plates orthogonal to their direction of motion. It is impossible to avoid collecting some neutral atoms on the collector plates for the ions because of the random motion of the molecules in a gas .This and other problems decrease the efficiency such that the maximum separation factor will be about 15. In practice, a separation factor of between five and ten is more likely, and this is far greater than the separation factor of processes that are already in use, such as gaseous diffusion and gas centrifuges where the separation factors are just above 1 [28].

An AVLIS module consists of the crucible holding the liquid uranium and the collector plates, and it is encapsulated in a vacuum system. All laser sources and mirrors are kept outside the volume containing the uranium steam. Each module can be typically 1 m high and 3 m long. The laser light is reflected many times by mirrors and may have a path length of more than 100 m [28].

Since only about 40 % of the U^{235}atoms are in their ground state, only these 40 % can be excited, and this lowers the efficiency of the process. But an additional about 30 % are in a low excited state and can be included by adding another laser with a slightly lower energy which excites these atoms into a state with the same energy as the atoms in the ground state are excited to (cf. Figure 8.1). In this way, 70 % of the U-235 atoms may be ionized instead of only 40 %. The four lasers that are needed all have slightly different colours corresponding to the slightly different energies required, but all are in the red-to-orange region of the spectrum [28].

6.5.1.2 Advantages and problems with the AVLIS process

One of the advantages of the AVLIS process is the high separation factor. This suggests that producing highly enriched uranium would be easier with this process than with many other processes. With a separation factor of ten, only three times of batch recycling will be needed to reach a product of 97 % U^{235}. But there are some obstacles. When the fraction of U^{235}in the feed is increased, a higher laser intensity is needed to maintain the same separation factor because there are more atoms present which need to be excited. When the feed contains more than 50 % U^{235}, it will be better to tune the lasers to ionize the U^{238}atoms. Then the intensity of the lasers will not have to increase as the enrichment level becomes larger, but can instead decrease. If the steam is too dense, it will be more difficult to collect alt the atoms, and some will be lost. On the other hand, if the density of the steam is to low, the atoms will not have the opportunity to collide often enough to reach the ground state before they enter the radiation zone [28].

Also only about 50 % of the atoms reach the radiation zone with the laser beams after being evaporated, the remaining atoms are collected on the walls of the chamber and must be recycled. This makes the process less efficient [28].

The AVLIS enrichment process will generate a small amount of waste and consume only about as much energy as the centrifuge process, 100–200 kWh/SWU [4,23]. The process does not require as much space as a centrifuge plant, however [29].

6.5.1.3 Photoionization of a uranium atom

level decays into electrons and ions, that is, it is ionized. High selectivity of the preferred ionization for the desired isotope is obtained at all cascade-excitation steps due to fine matching between the frequencies of tunable lasers and the energy levels of the uranium atom. U^{235} isotopes ionized in
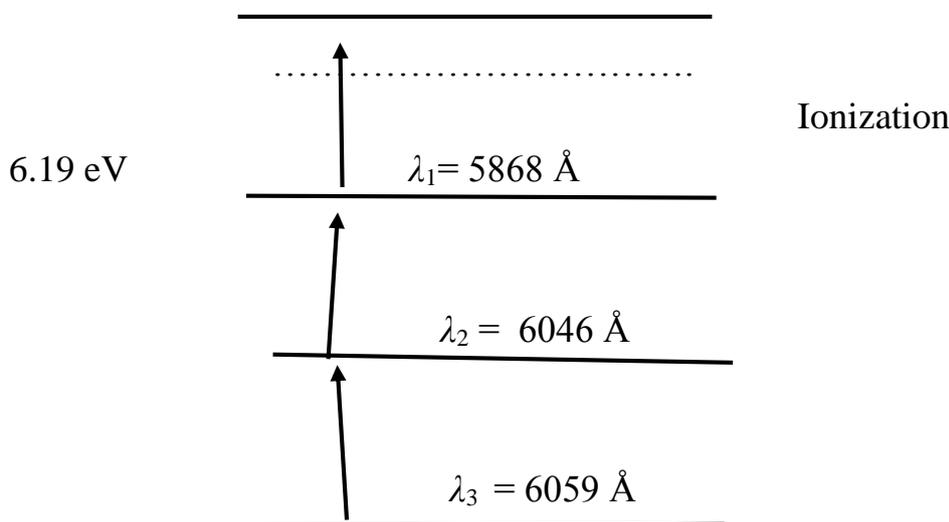
Figure (6.5) the ionization stage extracted.

This way are extracted from the interaction zone by an electric field and are directed to collector plates where they are neutralized and condensed as the final product of separation.

The AVLIS method is based on multistep isotope-selective ionization of atom.

Generally speaking, the upper excited level can be ionized in different ways. Conventionally, a laser radiation is used. Isotope-selective excitation of atom became possible by means of a frequency-tunable laser radiation with the spectral line width less than the specific frequency differences between the absorption lines of isotopes under consideration.

One important problem with the method considered is the choice of a photoionization scheme. It may be a two-level, three-level (see Fig(6.5), or four-level scheme. The choice is mainly determined by the laser source. Most of the atoms have such a structure of energy levels that the first-transition radiation (with the wavelength λ_1 in Fig. (6.5) should lie in the visible or UV spectral range.

For the second transition, the IR radiation may be used. The corresponding lifetimes are :
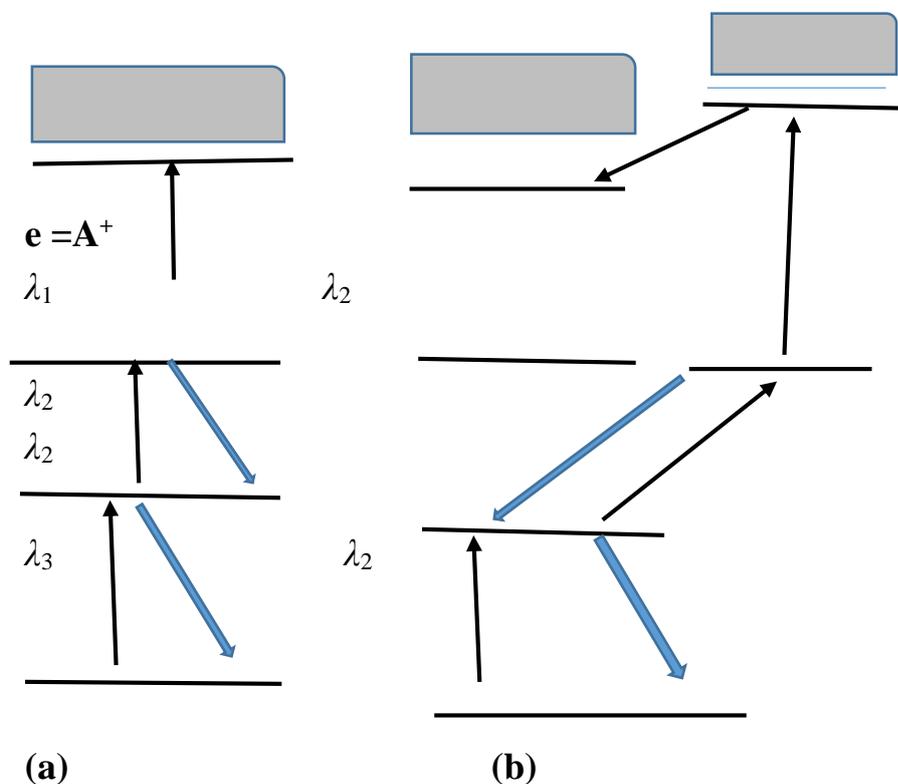
Figure (6.6) stepwise ionization: (a) direct photoionization; (b) ionization via the auto ionization state (heavy line).

The dotted lines designate the radiation decay channels for levels; the thick arrow refers to the decay of the auto ionization state into electrons and ions.

Usually not very long and lie in the range 3–200 ns (we do not consider excitation of metastable or Rydberg levels).

To avoid losses to spontaneous emission one should employ pulse-periodic sources of laser radiation with the pulse duration lying in the range mentioned.

The characteristic excitation time for a level is determined by the formula $\tau = (\sigma\Phi)^{-1}$, where σ is the absorption cross-section and Φ is the quantum flux. The energy of the pulse per unit area is $W = h\nu\Phi\tau = h\nu/\sigma$. The magnitude of σ is at least 10^{-15} cm^{-3} even in the case of Doppler broadening. For the characteristic quantum energy of 1 eV we have $W \approx 160$ **μJ cm**$^{-2}$. At the pulse repetition frequency of 10 kHz (the typical frequency for pumping pulses) we obtain that the required power

is in the range 1–$2\ \mathrm{W\ cm^{-2}}$. The radiation power of modern laser sources noticeably exceeds this value in UV, visible, and IR spectral ranges.

Energy levels are excited by moderate energy, whereas direct photoionization necessitates energy a few orders higher. This is explained by the small cross-section of photoionization, which is 10^{-17}–$10^{-18}\ \mathrm{cm^2}$. Under the above-mentioned conditions, the average power of the ionizing radiation is estimated as 100–1000 W.

The low cross-section of direct photoionization results in a weak absorption of the ionizing radiation. Hence, the optical density is a few orders lower than the corresponding value for the radiation exciting atomic levels. A serious problem of matching optical paths arises at various steps of photoionization. The photo ionizing process will mainly determine the energetics of isotope selection.

Ionization via the decay of auto ionization levels can be used instead of direct photoionization. Such levels lie higher than the first ionization potential. Emission lines from auto ionization states are observed in most of chemical elements. Presently, identification of auto ionization levels, and refinement of their positions and lifetimes are a subject of intensive investigations. The cross-section of such levels may amount to 10^{-15}–$10^{-16}\ \mathrm{cm^2}$. Hence, the problems mentioned above are to a certain degree tempered. Search for appropriate auto ionization levels[28,30]. is one of the most important problems in spectroscopy. By tuning the frequency of a tunable laser one can find from the electron yield due to ionization or luminescence processes the efficiency of exciting auto ionization levels. The radiation lifetime of auto ionization levels is usually a few orders longer than the time of atom auto ionization decay into electrons and ions. Hence, we can neglect the losses related to spontaneous emission from such levels.

Conventionally, the isotopic structure with hyperfine line splitting is rather dense and overlapped with Doppler broadening. In this case, the Zeeman effect can be used by applying a magnetic field in order to separate the lines by a frequency shift sufficient for selective excitation of atoms. In particular, this method is recommended for palladium isotopes.

One more method of photoionization is to excite long-living Rydberg states lying close to the ionization threshold. A relatively high ionization cross-section and proximity to the ionization threshold make employment of powerful infrared lasers (CO_2, CO, etc.) probable.

Under an electrostatic field applied, Rydberg levels shift. If then the electric field is switched off, some high levels may become higher than the ionization threshold and the atom may decay into electrons and ions.

A drawback of such ionization via Rydberg states is a high sensitivity of the state positions to weak induced external electric fields, which smears the levels and hinders their excitation.

6.5.1.4 Theoretical Description of the Method for Incoherent Interaction Between Radiation and Atoms

We have considered some basic photoionization methods. However, physics of level excitation may differ for radiation pulses with different durations and line widths. At early investigation stages, mainly a broadband radiation was used. The spectral width of radiation exceeded the inverse pulse duration time or inverse radiation lifetime. Under these conditions, the analysis may be performed in the incoherent approximation of the interaction between radiation and atoms.

The method of isotope separation considered in the case of incoherent interaction can be described in the framework of the kinetic model, which is widely used for the analysis in gas laser physics. We will assume Doppler distribution of atoms over velocities. Let us first consider the point model without taking into account spatial variations of parameters. For better perception we also assume that the spectral width of the atomic absorption line is far less than that of the radiation line and of Doppler profile. Later, we will make corrections for the case where such assumptions are not applicable.

Under the conditions mentioned, we may assume that binary impacts between atoms and photons corresponding to frequency v occur during the radiation pulse. For such cases, we will employ the density of atoms and photons per unit frequency interval and for other cases the total particle concentration will be assumed.

As is shown in Appendix A, the kinetic equations can be obtained automatically. It suffice to indicate all the processes in the form of chemical reactions with the corresponding rate constants. Reactions given below are the same for all isotopes. Excitation and depopulation of atomic levels by radiation is described by the following reactions:

$$N_i + hv_i \Rightarrow N_i + 1 ; \; \alpha = ck_i \left(g_i + \frac{1}{g_i} \right) \tag{6.7}$$

$$N_i + 1 + hv_i \Rightarrow N_i + hv_i \quad ; \alpha = ck_i \tag{6.8}$$
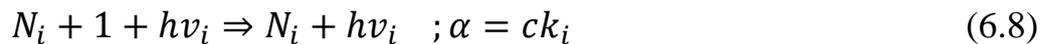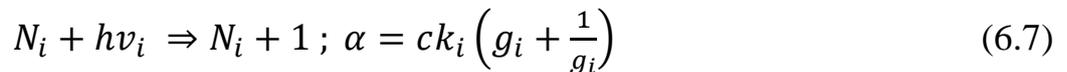
Here *Ni* is the atom at the *i*th level; *hvi* refers to the radiation in a transfer from the $(i + 1)$st to the *i*th level. To the right of the reactions, the corresponding rate constants are given, where c is the speed of light; *gi* is the statistical weight of the *i*th level; and $k_i = A_i + 1\lambda^2_i/(8\pi)$ (λi is the wavelength and $A_i + 1$ is the Einstein factor for the transition considered).The photoionization reaction looks like

$$N_{UP} + h\nu_{ion} \Rightarrow N^+ + e \tag{6.9}$$

where Nup is the upper level in the stepwise scheme of photoionization; $h\nu$ ion refers to the ionizing radiation; and $N+$ and e denote ions and electrons, respectively. The rate constant depends on the details of atom excitation to the autoionization state. It is worth noting that the corresponding experimental data seem to be more reliable than the theoretical formulae available.

If a spontaneous decay of level occurs at the preceding level in the scheme of stepwise ionization, then we can write:

$$N_i + 1 \Rightarrow N_i \quad ; \alpha = A_i + 1 \tag{6.10}$$

and if the decay occurs at the levels that are not taken into account in the model, then we can write:

$$\boldsymbol{N_i + 1 \Rightarrow \quad ; \alpha = (^1\!/_{t_i} + 1) - A_i + 1} \tag{6.11}$$

Here, $\tau_i + 1$ is the total radiation lifetime of the level.

In certain cases, one should also take into account the reaction of energy transfer in impacts from the atom of one isotope to the atom of another isotope:

$$N_i^* + N_j \Rightarrow N_j^* + N_i \tag{6.12}$$

Where the indices refer to different isotopes.

The concentration of atoms can be limited above due to the charge exchange reaction for ions:

$$N_i^+ + N_j \Rightarrow N_j^+ N_i \tag{6.13}$$

The rate constants for the reactions depend on the distribution function of particles over velocities. Calculation of rate constants for a particle flux is a complicated problem and beyond the scope of this search.

The reactions considered describe mainly a conventional point model of the process of pulse radiation. Nevertheless, this model neglects the possibility of super luminescence from one of the considered levels to the levels that are not directly excited by radiation in the process of stepwise photoionization. The latter factor is important if the upper state comprises Rydberg states. The cross-sections of induced transitions between them are so high that a super luminescence occurs at the density much lower than a typical operating concentration of atoms.

$$\alpha_{n(v)} = \int_0^\infty \frac{n_{ph}(v'-v)}{n_{ph}} l(v')dv' \tag{6.14}$$

$$\alpha_{s(v)} = \int_0^\infty \frac{N_i(v'-v)}{N_i} l(v')dv' \tag{6.15}$$

where n_{ph} is the photon density at the corresponding transition[33].

In the equations for level populations, the rate constants of excitation and de excitation by radiation are multiplied by α_N. In the equations for photon density, the rate constants are multiplied by α_s. In a one-dimensional model, variables depend on the coordinate along the laser beam. The only change to be made in writing the equations is to substitute the ordinary derivative by a substantial one in the photon density equation:

$$\frac{d}{dt} = \left(\frac{1}{c}\right)\frac{\partial}{\partial z} + \frac{\partial}{\partial t} \tag{6.16}$$

Where z is the coordinate along the laser beam.
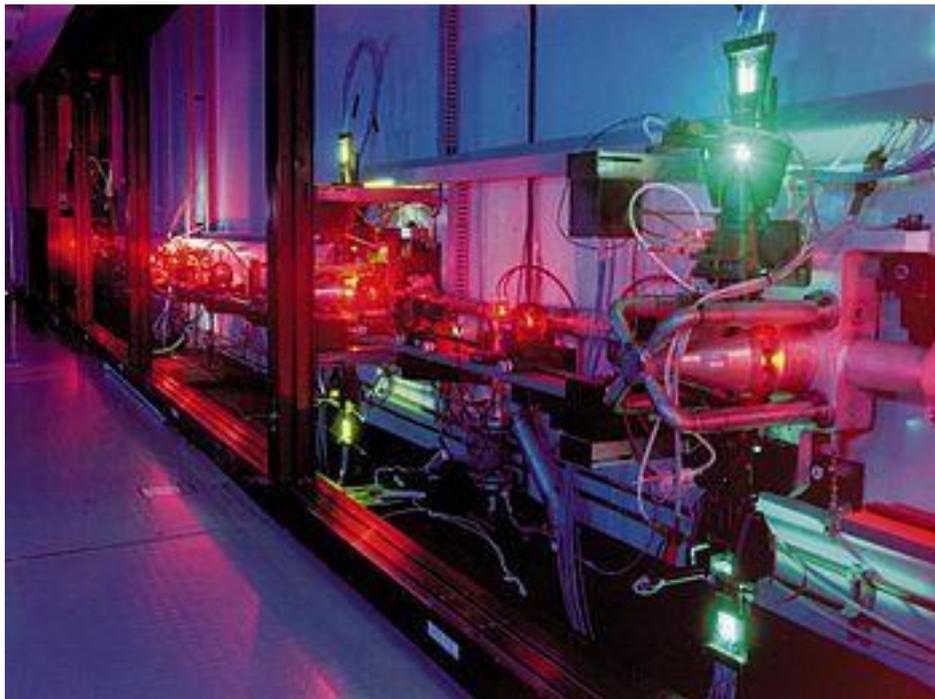
Figure (6.7) laser beam separation equipment

Laser isotope separation makes use of the quantum mechanical properties of atoms and molecules. Lasers are light sources that can provide an intense beam of light with a very precise wavelength or energy of the photons. If one can produce a laser capable of emitting light at the exact frequency that is needed to separate the two

isotopes of uranium, it will be a very efficient way of enriching. therefore possible to "practice" the use of LIS technology and LIS equipment without using uranium just by tuning the lasers to a different frequency.

Unlike many other isotope separation methods, LIS can be used to separate the isotopes of plutonium [28].

6.5.2 Molecular Laser Isotope Separation (MLIS)

6.5.2.1 The main principle

The energy differences between the vibrational states of a molecule are smaller than the differences between the electronic states of an atom, so for the dissociation of molecules infrared light must be used instead of visible light. Photo dissociation of a molecule (see Section A.5) can be done in one, two or more steps. When two steps are used in the molecular laser isotope separation (MLIS) process for enrichment of uranium, the first photon is in the infrared section of the electromagnetic spectrum. This is the one that selects the desired molecule by exciting the vibrational states of the molecules containing U^{235} and not U^{238}. Then a photon of higher energy, in the ultraviolet end of the spectrum, breaks the bond completely. Molecules which are already excited have a larger probability of absorbing the photon and be dissociated than those in the ground state, and in this way the selectivity of the first step is conserved. But it is not impossible also for the unexcited molecules to be dissociated by the UV photons, and this makes the process less effective. In this two-step process the excitation and the dissociation must occur on a time scale that is short compared to the lifetime of the intermediate vibrational state. Alternatively, several infrared photons of the same frequency may be used to dissociate the molecules and still be isotopic ally selective [30].

The MLIS process uses uranium hexafluoride gas because of its chemical qualities14, and because its handling is well known from its use in the gaseous diffusion method and the gas centrifuge method. Other molecules could also be used. The idea is to break the bond between the uranium atom and one of the fluorine atoms in the molecules containing U^{235} so that uranium pent fluoride and a free fluorine atom are formed. Uranium pent fluoride enriched in U^{235} will then precipitate15 as a white powder and can subsequently be collected [4].

As in the AVLIS process, it is the ground state that has to be occupied initially if the laser is going to be able to excite the molecules. The molecules in the uranium hexafluoride gas collide so often that almost none of them occupy the ground state. The molecules can be forced into the ground state by cooling, but before room temperature is reached uranium hexafluoride will precipitate. One way of getting around this is by super-cooling the gas. Super-cooling means cooling below the sublimation point without crystallization taking place. This is a very unstable state, and if just a small crystal is present or the gas is disturbed, then crystallization will occur. Gaseous uranium hexafluoride mixed with a carrier gas and a scavenger gas is expanded through a nozzle at large speed, this cools the gas down to low temperatures without the uranium.

Hexafluoride precipitating. Suitable carriers are hydrogen or a noble gas. A scavenger gas (such as methane) is used to capture the fluorine atoms that are

released as a result of the dissociation of the uranium hexafluoride molecules containing U^{235} [25].

By this process, 95 % of the molecules will descend to the ground state, and only one laser is needed to excite most of the molecules (unlike in the AVLIS process where two lasers were needed and only 60 % of the atoms were excited). [28]

The energy consumption of the MLIS process is estimated at about 235 kWh/SWU which is slightly higher than for the AVLIS process. The use of uranium hexafluoride and low power consumption are some of the advantages of the MLIS process [23, 25].

6.5.2.2 Potential for HEU Production

It was a concern when developing the process that the uranium pent fluoride and the uranium hexafluoride molecules might exchange fluorine atoms before being separated, but this turned out not to be a big problem [30]. Also as the enrichment in the feed increases, this problem will decrease because there will be fewer U^{235} atoms present to change places with the U^{235} atoms. This makes the MLIS process more suitable for cascading than the AVLIS process. Also removing the uranium pent fluoride and converting it back to uranium hexafluoride does not pose great difficulties. And the amount of U^{235} can always be made to match the lasers, independent of the enrichment level, because the uranium hexafluoride gas is diluted by a carrier gas. All of this implies that using the MLIS process for HEU production may be possible in relatively small plants Z.

6.5.3 Separation of Isotopes by Laser Excitation (SILEX)

The SILEX process was developed in Australia by the company Silex Systems Limited (Silex). It is an MLIS process using uranium hexafluoride as feed [32]. Many details of the process are classified, and it is difficult to distinguish SILEX from the general MLIS technique described above. The technique uses a pulsed CO_2 laser of wavelength 10.8 μm which is converted to about 16 μm by use of a par hydrogen Raman converter [31]. The 50 Hz pulse rate of the lasers is not enough to produce significant enrichment; it must be increased at least ten times [33]. Both the product and the tails are removed mechanically, and the separator system is cascaded [34].

SILEX has advantages to other processes for uranium enrichment such as low power consumption, simple and practical separation modules and versatility [35]. Because of this, the SILEX method is suggested as the third generation of uranium enrichment, where gaseous diffusion and gas centrifuges constitute the first and second generations, respectively. The separation factor is classified, but announced by the company Silex to be somewhere between 2 and 20 whereas it is 1.3 for gas centrifuges and 1.004 for gaseous diffusion [35].

6.5.3.1 Laser Assisted Processes

The information in this section is from "Uranium Enrichment and Nuclear Weapon Proliferation" [28].

The laser's ability to excite only one kind of molecule can be utilized in several different ways in addition to those mentioned above. One example is that they may raise the temperature of one component of a gas mixture so rapidly that the rest of the mixture is not heated notably during the process. This could be done to uranium hexafluoride molecules by use of an infrared laser similar to the one used in the MLIS process. When the molecules are excited into a higher vibrational state, and the molecules collide with others, much of the vibrational energy can be converted into kinetic energy, which makes the molecules move and diffuse faster. This is a quality that can be used to enhance the effect of some of the methods which use uranium hexafluoride.

One example is the thermal diffusion process. If only uranium-235 hexafluoride molecules are heated, then fewer stages are needed to achieve the same separation of U^{235} and U^{238} than without the use of lasers. Another method is to combine it with an aerodynamic process.

Another proposed method involves letting a stream of a gas mixture of uranium hexafluoride and argon be irradiated by an infrared laser. At the end of the tube, there are very cold plates (below -150 °C) on which the gas molecules will condense if they collide with it. Because the molecules containing U^{235} will have a higher velocity, they are more likely to collide with the plates. When the tube is evacuated, the plates can be heated to free the uranium hexafluoride in gaseous form again and the gas can be sent on to the next stage in the process.

6.5.3.2 Countries Using SILEX Technologies

More than 20 countries are known to have done research on laser isotope separation techniques.16 France, the United Kingdom and the United States have developed LIS programs that could move beyond the lab into the pre-industrial phase and ultimately into commercial production. In August 2002, it was announced by the dissident group National Council of Resistance of Iran that Iran had started an LIS program and developed a laser enrichment facility. This enrichment program was not detected by the International Atomic Energy Agency (IAEA), but had supposedly only enriched milligrams of uranium. In September 2004, IAEA discovered South Korean experiments; in 2000, scientists at the Laboratory for Quantum Optics at the Korea Atomic Energy Research Institute had separated about 0.2 g of U^{235}enriched to levels between 10 % and 77 %. It seems that LIS methods are hard to upgrade to commercial scale, but scientists would not need a commercial-scale plant to enrich enough uranium for a single nuclear weapon if they had one or two years in which to do it. [34] The challenges posed by the AVLIS technique are mainly technical. The complex laser systems and the fact that uranium vapour is very corrosive, lead to technical difficulties that make the process hard to apply on a commercial scale. In June 1999, the United States was on the verge of commercialization when it was decided to cancel its AVLIS

program. About two billion dollars had then been spent on the program. Faced with stiff competition from gas centrifuges, large-scale LIS processes have not been developed by any of the countries who have sought this path for uranium enrichment. [34] The only potential large-scale LIS project seems to be the one conducted by the Australian company Silex .

Silex Systems Limited (Silex) was established in 1988. The principle was demonstrated to work in 1994, and in 1996 Silex reached an agreement for the development and licensing of SILEX technology with the United States Enrichment Corporation (USEC). In 2000, the process was demonstrated at prototype level, and in 2001 the technology was classified by the United States and Australian governments. The classification of the process has been interpreted by some to mean that the process can be used to produce highly enriched uranium for weapons use, but in 2007, it was claimed that the SILEX process was not mature enough to enrich significant amounts of uranium to the levels required in nuclear weapons [30]. USEC withdrew from the project in 2003, but in May 2006, General Electric Company signed an exclusive commercialization and license agreement for the SILEX uranium enrichment technology [35].

6.6 Electromagnetic processes

The electromagnetic processes for the separation of uranium isotopes all have in common that they make use of uranium ions[8]. Ions of different mass but equal charge will behave differently in a uniform magnetic field. The first process, known as EMIS, is relatively simple in theory, and the main challenge is that it is relatively slow. The two other methods are plasma processes (Ions have a different numbers of electrons than they have protons; this means that they have a net electric charge).
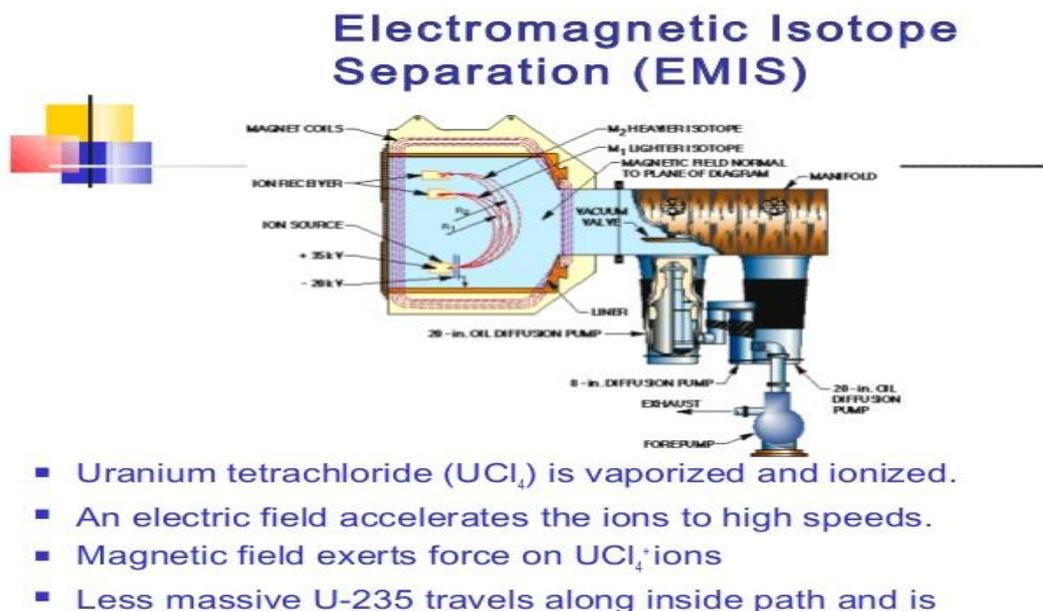
Figure (6.8) electromagnetic separation (EMIS).
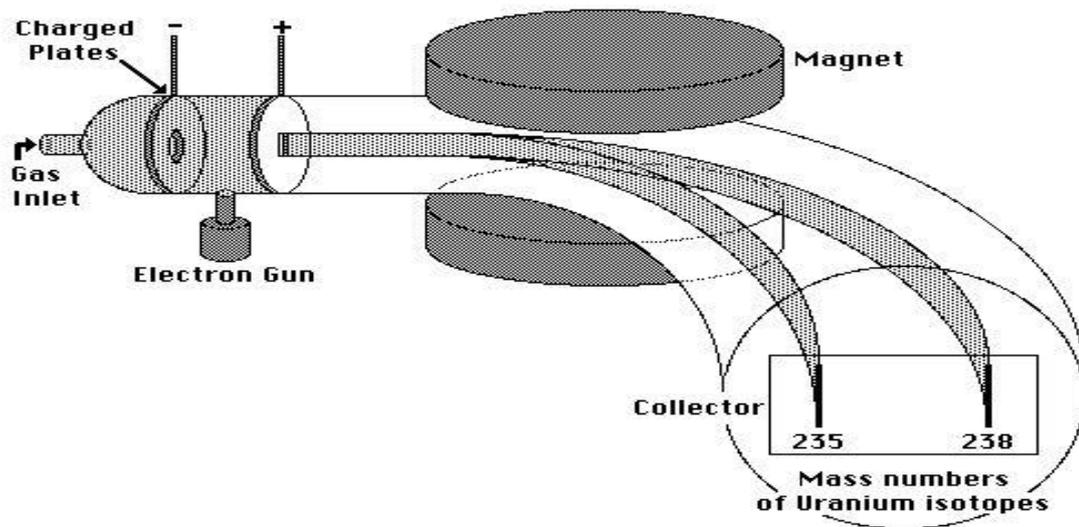
Figure (6.9) Simple drawing of a mass spectrometer. The particles enter in the bottom with the same kinetic energy and the same electric charge, and the beam will then split in two because of the different masses of the isotopes. The magnetic field points into the paper.

6.6.1 Mass Spectrometer: EMIS (calutron)
The Main Principle

In a uniform magnetic field, a charged particle with a velocity component orthogonal to the field will follow a circular or helical path. If it has no velocity component along the field, the trajectory will be circular in the plane orthogonal to the magnetic field; this is the case in the mass spectrometer. The radius of the circular path is determined by the strength of the field, the electric charge, the kinetic energy of the particle and the particle's mass. A beam of uranium ions with equal charge and kinetic energy being sent into such a uniform magnetic field will split into two beams because of the different masses of the isotopes. The beams will form circles, and the one containing U^{238} will have a larger radius than the one containing U^{235}. This is illustrated in Figure 6.1. The isotopes can be collected for example after half a rotation in the field. The mass spectrometer method is often referred to as EMIS (electromagnetic isotope separation), and the spectrometer is referred to as a calutron, which is what the American machine developed at the University of California in the early 1940s was called [35].

6.6.2 Equipment and Efficiency

In principle, a mass spectrometer consists of a large magnet that can provide a uniform magnetic field at a sufficient strength, a vacuum pump, an ion source and collectors. The ions of the two uranium isotopes U^{235} and U^{238} differ in mass by 1.27 %, and they will have radii that differ by only 0.64 %.

The uranium ions can be made from uranium tetrachloride ("green salt"). Solid uranium tetrachloride is electrically heated to produce uranium tetrachloride vapor. The vapor molecules are bombarded with electrons, and this knocks loose U+ ions. The ions are then accelerated over an electric potential to high speeds and follow circular paths in the magnetic field. Because they are accelerated over the same potential they acquire the same kinetic energy.

The EMIS process has a very high separation factor (somewhere between 30 and as high as 80 000), but a very low throughput. It also has a very high energy consumption (3800 kWh/SWU for the early machines, but this value may be lower today).

One limitation is the ion beam intensity because the charges on the ion beam particles will repel each other. To avoid this problem, the ions can be allowed to collide with a background gas so that electrons are released and neutralize the repulsion, but if the gas is too dense and the collisions became too frequent it would disturb the ion beam too much. The typical currents are a few hundred mill amperes, and at this rate it would take more than 400 years to collect a critical mass [4].

Another limitation is the fact that only a small amount of the feed material can be converted into product in one run. Only about 50 % of the uranium tetrachloride feed is converted to uranium ions, and only about 50 % of the ions are actually collected in the end. This is a problem if valuable partially enriched uranium is used as feed, and the insides of the vacuum chamber have to be cleaned so the material can be recovered. This is a time-consuming process which decreases the efficiency.

 *Note that EMIS is an isotope separation method which can be used also to separate the isotopes of plutonium.

6.6.3 Countries Using EMIS Technologies

The method was used, among others, by the United States during World War II to enrich uranium for their uranium bomb, and the machines were called "cauldrons". Because of the low through-put, over 1000 cauldrons were used to help produce uranium for the first American bomb. This cost roughly a billion dollars over three years [20]. The enrichment was done in two stages; in the first stage natural or slightly enriched uranium was used to increase the enrichment to 12–20 %; and in the second stage the product from the first stage was further enriched to weapons grade. EMIS could be used to produce limited amounts of weapons-grade uranium, and it may be especially attractive for further enrichment of partially enriched material. The United States, the United Kingdom, France, the Soviet Union and China all tried this enrichment method. It was also pursued by the Iraqi uranium

enrichment program, and this happened after the process had been discarded by other countries. The process is both energy demanding and labor intensive, and it is not economically competitive with other technologies.

6.7 Ion Cyclotron Resonance

This process employs the same principle as used in the mass spectrometer, only this time the particles also have a velocity component along the magnetic field. The motion of the charged particles becomes a helix with its axis along the magnetic field. The frequency of the rotation is determined by the strength of the magnetic field and the mass and charge of the particles, and is called the cyclotron frequency. If the uranium ions have the same charge, then the U^{235} ions will have a larger cyclotron frequency than the U^{238} ions. If the ions are subjected to an oscillating electric field in the plane orthogonal to the magnetic field, which has the same frequency as the cyclotron frequency9 of the U^{235} ions, then their energy will increase (the ions are excited) and the radius of the helix for these ions will increase while the radius of the U^{238}ions remains about the same. A typical unexcited ion will have a helix radius of the order of a millimeter, but excited U^{235} ion can be made to move in circles several centimeters in diameter.

The ions with the larger radius in their orbit can be collected by plates parallel to the helix axis while most of the ions which still have a small radius can pass without being caught by the plates.

6.7.1 The Basic Work

The ions can be produced from a feed plate of solid uranium. The atoms are vaporized by bombarding the plate with energetic ions in a process called sputtering. A microwave antenna located in front of the plate, energizes free electrons which collide with neutral uranium atoms in the vapor sputtering off the plate. This in turn displaces electrons from the uranium atoms and produces a plasma of $U^{235}$and U^{238} ions. The ions then move in their helical paths toward the collector inside a cylindrical vacuum chamber. The magnetic field is produced by *a* super conducting magnet located around the outside of the chamber [21,20].

The strength of the magnetic field is about 2 T which gives the U^{235}ions a cyclotron frequency of 130 kHz. The frequency of the oscillating electric field must be stable to an accuracy of 1 % or better to distinguish U^{235} form U^{235}. It is difficult to produce a magnetic field which is uniform to better than 1 % over a large area. Modern magnet technology can achieve this goal (The number of rotations in the helix done by the ions per second) only with relatively sophisticated and expensive solenoid designs. Because plasmas consist of charged particles, they shield electric fields. The characteristic shielding length of uranium plasma is a few micrometers.

This means in practice that even though the electric field can be applied in different ways, it must be applied indirectly.

To separate the two isotopes properly, the ions must be sent through several revolutions. The influence of the electric field on the U^{235}ions will average to zero while the U^{235}ions will get a substantially larger radius. There should be about 80 cyclotron periods before the ions reach the collector. If the separation tube is 1 m long this means that the axial velocity of the ions cannot be more than 1 600 m/s which corresponds to a temperature of about 75 000 K.10 This again means that the temperature of the plasma has an upper limit for the method to work. When the ions collide, they will be thrown out of phase with the electric field and loose energy. This puts a limit on the density of the plasma; the ion density must be below 106 ions per m3. This means that with a beam diameter of about a meter, using natural uranium as feed and assuming that all the U^{235} ions are collected, then about 70 kg of U^{235}could be collected per year

(The process needs large amounts of liquid helium11 to maintain the magnets in their superconducting state).

The process is a batch process, and has to be repeated to produce HEU from natural uranium feed. The enrichment factor is high, and the energy consumption of the process is quite low, about 330 kWh/SWU.

6.7.2 The Countries Used This Technique

The only countries known to have carried out serious experimental programs are the United States and France. The technique became a part of the Advanced Isotope Separation research and development program of the American Department of Energy in 1976, but development was stopped in 1982. The French developed their own version, and this program ended around 1990.

6.8 The Plasma Centrifuge Method

The information in this section is mainly from "Uranium Enrichment and Nuclear Weapon Proliferation" [20].

In the plasma centrifuge method, a plasma is set into fast rotations. As in a gas centrifuge, the heavier isotopes will gather closer to the wall of the centrifuge than the lighter isotopes. A main difference between the gas centrifuge and the plasma centrifuge is that the plasma container does not have to spin like the rotor in the gas centrifuge. This is a feature of the plasma centrifuge method which makes it easier to operate at a high rotational velocity. The bearings will not wear

(The Kelvin temperature scale has the same unit size as the Celsius scale, but it starts at absolute zero. This means that water freezes at 273 K.

Liquid helium has a temperature of 4.2 K (-268.9 °Celsius) at atmospheric pressure) out and the walls do not have to withstand the same amount of tension as in the rotating gas centrifuge. But as the rotational speed increases in the plasma

centrifuge, energy consumption and problems with collisions between ions and neutral particles increase.
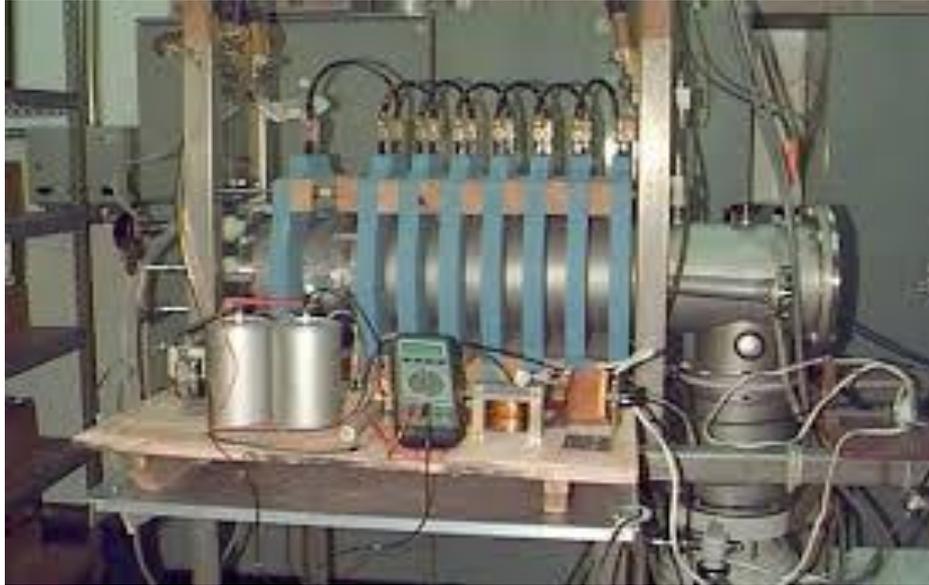
Figure (6.10) plasma centrifuge processing device.

The plasma is made to rotate by creating an electric discharge perpendicular to a strong magnetic field. As in the EMIS and the ion cyclotron resonance processes, the magnetic field makes the charged particles move in circles. By adding a radial electric field all the particles in the plasma (independent of whether the charge is negative or positive) will rotate in the same direction and very high speeds can be attained. In the rotating plasma, the different isotopes will have a velocity relative to each other. In collisions between ions, the heavier isotopes tend to gain speed and the lighter to slow down. This increases the velocity difference between the isotopes and enhances the separation effect [35].

The energy consumption of the plasma centrifuge method has been said to be about ten times that for gaseous diffusion, and even though this probably can be reduced, it may still be larger than that of mechanical centrifuges. One suggestion for reducing the energy consumption is to use a partially ionized plasma, then the rotation must be transferred to the neutral gas by collisions.
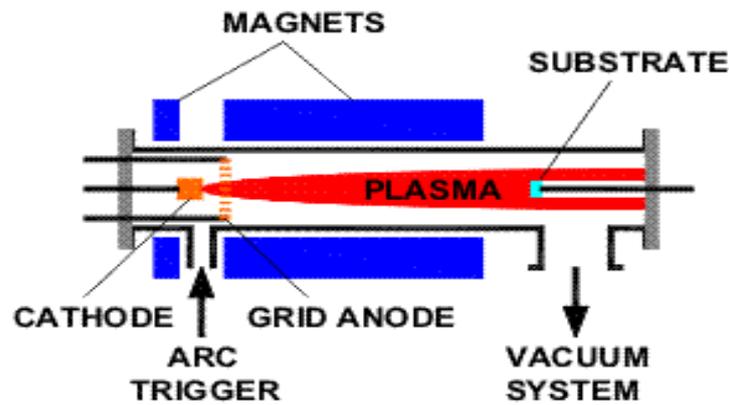
Figure (6.11) Schematic view plasma centrifuge processing.

The separation factor of a plasma centrifuge is not too great, but in some designs it may be possible to enhance it by setting up a counter current flow pattern as in the gas centrifuges.

Several countries have researched plasma centrifuges, also for separation of isotopes of other elements than uranium. There are a number of variations in the design, but no one has been used for enrichment of uranium to any degree worth mentioning.

6.8 Aerodynamic processes

There are two aerodynamic processes that are developed to a degree that makes them possible to use: The separation nozzle process developed in Germany and the vortex tube separation process developed in South Africa. They both rely on centrifugal forces to separate the isotopes, but unlike in the gas centrifuge process there are no spinning parts [33].

A stream of uranium hexafluoride gas is sent into a curved wall which bends it. During this bending, centrifugal forces press the heavier gas molecules closer to the wall than the lighter molecules. Then the stream is separated in one heavier and one lighter fraction.

The centrifugal forces are greater the larger the speed of the gas stream. To make use of this, the uranium hexafluoride is diluted by a light carrier gas such as hydrogen or helium. The velocity of sound is higher in a lighter gas than in a heavy gas, and this means that the gas can travel faster without energy being lost in sonic shock effects. Also the light carrier gas makes it easier for the isotopes of uranium to separate.

Both of the aerodynamic processes described below have a high energy consumption because of the high proportion of carrier gas relative to uranium hexafluoride. The processes are not considered economically competitive because of this high energy consumption.

A complicating feature of the aerodynamic processes is the handling of hydrogen. If the temperature is too high, hydrogen will react with uranium hexafluoride, so the temperature must be kept under control.
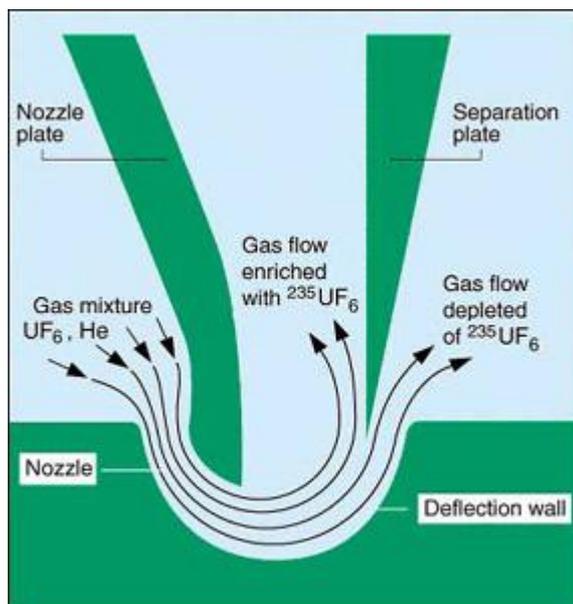
Figure (6.12) Schematic view of a separation nozzle. The smaller the nozzle is, the less turbulence there will be in the gas flow. The sketch is reprinted here with permission from Informationskreis KernEnergie, Berlin (www.kernenergie.de).

6.9 The Separation Nozzle process

The separation nozzle process, also known as the Becker nozzle process after its inventor, was developed in Germany and has been used both there and in Brazil, but both countries have since abandoned this activity. Gaseous uranium hexafluoride diluted by hydrogen or helium is compressed and directed along a curved wall at high speed, and the heavier molecules gather closer to the wall. The centrifugal forces on the gas in the nozzle can exceed those in a centrifuge by more than a thousand times. Then the stream is split by a knife edge, and the heavy and the light fraction is withdrawn separately [33].

The smaller the physical dimension of the separation nozzle, the more economic it is, and the radius of curvature may be as small as 10 μm. Production of the nozzles is done by photo etching of metal foils and then stacking them. A stage typically consists of a vertical cylindrical vessel containing the separation elements, a cross piece for gas distribution, a *gas* cooler to remove the heat of compression and a centrifugal compressor driven by an electric motor .

The energy costs for a separation nozzle plant is about 3600 kWh/SWU, and originate mostly in the re-compression of the depleted and enriched gas streams.

The separation factor of this method is bout 1.015 which is lower than for gas centrifuges but higher than for gaseous diffusion.

6.10 Gas Centrifuge Techniques

The enrichment of uranium involves the separation, to some degree, of a binary mixture. This can be represented diagrammatically:
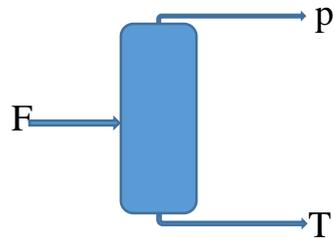
Where   the box represents a separating device

     F   is   a feed stream

     P   is   a product stream which is enriched in the desired isotope

     T   is   the tails stream which is depleted in the desired isotope.

Since no material is lost, clearly

$$F = P + T \tag{6.17}$$

So, the quantity of the desired isotope remains constant:

$$FV_F = PN_{P+}TN_T \tag{6.18}$$

Where N_F, N_p and N_T are the concentrations of this isotope in the feed, product, and tails streams, respectively.

The separating device has to expend some work to undertake this separation.

For a given feed flow, it is found that the work required is dependent on the concentration of the desired isotope in all three streams, F, P and T.

Unfortunately, there is no simple relationship, such as a ratio or difference, between these concentrations and the work expended [39].

The separation of a mixture into its component parts, even to a limited degree, reduces the disorder of the system. Conversely, mixing two substances increases the disorder. The thermodynamic measure of disorder is entropy. This concept can be used to derive a value function "V(N)"chicly is a measure of the disorder of a mixture having a concentration of the desired isotope N. The description of separative work as a change in the valuefunction of the components of the mixture has been attributed to Dirac in 1941 in an unpublished manuscript cited by Cohen

Since each of the three streams, F , P and T can be assigned a value or potential V(N)" then the work done can be characterized by the difference between the value of the feed and the resulting product and tails streams:

$$\delta U = PV(N_P) + TV(N_T) - FV(V_P) \tag{6.19}$$

Where δU is the separative work and other symbols have the same meaning as before.

It should be noted that the original derivation of separative work assumed that the separation factor, defined as the quotient of U^{235} abundance ratios in product and tails streams, was very small. For gas diffusion plants where the separation factor is less than 1.004, this is justifiable. Centrifuges, however, have much higher separation factors, perhaps 1.4, which makes the concept rather less appropriate.

6.10.1 Requirements for Separative Work

Most commercial power reactors using enriched fuel require product enriched to about 3 - 4%. Assuming natural feed (0.711% U^{235}) and the tails stream depleted to 0.25% U^{235}, then a typical 3.2% product requires

4.21 kgSW per kg product and 6.40 kgU feed. A typical 1,000 MWe reactor will require a reload of about 30 tU at each annual shutdown, i.e.

Figure (6.13). A bank of autoclaves used for feeding UF₆ to a plant.

120 tSW/a. First cores require about three times the quantity of fuel but at a lower average product assay giving a separative work requirement of around 230 tSW. These typical figures conceal a considerable variation in practice due to different technical designs, operating performance records and ratio of on-line to shut-down times.

6.10.2 Basic Separation Principles

(a)

The centrifuge exploits the small difference in mass which exists between the uranium isotopes in the molecules $^{235}UF_6$ and $^{238}UF_6$. Fortunately the separation

effect is proportional to the absolute mass difference of the molecules, *dM,* rather than to relative difference, dM/M, which is the case for gas diffusion.

   The separation effect is achieved by pressure diffusion through the gravitational field generated within the centrifuge rotor. In simple terms the concentration of the heavy isotope tends to increase towards the periphery of the rotor whilst that of the light isotope tends to increase towards the center. This method of separating isotopes was first suggested in 1919 by Lindeman and Aston, shortly after the discover)" of isotopes, with the prophetic words gives the derivation of the simple radial separation factor:

$$\alpha_0 = Exp(dMV^2/2RT) \tag{6.20}$$

Where   dM =   molecular weight difference

   V =   peripheral velocity

   R =   universal gas constant

   T =   absolute temperature

For the practical application to uranium enrichment, this factor is thus

Thus even a centrifuge of very modest speed has a separation factor greatly exceeding that of the gas diffusion process which has an ideal value of 1.0043 [33.36].

The above expression also indicates the desirability of operating at a lower at least moderate temperature; nevertheless, an adequate temperature margin must always be maintained to ensure that the process gas UF6 does not condense

$$\alpha_0 = Exp\left(\frac{dMV^2}{2RT}\right)\sqrt{2l}\,/d \tag{6.21}$$

Where

   l = rotor length

   d = rotor diameter

 (b)

A good model for understanding the way in which a centrifuge separates a mixture of isotopes is to imagine a sample of gas in a room under the influence of gravity. Since each molecule is being pulled downwards, a certain amount of work must be done to lift the molecule to some height h.

This work represents an increase in the molecule's 'potential7 energy, and this change is given simply by

$$PE = mgh \tag{6.22}$$

 Where m is the molecule's mass and g is the acceleration due to gravity, equal to 9.8 m/s^2 at the Earth's surface. If the temperature in the room is the same everywhere, molecules at all heights have the same average kinetic energy, but

those near the ceiling have a higher potential energy than those near the floor. The same theory that predicts the equipartition of energy predicts that higher energies are less probable than lower ones. It predicts that the density of particles near the ceiling will be less than the density near the floor by the factor

$$N(h)\big/N(0) = Exp - (^{mgh}\big/_{RT})$$ (6.23)

Where R is called the gas constant (8.3 joules per degree Kelvin), and T is the temperature measured from absolute zero (0 K = -273°C). If two different species are present in the mixture, then an equation like 6.2 can be written for each one and the ratio taken:

$$\left[\frac{N_1(h)\big/N_1(0)}{N_2(h)\big/N_2}\right] = Exp - \left[(m_1 - m_2)^{gh}\big/_{RT}\right]$$ (6.24)

If the factors on the left are slightly rearranged, the ratio can be seen to be equivalent to the ratio of the relative isotopic abundances at the heights h and zero. So this equation can be rewritten as

$$R(h)\big/R(0) = Exp - \left[(m_1 - m_2)^{gh}\big/_{RT}\right]$$ (6.25)

This calculation suggests that it should be possible in principle to separate isotopes of uranium by filling a room with UF6, allowing it to come to thermal equilibrium, and then simply skimming off the top portion of the gas. Experiments similar to this were in fact performed over 80 years ago. However, this is not a practical process for uranium, since the effect is extremely small. For example, in a room 3 metres high at normal temperature the ideal separation gain for UF$_6$ would be only 3.4×10^5, Over 100 times smaller than for gaseous diffusion.

It was recognized very early that a rapidly rotating centrifuge could provide a much stronger force field and therefore increase the separation gain many times. In a centrifuge the acceleration of gravity is replaced by the 'centrifugal' acceleration, can be shown to be:

$$R(r)\big/R(0) = Exp - \left[(m_1 - m_2)(\omega r^2) / 2RT\right]$$ (6.26)

Where r is the distance from the center of the centrifuge, and ω is the angular velocity in radians per second. Notice that the sign of the exponential factor has changed, implying that the isotopic abundance of the lighter species increases towards the center of the centrifuge. The wall of the centrifuge is then analogous to the floor in the previous example.

To see what equation (6.26) predicts for uranium enrichment consider a hypothetical centrifuge with a radius of 10 cm and an angular frequency of

800 revolutions per second ($\omega = 800 \times 2\pi = 5\,000$ rad/s). The acceleration at the wall of this centrifuge is 2.5 X 10^6 m/s^2 or more than 250 000 times as strong as gravity. Using $m_1 = 0.349$ kg and $m_2 = 0.352$ kg and assuming a temperature of 330 K (57°C) the ratio of abundance between the center and outer wall is found to be 1.147. The ideal value of g $= 0.147$ for this centrifuge can be seen to be over 16 times as large as the corresponding value for gaseous diffusion. Note also that the separation gain depends on the simple difference between the isotope masses, not on their ratio. This means that the advantage of the centrifuge over gaseous diffusion improves as the isotopic masses increase.

The essential characteristics of the three-pole separation system may thus   be summarized as:

1. Cylinder rotating at the highest practical peripheral speed;
2. means of introducing feed material, UF$_6$, towards the center of therotor (a three-pole machine); sometimes two feeds of differing concentrations are introduced at the ends, giving a four-pole machine of potentially higher efficiency;
3. elementary separation effect, ceo, m the radial direction;
4. countercurrent circulation giving an enhanced separative effect, a/3, in   the axial direction;
1. means of withdrawing the enriched product, P , and depleted tails, T, from the ends of the rotor.
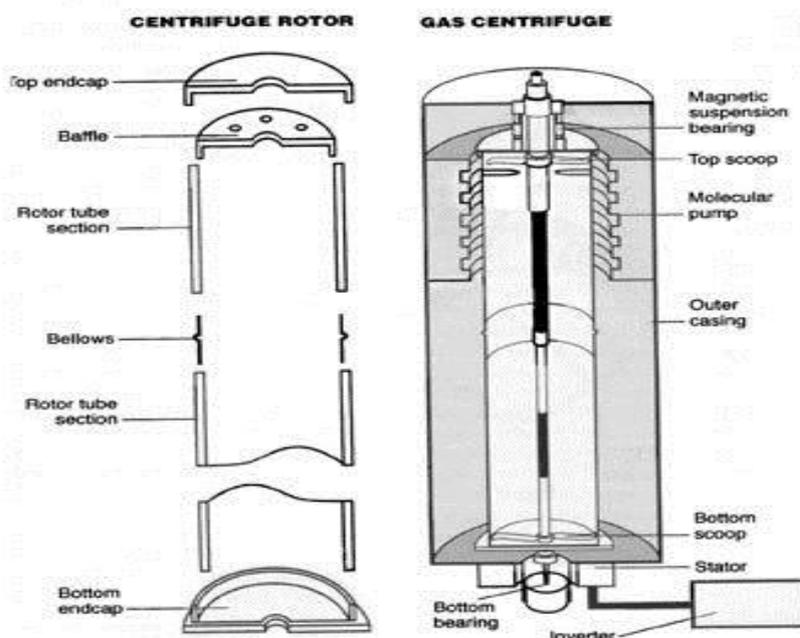
Source: Albright, D. and Hibbs, M., 'Iraq's shop-till-you-drop nuclear program', *Bulletin of the Atomic Scientists*, vol. 48, no. 3 (Apr. 1992), pp. 32 and 33.

Figure (6.14) Schematic view gas centrifuge.

6.10.3 Separative Power:

$$\delta U = \rho . D \left[\frac{\Delta M V^2}{2RT}\right]^2 . \frac{\pi}{2} L \qquad (6.27)$$

Where:

ρ =   gas density

D   = coefficient of self-diffusion

ΔM = Difference of molecular weights of isotopes
Being separated

V   =   peripheral velocity of rotor

R   = universal gas constant

L   =   rotor length

This clearly shows:

1. The strong dependence on speed since $\delta U \propto V^4$.
2. The proportionality of output to rotor length,
3. The independence of output to diameter.
4. The independence of output to gas pressure since $\propto \frac{1}{\rho}$.
5. The desirability of operating at a low, or at least moderate, temperature.

6.10.4 Cascades

In addition to the centrifuge itself, industrial deployment required the development of suitable cascades and the associated theoretical basis for their design.

The enrichment of uranium to, say, 3.5% for reactor use and the associated depletion of the tails to about 0.25% ^{235}U implies an overall separation factor of 14. The separation factor $\alpha\beta$ of a centrifuge is typically $1.5 < \alpha\beta < 3$. Consequently it is necessary to arrange a number of centrifuges in series in order to obtain the necessary overall factor by compounding the individual centrifuge separation factor. Each step in such an arrangement is termed a "stage". The stages are connected such that the product is fed forward to become the feed of the stage above whilst the tails is fed back to become the feed of the stage below.

At each stage there must be a balance of material flowing into and out of the stage and a balance of isotope flows. As the material becomes more enriched towards the product end of the cascade and more depleted towards the tails end of the cascade, the stage flow decreases. If all centrifuges are [41].

to be made identical, which is usual for manufacturing convenience, then in order to keep each machine operating close to its optimum working point, it is necessary to group centrifuges in parallel in order to maintain the required stage flows. The theoretical design basis for such an arrangement has parallels in the chemical industry and was applied at an early stage to the design of diffusion plants and later to centrifuge plants.

By way of example, Whitley shows the stage arrangement for a centrifuge having equal enrichment and depletion factors $\alpha = \beta = 1.2$ connected to produce 3% product and 0.3% tails.

For comparison, a gas diffusion plant would require in excess of 1000 stages for the same duty rather than the 12 used in the centrifuge plant. However, the centrifuge, being a low-throughput device, would require a number of parallel cascades to provide an industrially-useful output, whereas a gas diffusion plant would comprise a single cascade only.

Figure (6.15) UF$_6$ Feed Station.

Figure (6.16) Centrifuges in the Cascade Hall (a)

Figure (6.17) Centrifuges in the Cascade Hall (b)

6.10.5 Large Scale Plant Design
Basic Principles

The translation of the basic technology, described above, into large-scale

Industrial plants is a significant achievement in process engineering. Important criteria in plant, design are:

1. Safety : environmental and operator protection as well as preservation of the equipment investment
2. Reliability : ability to operate without breakdown for many years and to be tolerant of external malfunction to, saj', electrical power supplies
3. Flexibility: easy operational adjustment to suit customer requirements for product assay, tails assay, and quantity at short notice
4. Economy: competitive and robust economics are essential not only for completed plants but during the critical build-up phase where capacity, and hence investment, must be matched to business secured.

6.10.6 Process Description

A two-stage pressure reduction is undertaken and UF_6 distributed to the centrifuge cascades at sub-atmospheric pressure, see Figure (6.17). The cascades undertake the full separation task and are small in size relative to plant capacity.

The centrifuges are driven by means of a medium-frequency A.C. power supply individual to each cascade, and are temperature regulated via a centralized cooling system. Enriched and depleted material streams have substantially similar take-off systems. Cold traps, or desublimers, condense the UF_6 as solid. When full, the gas stream is directed to an adjacent unit and the desublimer is heated to transfer the

material to standard 30" or 48" containers, as appropriate, in the collection station, see Figure 10. The UF_6 collection stations are simple enclosures wherein the containers may be chilled in order to collect the material. Air cooling is used for product stations and water chilling for tails.

The cascades are designed to have a very high efficiency over a certain range of product and tails assays [40].

In addition to the enrichment facility proper, each Urenco enrichment plant

is also provided with a central service building CSBJ. This facility provides a range of technical services, the most important being the blending, weighing, sampling and analysis of product. Although it is the aim to produce the correct product assays in the plant, blending may be required to meet special or short-notice orders. The CSB is equipped with the blending facilities to suit an appropriate proportion of the throughput. The CSB also accommodates container inspection, washing and testing facilities, general stores, plant maintenance workshop and a decontamination facility.

These functional areas can be seen in the schematic arrangement, Figure (6.14) centrifuge workshop is required since a "zero maintenance" approach is applied.

Centrifuges have an average expected life greater than ten years and cascades are tolerant of failed machines, which are left in situ The installation of many thousands of centrifuges in a single plant poses unique problems to the engineer. An important design problem is the security of mounting of the centrifuges in a compact and economic way such that in the event of a failure (crash) the considerable energy release sch : does not disturb adjacent machines leading to subsequent failures in a "domino" effect. The complexity of the distribution networks for process gas, cooling and electrical power to each machine is an important aspect of the optimization of the design concept.

6.10.7 Main Plant Systems

The interrelation of the various process systems is   The principal systems are:
1. Centrifuges
2. Cascades
3. UF_6 feed, product and tails
4. Control and instrumentation
5. Auxiliaries
6. Electrics
7. Building

6.11 Thermal diffusion
The main principle

The thermal diffusion process is based on the same principle as the counter current flow pattern in the gas centrifuges, and employs uranium in the form of uranium hexafluoride. In this process a thin container of gaseous or liquid uranium hexafluoride is oriented vertically (see Figure (6.18)). One side is then heated and the other side is cooled, and this gives rise to a thermal convection current with an upward flow along the hot surface and a downward flow along the cold surface. Under these conditions, the lighter U^{235} gas molecules will diffuse toward the hot surface, and the heavier U^{238} gas molecules will diffuse toward the cold surface.
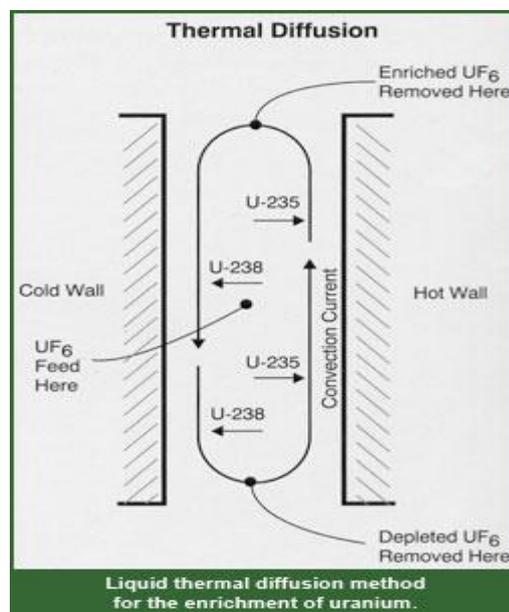
Figure (6.18) liquid thermal diffusion method for the enrichment of uranium.

The result of these motions is that the U^{235} molecules concentrate at the top of the film, and the heavier U^{238} molecules concentrate at the bottom.

6.12 Chemical separation methods

Two chemical processes have been developed to a point where they could be considered possible to use for the enrichment of uranium. The CHEMEX process was developed in France, and an ion exchange method has been developed in Japan. The chemical approaches to the separation of uranium isotopes are attractive because of their low energy consumption; there is no need for powerful compressors or pumps. Furthermore, these processes are based on standard chemical engineering technology, which makes them favorable.

Both processes are based on the fact that U^{235} is more likely than U^{238} to be in a higher oxidation state. This means that if natural uranium is present in different chemical states or compounds, then there will be an accumulation of U^{235} in the higher oxidation states. If it is possible to separate the states (for example if they are different types of molecules that do not mix very well) then the compound which is enriched in U^{235} can be extracted. For the separation effect to be the greatest, the two compounds used should have uranium in as different oxidation states as possible, and the temperature should be as low as possible. Unfortunately these are criteria which also make the process go slower and lead to the need of good catalyst (A catalyst is a substance which facilitates a chemical process without being consumed itself).

6.12.1 The general concept

A separation stage consists of mixing of the two compounds of uranium for a long enough time that chemical equilibrium may be established. This can be done by letting the two substances flow in opposite directions in a column. The main difficulty of chemical enrichment methods is the reflux problem. When the substances emerge at the ends of the column, some can be extracted as product and tail, but much has to be sent back into the column to maintain the counter current flow pattern. Before they can be sent back into the column, the enriched substance has to be [39,40].

Transformed into the form of the depleted substance and vice versa. This is a non-reversible process and the main source of power consumption in the chemical enrichment processes. Even a small material loss in the reflux processes can reduce the efficiency of the plant significantly.

6.12.2 Basic principles

The chemical-exchange method of isotope separation depends on the very small tendency of different isotopes of an element to concentrate in different molecules when there is an opportunity for exchange between the molecules. As an illustration consider two uranium compounds AU and

BU which are mixed together and allowed to come to equilibrium. In general each compound will have two isotopic forms: A U^{235} and A U^{235}; and B U^{235} and B U^{238}. Therefore, in any mixture of the two uranium compounds there will be four different species present. If these species are chosen properly, they can exchange uranium atoms. Chemists depict such an exchange equilibrium in the form of an equation:

$$A^{238}U + B^{235}U \rightleftharpoons B^{238}U + A^{235}U \qquad (6.28)$$

Where the double arrows imply that the reaction can proceed in both directions.

When the system is in equilibrium each species is present in a certain concentration, usually measured in moles per liter and denoted by the symbol [AU]. The equilibrium is characterized by an equilibrium constant K which gives the relation among the four concentrations [37].

$$K = [A^{235}B][B^{238}A]/[A^{238}B][B^{235}A] \qquad (6.29)$$

The chemical equation is customarily written to make K greater than 1, so this implies that in equilibrium the product of concentrations with U^{235}in and U^{235} in B is greater than the product with U^{235} and U^{235} interchanged. Therefore if the two compounds AU and BU initially contain identical isotopic ratios, after the equilibrium is established compound A will be slightly enriched and B slightly depleted in U^{235}.

Unfortunately it is not easy to explain why the value of K is different from 1. Indeed, the equipartition theorem of statistical mechanics, which was used to explain the separation effect in gaseous diffusion, predicts that K should always equal 1, and that no isotope separation should occur.

The fact that it does occur is explained by the theory of quantum mechanics and the connection between the energy of a molecule and its vibrational frequencies. No such connection exists in the older classical physics.

The quantum theory states that the unit of vibrational energy of a molecule is proportional to its frequency, and that the molecule can only absorb energy in amounts which are a multiple of this basic unit. This means that the classical equipartition theorem cannot be valid for such a system, since the theorem requires that any amount of energy, no matter how small, which is added to the system must be shared equally among all the molecules. This situation exists to a good approximation if the temperature is high, but as the temperature is reduced the equipartition principle breaks down. In practice this results in the lighter isotope tending to concentrate in the more loosely bound molecule, while the heavy isotope is more likely to be found in the tightly bound molecule [36].

This explanation immediately suggests two requirements on the compounds for a good chemical-exchange separation:

 1. The two compounds should be very different, with the uranium tightly bound in one and loosely bound in the other. Free uranium ions fit the second requirement best of all.

 2. The equilibrium should be established at the lowest possible temperature to maximize the separation effect. Unfortunately, both of these requirements are incompatible with the requirement of easy exchange of the uranium. Two very different compounds do not exchange components effectively, and reducing the temperature reduces the rate at which equilibrium can be established.

This problem can be dealt with by using suitable catalysts. These are compounds or materials which do not participate directly in the chemical reaction but whose presence in some way facilitates or speeds up the reaction. The ultimate success of the French and Japanese chemical enrichment processes seems to have been largely the result of the discovery of suitable catalysts. These will be discussed in more detail below. The equilibrium constant K of equation (6.29) is identical to the single-stage separation factor q. This can be demonstrated as follows [37].

Consider a stage to be a single operation of mixing the two compounds AU and BU, with both having equal proportions of light and heavy uranium isotopes. After equilibrium is achieved the compound AU will be slightly enriched in the light isotope and BU will be slightly depleted. Compound

AU can therefore be called the product, and BU the tails coming out of this stage. The relative isotopic abundances R_P and R_w in the two compounds are simply the ratios of the concentrations of the two isotopic species in each compound:

$$R_{P=}[A^{235}U] \big/ [A^{238}U] \tag{6.30}$$

$$R_{W=}[B^{235}U] \big/ [B^{238}U] \tag{6.31}$$

and the separation factor is given by:

$$q = R_P/R_W \tag{6.33}$$

Inserting equations (6.31) into (6.30) and rearranging the factors reproduces equation (6.312) and shows that:

$$q = k \tag{63.33}$$

This relationship holds as long as each compound contains only one atom of uranium. In more complicated compounds the formula must be modified, but such refinements are not needed for a basic understanding

For isotopes of light elements the value of q can be quite large, and in fact isotopes of hydrogen, boron and nitrogen are most effectively separated by chemical means. For uranium, however, q is limited to values at or below 1.003, and a great many stages are required to achieve useful enrichments. For example, if q = 1.002, over 2 700 stages are required in an ideal cascade to produce 3 per cent enriched product with 0.2 per cent tails. But chemical separation 'stages' are quite a bit simpler than the other types discussed so far, and this number is not as prohibitive as it may seem.

A chemical separation stage consists simply of the thorough mixing of two substances for a sufficient time to allow chemical equilibrium to be established. This is most efficiently done in a device called a 'countercurrent column' in which one of the compounds is carried upward and the other downward. Any level in the column is associated with a given isotopic composition of both compounds, so that only compounds of the same composition are brought into contact. The very slight isotopic transfer causes one compound to be slightly enriched and the other depleted as each moves on to the next contact.

For an efficient exchange the two compounds AU and BU must be both easily brought together and easily separated. This is accomplished most efficiently if the two compounds are in different phases. One might be a liquid and the other a gas; or both could be liquids which are immiscible; or one could be a liquid or gas and the other a solid. The liquid-gas system operates very much like a standard fractional distillation column, the liquid-liquid system like a solvent extraction process, and the liquid solid system works like an ion-exchange column or 'chromatography'. All of these techniques are widely used and very well understood by the petroleum and chemical industries, and this great wealth of experience adds to the attractiveness of chemical separation. Another attractive feature is the potentially very low energy consumption of the process. Since chemical exchange is an equilibrium process there is no need for powerful compressors or pumps for the preparation of phases for contacting.

These advantages are counterbalanced by the problem of processing the two compounds at the ends of the column, the problem of 'reflux'. To illustrate this problem, suppose that compound A is moving upward in the column and B downward (see figure 6.14). This means that the highest enrichment in ^U is at the top, and the enriched uranium emerges from the top in the compound AU. Some of this can be removed as product (or sent on to another column), but most of it must be refluxed to maintain the large countercurrent flow. However, before it can be sent back down the column the AU must be converted to BU. (The opposite must be done at the lower end, the tails reflux.) This is in general a non-reversible chemical   process, which can require large amounts of energy and which must be done very carefully to avoid losses of enriched uranium. Because the reflux ratio is generally very large (i.e., the amount recycled is many times the amount extracted), even a small percentage loss of material in the reflux reaction can significantly reduce the efficiency of a plant [8,40].

With this general introduction to the theory of chemical enrichment it is now possible to consider the two most promising processes in more detail. The first is the Japanese process which depends on an exchange of uranium between a liquid solution and a finely divided ion-exchange resin. The second is the French solvent

extraction process which uses an exchange between two immiscible liquid phases, one aqueous and the other organic. A third process, studied extensively in the USA, but apparently considerably less developed than the first two, is also a liquid-liquid process based on exchange between solutions of UF_6 and $NOUF_6$.

6.12.3 French Process (CHEMEX)

In the French process, two different compounds including uranium are dissolved in different liquids. These liquids are immiscible18 in order to make possible the later separation of the compounds; one is in an aqueous phase, and one is in an organic phase.

The two liquids flow through each other in a column. The liquids are agitated so that the organic one forms small droplets to increase the reaction surface; this is done either by the use of an external mechanical device or a series of reciprocating discs. Limiting factors are the rate of the chemical reaction and the rate of diffusion in and out of the oil droplets for the two chemical species. The chemical reaction rate can be sped up by changing the uranium compounds or by the use of catalysts and raising the temperature. Some compounds from the group known as "crown ethers" have good qualities for uranium enrichment because they both extract the uranium from one liquid phase to another and enhance the separation effect. The diffusion rate of the uranium compounds in and out of the oil droplet can be made higher by decreasing the size of the droplet. But there seems to be a practical limit at a diameter of a few millimeters, and this makes the process slower than the Japanese process discussed below.

The French enrichment facility is proposed to be constructed of two identical columns connected in a way that reduces the equilibrium time. The standard dimensions of the exchange column in the CHEMEX process is 1.6 m in diameter and 35 m high, and the capacity is 3000–5000 SWU per year. The concentration of uranium is greater in this process than in the Japanese process. The inventory is about one order of magnitude larger than that for a gaseous diffusion plant of about the same capacity. There is also a need for large volumes of chemicals. Some of them, such as the crown ethers, can be expensive [26].

At the ends of the column, the two uranium compounds have to be transformed into each other, in order to have reflux. The chemical converters must be very efficient; they are the main sources of energy consumption in this process.

The uranium compounds, the catalysts and the reflux reactions are kept secret. The components needed in the process are made from plastic materials because of corrosion problems. The estimated energy consumption is about 600 kWh/SWU.

6.12.4 Japanese Process (ACEP)

The Japanese ion exchange process known as the Asahi Chemical Exchange Process (ACEP) has been under development since 1972. It is based on a solid uranium adsorbent which has been Developed by Asahi Chemical; this selectively adsorbs19 hexavalent uranium. The breakthrough of this process was the discovery of a uranium adsorbed of high reaction speed. The adsorbed is a resin20 in which the individual particles have the forms of spheres with a porous surface to increase the contact surface. It has the function of holding one of the two uranium compounds on its surface.

First the resin is charged with an oxidizing agent. A solution containing a uranium compound in which uranium is in the 4+ oxidation state is introduced. Then the uranium is oxidized to the 6+ state and replaces the oxidizing agent on the resin. This process will leave a sharp boundary between the part of the column where the resins are occupied by the oxidizing agent and uranium, this boundary will move slowly down the column when more uranium solution is added at the top of the column. After some time a new solution is added at the top of the column, this one containing a reducing agent. This will act on the uranium absorbed on the resin and reduce it back to the 4+ oxidation state and then replace it on the resin. The 4+ uranium goes into the solution and continues down the column with it. The 4+ uranium will then come into contact with the 6+ uranium on the resins further down the column, and the isotopic change will take place. There will eventually be a higher concentration of U^{235} in the compound adsorbed on the resin than in the solution. The portion of the column where the isotope exchange takes place is well defined.

Because also the reducing agent forms a sharp boundary with the uranium compound [35].

The process is carried out by the use of four columns; the actual ion-exchange process takes place in the first three columns which are connected in series, and the recharging of the agents is done in the fourth. Each column should be a few meters high. Very little energy is needed in the ion-exchange columns unless the temperature has to be raised. The main energy consumption comes from recharging the oxidizing and reducing agents and the pumping of the recycled solution. The energy consumption of the process is estimated to be about 150 kWh/SWU. Because of good exchange kinetics, the ACEP process requires smaller volumes than the CHEMEX process.

To find suitable resins, oxidizing and reducing agents, processes for recharging these as well as a suitable catalyst for the exchange process has taken many years.

6.12.5 Third process

There is also a third process that has been studied by several countries. It appears to be attractive, but there are problems related to developing it past the laboratory stage. The problems seem to be mainly large energy costs and losses of material in the recycling of the compounds. The development of this process seems to be far behind the two processes discussed above. It makes use of $NOUF_6$ dissolved in dry hydrogen fluoride and UF_6 dissolved in Freon or some other saturated fluorocarbon in the liquid phases. It is the execution without large energy costs and (When something is adsorbed it forms a layer on the surface of the absorber. 20 A resin is a substance that acts as an ion exchanger. It holds ions in its structure and is therefore able to hold ions with the opposite charge to its surface. When new ions are adsorbed, old ones are released.) losses of material of the reaction that recycles $NOUF_6$ which seems to be the biggest challenge.

6.13 Conclusion

In this broadcast I studied and discussed the uranium enrichment techniques used in many countries, which have been obtained from enriched uranium at different rates and low rates of enrichment.

It should be noted that the enrichment of uranium is linked to the engineering, economic, political and technical aspects of complex, so this study is characterized by discussion of the aspects related to physical concepts only.

All the results in this broadcast are drawn from empirical experiments and scientific studies in the United States, France, Japan, German, Russia, the United Kingdom and the Swiss Institute of Physics Research, which have been referred to as references.

In order for the fertilization process to be studied, we need relatively long periods of time. The details of the enrichment techniques are highly confidential and are only available in the major industrialized countries, so it is difficult for researchers to obtain accurate information and carry out direct experiments.

6.14 Recommendation

Sudan has a large contingent of natural raw uranium, which makes it eligible to enter the nuclear industry without the need to import from abroad if the development of a special program to take advantage of it in the near future to do so:

I recommend the provision of specialized scientific material in the field of uranium enrichment at the level of universities and related scientific institutes, and the establishment of periodic scientific lectures and workshops for the exchange of information and the provision of references and books and provide opportunities for testing.

In addition, opportunities for cooperation between national institutions and their foreign counterparts in the uranium enrichment industry should be created as much as possible.

Laboratory simulations should be provided to help students and researchers (simulators, for example) understand basic concepts and theories of uranium enrichment and production.

Through my research and research on uranium enrichment techniques, I believe that the most suitable model that can be applied in Sudan is the method of enrichment by which method.

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