Seventh Conference of Nuclear Sciences & Applications
6-10 February 2000, Cairo, Egypt

NS-2 Proposal for Analysis of the Safeguarded Nuclear Materials ^{235}U and ^{239}Pu By Delayed Neutrons Technique

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ABSTRACT

This paper introduces, describes and initiates a very sensitive and rapid non-destructive technique to be used for analysis of the safeguarded nuclear materials ^{235}U and ^{239}Pu. The technique is based on fission of the nuclear material by neutrons and then measuring the delayed neutrons produced from the neutron rich fission products. By this technique, fissile isotope content (^{235}U) can be determined in the presence of the other fissile (e.g. ^{239}Pu) or fertile isotopes (e.g. ^{238}U) in fresh and spent fuel. The time consumed for analysis of bulk materials by this technique is only 4 minutes. The method is also used for analysis of uranium in rock, sediment, soil, meteorites, lunar, biological, urine, archeological, zircon sand and seawater samples. The method enables uranium in a sample to be measured without respect to its oxidation state, organic and inorganic elements.

Keywords: Nuclear Materials / Safeguards / Delayed Neutron Activation Analysis.

INTRODUCTION

The ratification of the nuclear Non-Proliferation Treaty (NPT) makes it necessary to develop nuclear safeguards methods and measures to detect and prevent the diversion of fissionable material from the front-end and back-end of the nuclear fuel cycle.

Analysis of the safeguarded nuclear materials is carried out by two techniques: Destructive (chemical) assay (DA) and Non-Destructive assay (NDA)[1,2]. Delayed neutron activation analysis (DNAA), as one of the NDA techniques, has been employed in several part of the nuclear fuel cycle, including borehole logging as a uranium exploration tool and analysis of uranium ore samples. Delayed neutrons (dn) have provided a very useful signature for Non-destructive assay of fissionable materials (^{235}U and ^{239}Pu) and have served as the major Work-Horse of NDA by active interrogation technique in safeguards and nuclear materials management program [1-4]

Egypt has all the capabilities to install this technique to be used for assaying of the safeguarded nuclear materials and environmental samples for safeguard purposes.

The author had been using this technique for analysis of uranium in uranyl nitrate solutions and in environmental samples.

THEORETICAL ASPECTS OF THE METHOD

The neutrons produced in nuclear fission can be divided into two groups; prompt neutrons and delayed neutrons [2,5,6]. The prompt neutrons are emitted not later than 10^{-13} seconds after fission of the nucleus and form about 99% of the total number of neutrons. Delayed neutrons represent only a small part (<1 %) of the total number of fission neutrons. They are emitted at the end of the β-and K-transformations of fission products; 10^{-1} - 10^{2} second after fission [2,6,7,8,9].

Delayed neutron yields are function of the particular fissioning isotope and the energy of the interrogating radiation [1]. The total delayed neutron fraction for fissioning induced by thermal and fast neutrons are shown in table 1 [10]

Fission	Thermal			Fast	
Isotope	^{233}U	^{235}U	^{239}Pu	^{238}U	^{232}Th
Total dn fraction	0.0026	0.0065	0.0021	0.0148	0.0203

The correct explanation of the delayed neutron (dn) has been given by Boher and Wheeler [11]. They supposed that a proportion of the neutron-rich isotopes (known as delayed neutron precursors - Fig.1) formed in fission had sufficiently large β-decay energies to populate excited states in the daughter nuclei above the neutron binding energies of these nuclei (called neutron emitters). Hence dn emission could occur, the neutron activity having the β-decay half-life of the precursor nuclide. Excited states in the emitter above the neutron binding energy are populated by β-decay of the precursor. These levels can then deexcite in one of three ways [4]; gamma emission , $_0^1n$ emission to the ground slate of the final nucleus and neutron emission to an excited state of the final nucleus and then emission to ground state.

Most applications of dn use an approximate temporal groups representation of measured aggregate data. These groups (usually 6 according to their half-lives of less than 1 to 55 sec.) have no true physical basis but rather originated as fits to measured delayed neutron emission following fission pulse and saturation irradiation experiments in critical assemblies [1,4.12,13]. The delayed neutron groups, their half-lives and example of dn precursors are given in table 2 [10]

dn Groups	Half-life (sec.)	dn precursors
1	55	^{87}Br
2	22	^{88}Br , ^{137}I
3	6	^{89}Br , ^{138}I
4	2	^{90}Br, ^{139}I, ^{144}Cs
5	0.6	^{140}I
6	0.2	^{93}Br

DELAYED NEUTRON EMISSION PROBABILITIES

Amiel and Feldstein, using statistical consideration, obtained the simple expression [3,14];

$$\mathbf{Pn = C\,(Q_\beta - B_n)^m}$$

Where, Pn is the n emission probability

Q_β is the decay energy of the precursor

Bn, is the neutron binding energy of the emitter

C and m are constants

This expression assumes that the level density is a function of $(Q_B - Bn)$ and that γ-ray competition due to spin and parity effects can be ignored [13]. Delayed neutron emission probabilities are also given in recent publications [15,16]

Separation of isotopic responses are achieved by varying the average energy of the interrogating neutrons using target shields consisting of shells of W, Pb, graphite and polyethylene [18].

Sub-threshold and super-thresholds neutron interrogation makes it possible to separate directly the response of the fissile isotopes (e.g. ^{233}U, ^{235}U, ^{239}Pu) from the fertile isotopes (e.g. ^{238}U and ^{232}Th)[18] (see fig.2,3).

To discriminate between ^{235}U and ^{239}Pu isotopes, their different energy-dependent fission cross-sections are used (see fig.2,3). As a matter of fact the ^{239}Pu has a pronounced resonance at 0.3 eV while ^{235}U fissions by 0.025eV thermal neutron [1,6,17,18].

EXPERIMENTAL WORK

MATERIAL OF THE METHOD

(1)$_0^1$n source (^{252}Cf or n generator or nuclear reactor) [3,17,19]
(2)Pneumatic system (Rabbit system) in the case of bulk materials
(3)The nuclear materials to be analyzed.
(4)Neutrons counter with its associated electronics (Power supply, Amplifier~..).

METHOD OF THE ANALYSIS

The following typical steps were carried out for analyzing of bulk nuclear materials samples with using nuclear reactor as a neutron source;

1-Very minute mass (ug to g) of the sample was weighted and packed in polypropylene vial.
2-The vial was put in a polystyrene capsule to withstand the thermal and the radiological stresses inside the reactor core.
3-The polystyrene capsule was transported automatically to the reactor core to be irradiated for 2 minutes at a thermal flux of 1.7×19^{12} n / cm^2 s.
4-the sample was brought from the reactor core to the measuring position in the neutron counter after 20 sec. delayed time (transportation time) using a pneumatic system. The twenty seconds delay time was adjusted to avoid any contribution from the non-fission neutrons produced from the reaction [20].

$$^{17}O(n,p) \rightarrow {}^{17}N \xrightarrow{\beta} [{}^{17}O]^{*} \xrightarrow{4.4\,Sec} {}^{16}O + n$$

5-The β-delayed neutrons resulting from the uranium in the sample were counted for one minute by using neutron counter (assembly of He-3 proportional tubes).
6-The β-delayed neutrons resulting from different standard uranium concentrations were used for drawing a calibration curve (similar to fig. 4) [20, 21]
7-Finally concentration of uranium in the sample was deduced from the calibration curve.

The lower detection limit of uranium to be obtained with the abovementioned conditions is 3ng.

With some modifications, the analysis of the bulk materials (e.g. in fuel fabrication and reprocessing plants) and item materials (e.g. in reactors) can be carried out by using neutron generator or ^{252}Cf neutrons source [3,17,19,22, 23,24,25]

SOURCES OF ERROR

The sources of error are mainly due to [1];

1) Calibration standards
2) Quantity of special nuclear materials
3) Background

Calibration standards must resemble samples as closely as possible in order that the response per unit mass of special nuclear material (SNM) for the sample is the same as that predicted by the

calibration curve. Increasing quantity of SNM can result in self-attenuation, and the response will then be a non-linear function of the SNM quantity. Counting background that must be subtracted or otherwise accounted for. Contribution to this BG include radiation from the sample such as neutrons from (α ,n) reaction and spontaneous fission.

APPLICATIONS OF THE METHOD

The method has wide applications in different fields. Some of these applications are;
1-Augustson et al has measured the ^{235}U content in a spent MTR fuel element (the same fuel type of the Egyptian second reactor) using the delayed neutron yield technique. The dn measurements agreed within 1.5% with the U content as predicted by reactor burn up calculation [23].
2- ^{235}U and ^{233}U content in HTR fuel elements have also determined by delayed neutron analysis [26]
3-DNAA has also been used for analysis of uranium and thorium in aqueous solutions[20]
4-The concentration of uranium in seawater has been determined using DN. The method enables uranium in a sample to be measured without respect to its oxidation state, organic and inorganic elements. The sensitivity of the method is 2 ng/ml with an uncertainty of ~15% [27]
5-Monitoring of unauthorized movement of special nuclear material (nuclear materials smuggling) by active neutron interrogation package monitor [19]

TYPES OF THE NUCLEAR MATERIALS IN EGYPT

The nuclear materials in the Egyptian nuclear facilities can be classified and summarized as given in figures 6 and 7.

These nuclear materials can be with high accuracy verified by DNAA technique. The unburned ^{235}U and traces of build up ^{239}Pu in spent fuel of the first research reactor are mainly estimated based on the reactor operation parameters (flux, power, burn-up ...). By using the DNAA technique, they can be experimentally determined with taken into consideration the safety precautions to handle the spent fuel.

The method can easily detect what is known as *S/R* difference (the difference between (imported) shipped (S) and received (R) nuclear materials).

Recently, collection of environmental samples (e.g. air, water, vegetation, soil, smears) at a set of locations specified by the agency for the purpose of assisting the IAEA to draw conclusions about the absence of undeclared nuclear material or nuclear activities over a wide area is considered a part of the agency routine inspection (the additional protocol measures – INFICIRC/540- article18-g) [28]. The DNAA is one of the techniques to be used by the agency s clean laboratory for analysis of the safeguarded nuclear materials and environmental samples.

CONCLUSIONS

Based on the abovementioned facts, the method is considered as one of the most sensitive, accurate and rapid NDA technique. It has wide and versatile applications. The technique is highly recommended to be used for analysis of the safeguarded nuclear materials in Egypt. Uranium analysis for environmental safeguard purposes can also be carried out with high accuracy by DNAA. The method enables uranium in a sample to be measured without respect to its oxidation state, organic and inorganic elements. The sensitivity of the method is 3ng. Using this technique parrall to the other NDA and DA (e.g. mass spectrometry) techniques can fulfill establishing a good state system for accounting for and control of nuclear materials (SSAC) and our commitments toward the international safeguards (IAEA).

ACKNOWLEDGMENT

The author would like to thank Prof. Dr. K.L.Kratz, who had been supervised me during my working in Germany. Prof. Dr. M.S. El-Tahawy, Prof. Dr. A.G.Emara Prof. Dr. A. I. M. Aly and M.A. Abdel Aziz are also acknowledged for their valuable discussion and encouragement.

REFERENCES

(1) Donald R.Rogers " Handbook of Nuclear Safeguards Measurement Methods" ,NUREG/CR — 2078- NRC - USA(1983).
(2) D.Reilly ,Norbert Ensshin, Hasting Smith and Sarah Kreiner" Passive NDA of Nuclear Materials "NUREG / CR-5550-NRC-USA (1991).
(3) S.Amiel ," Nondestructive Activation Analysis " Elsevier Scientific Publishing Company USA (1981).
(4) G. Robert Keepin, Nuclear Technology , Vol.13 , April (1972).
(5) V.M. Gorbachev , V.5. Zamyatnin and A.A. Lbov "Nuclear Reactions in Heavy Elements A data handbook , Pergammon Press -USA (1980).
(6) Cyriel Wagemans "The Nuclear Fission Process ", CRC Press — QC790 —N83 (1991).
(7) Paul Kruger" Principles of Activation Analysis ", Dept. of Civil Engineering, Wiley — Inter Science - USA(1971)
(8) W.E. Burcham and M. Jobes "Nuclear and Particle Physics" , Longman Group Limited, John Wiley & Sons, Inc. , UK(l995).
(9) Raymond A. Serway "Physics for Scientists and Engineers with Modern Physics" , 4'~' ed., Saunders College Publishing , USA(1996).
(10) E. Stephen Binney, I. Robert Scherpelz , Nuclear Instruments and Methods, 154 (1978).
(11) N.Boher and J.A.Wheeler, Physics Review 65, 426 (1939).
(12) G.R. Keepin " Physics of Reactor Kinetics "Addison Wesley, Publishing Co., Reading Massachusetts ,USA(1956).
(13) LTomhinson ,Nuclear Technology, Vol.13 — April (1972).
(14) S.Amiel and H. Feldstein, Physics Letter JiB , 59 (1970).
(15) A.N. Gudkov, S.V. Krivasheyev ,A.B.Koldobski ,ViV.Kovalenko , E.Yu.Bobkov and V.M.Givun , Radiochimica Ada 57 , 69-75 (1992).
(16) J.C.Wang, P.Dendooven , M.Hannawald , A.Honkamen , M.Huhta, A.Jokinen , K.L.Kratz, G.Lhersonneau, M.Ginonen , H.penttila, K.Perajarvi , B.Pfeiffer and J.Aysto , Physics Letters, B454 (1999).
(17) J.Csikcai, "Handbook of Fast Neutron Generators", Volt, CRC, press Inc., Florida (1987).
(18) W.S.C. Williams "Nuclear and Particle Physics", Oxford Science Publications,USA (1991).
(19) R.L.York, E.G. Rooney , D.A. Close and H.E. Williams "Active Neutron Interrogation Package Monitor" The 6"' International Conference on Facility Gperations — Safeguards Interface —20 —24 Sept. Wyoming -USA (1999).
(20) G. Huber, S. Lenz and K.L. Kratz " Determination of the Uranium and Thorium Contents in Aqueous Samples with Neutron Activation Analysis " The 3"' International Conference on Nuclear and Radiochemistry , Vienna 7-11 Sept. IAEA — ASER (1992).
(21) Sayed A.M. El-Mongy " Determination of Baseline Natural Radioactivity Levels in the Suez —Canal Water Stream " , Ph.D Thesis in Radiochemistry An-Sham Uni. ,Cairo, Egypt. (1995).
(22) W.Rosenstock, T. Koble, P.Hilger" Measurement of the Time Structure of Delayed Neutrons after Induced Fission in Nuclear Material", 21" Annual Meeting, Symposium on Safeguards and Nuclear Material Management (ESARDA), 4 -6 May, Spain(t999)
(23) R.H. Augustson. C.N. Emery and C.R.Weisbin ,Nuclear Technology , Vol.14 (1972).
(24) D. Stegemann, Nuclear Technology, Vol.13, April (1972).

(25) Qine Schengzhang, P.M. Riard, E.L. Adams and R.H.Augustsan, "Determination of U and Pu in Simulant Mixture Samples by Cf-252 Shuffler", ESARDA Bulletin No. 26, March (1996).
(26) P.Cloth ,N.Kirch and F.J.Krings "Non-destrnclive Measurement of ^{235}U and ^{233}U Content in HTR Fuel Elements by DN Analysis" Safeguarding Nuclear Materials Proceeding of A Symposium — SGNM — IAEA — Vienna Vol.2 , 20-24 Gct.(1975).
(27) V.P. Novichkov, S.V.Krivasheev and Al. Murashov Soviet Radiochemistry — Russian original Vol.34 ,No.2 ,March — April (1992).
(28) IAEA," Model Protocol Additional in the Agreements between States and the IAEA for the Application of Safeguards", INFCIRC /540, Vienna, Austria (1997).

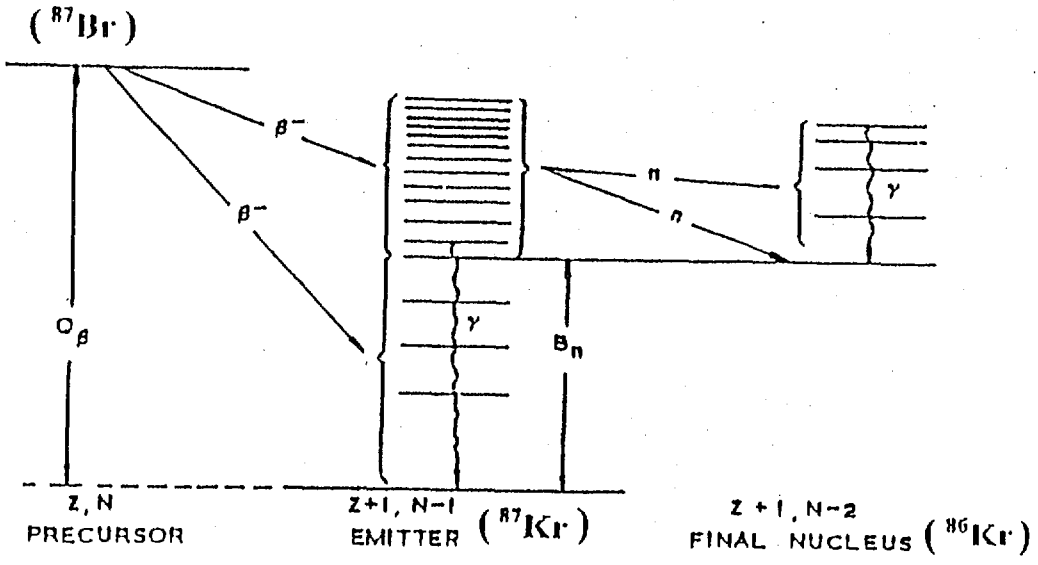

Fig.1 : Schematic Representation of Delayed Neutron Emission
Qβ is the beta decay energy of the precursor
Bn is the neutron binding energy

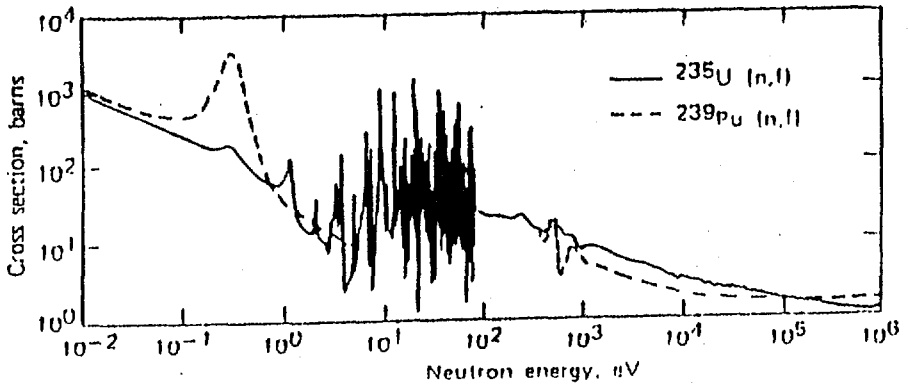

Fig.2 : The fission cross-section of ^{235}U and ^{239}Pu (ref.1)

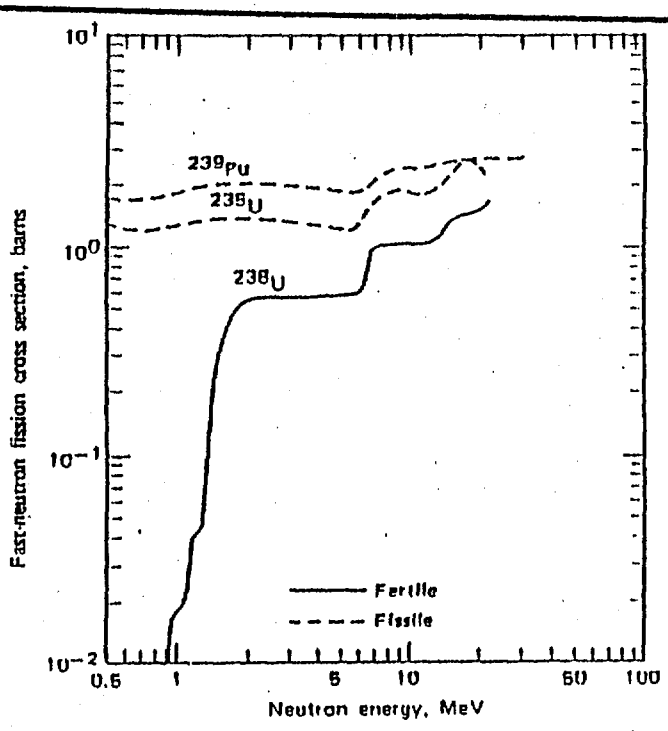

Fig.3 : The fission cross-section of ^{235}U, ^{239}Pu and ^{238}U (ref.1)

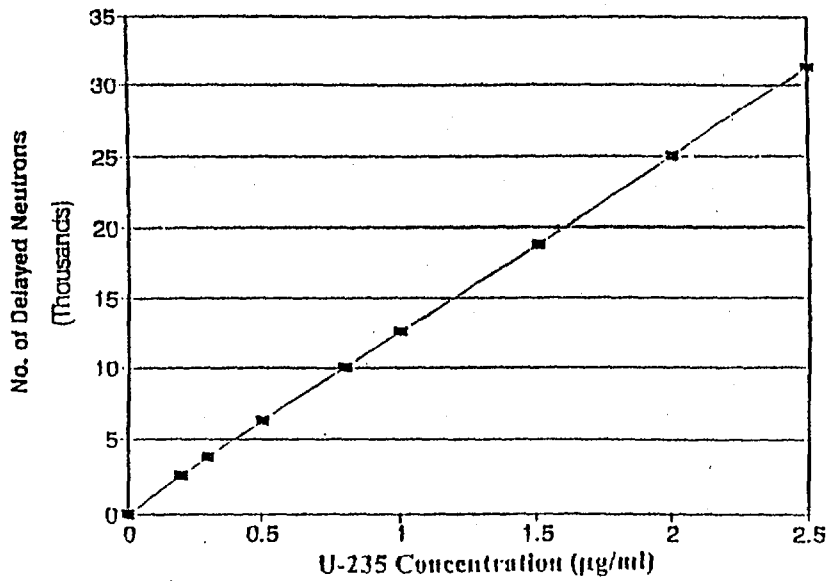

Fig.4 : Calibration curve for ^{235}U standards

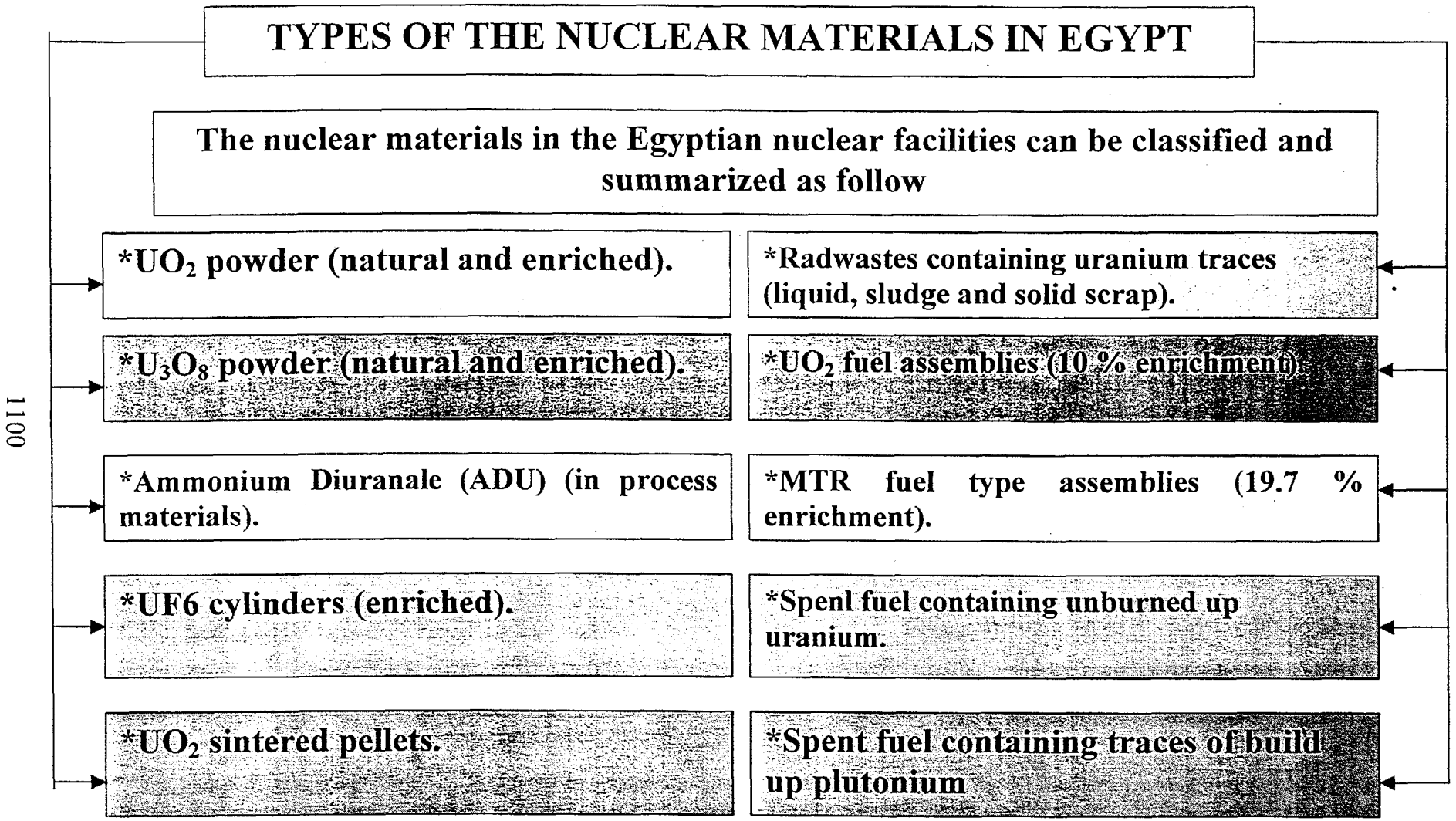

Fig. 5 : Types of the nuclear materials in Egypt

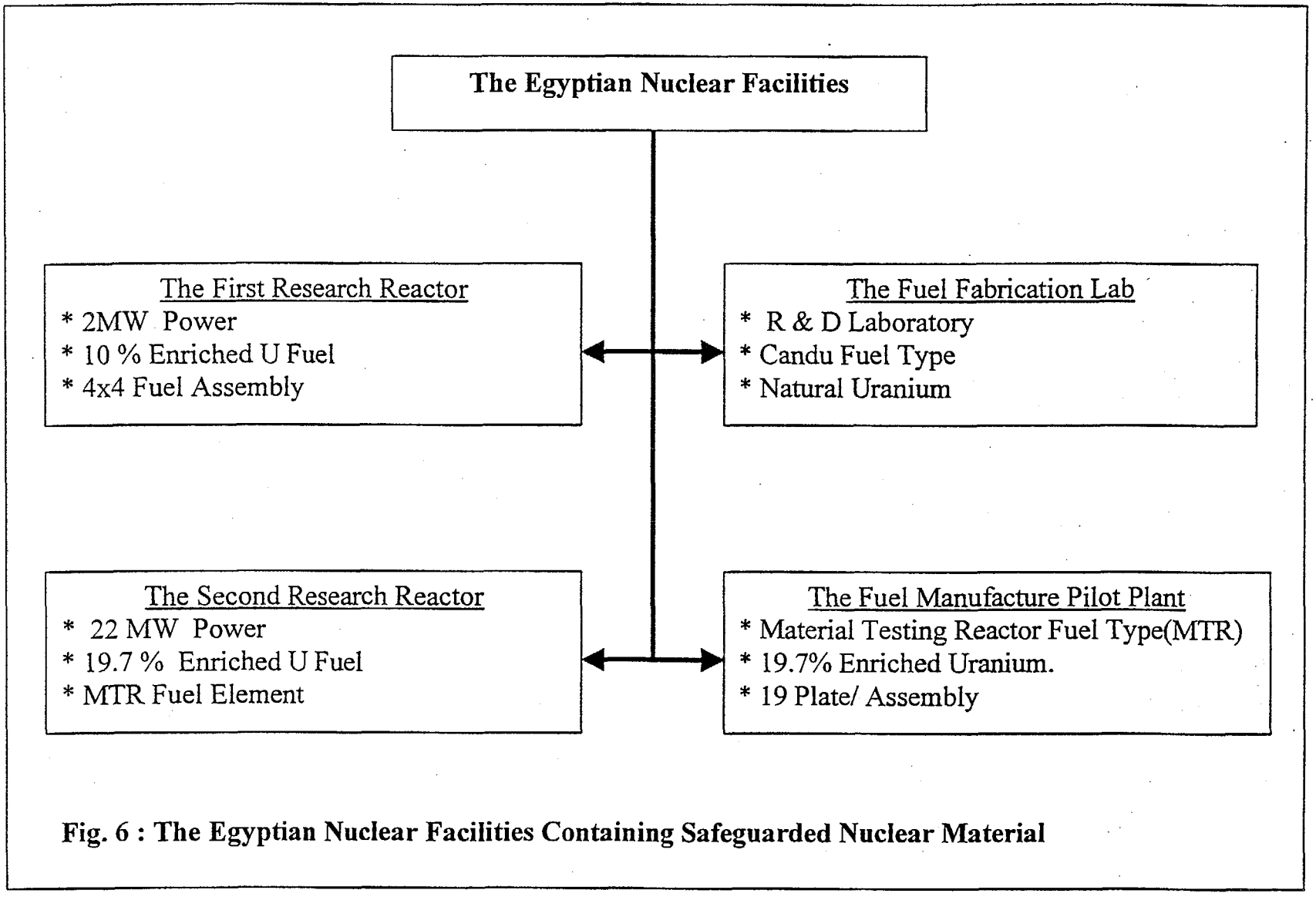

Fig. 6 : The Egyptian Nuclear Facilities Containing Safeguarded Nuclear Material