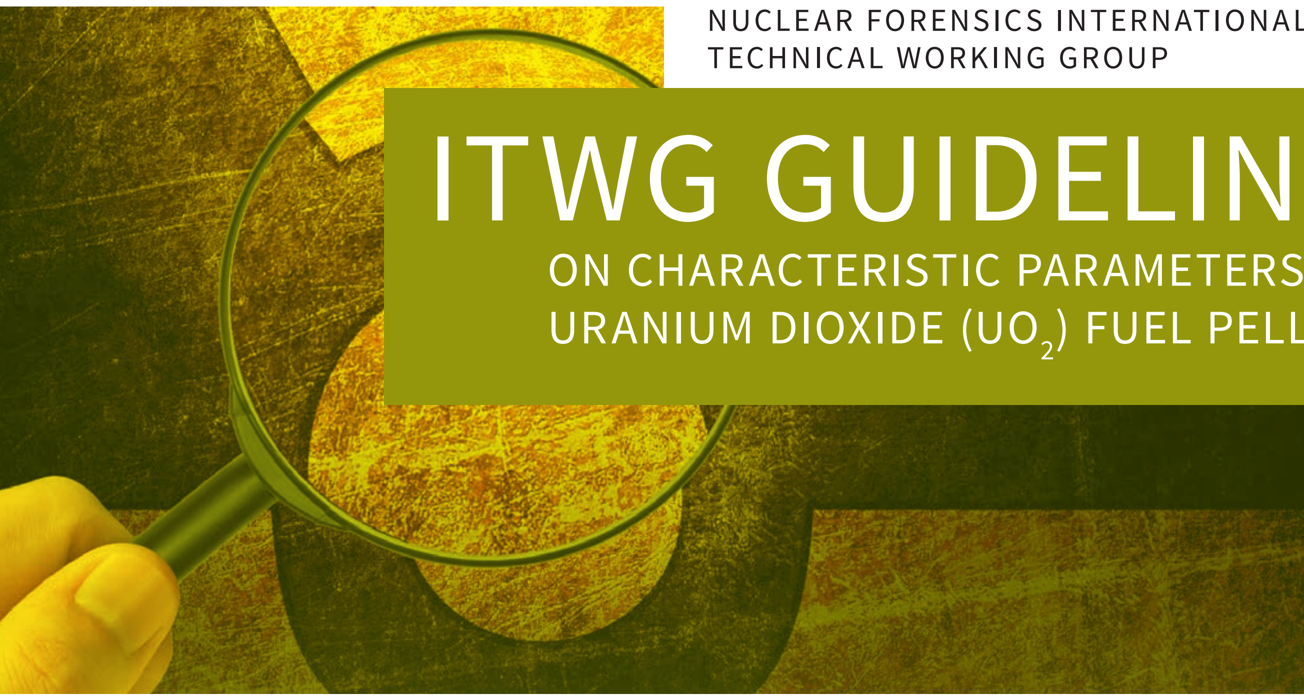
NUCLEAR FORENSICS INTERNATIONAL TECHNICAL WORKING GROUP



ITWG GUIDELINE

ON CHARACTERISTIC PARAMETERS OF URANIUM DIOXIDE (UO_2) FUEL PELLETS

EXECUTIVE SUMMARY

Low enriched uranium dioxide (UO_2) in form of ceramic pellets is typically used in commercial nuclear power reactors as fuel. In order to find out the intended use of UO_2 pellets, and subsequently to narrow down the possible production facility, UO_2 pellets contain a few characteristic parameters (i.e. signatures) that can be helpful in the origin determination. Such signatures include e.g. dimensions, ^{235}U enrichment and impurities. The following guideline will shortly describe each of the signatures and discuss about the information they may provide for nuclear forensic investigations.

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1. INTRODUCTION

Fuel pellets were found out of regulatory control frequently in the 1990s when the phenomenon of illicit trafficking of nuclear materials started. Therefore, a lot of effort was put in studies on their characteristic parameters with a view to trace back the origin [1]. Fuel for commercial power reactors is produced by compacting uranium dioxide (UO_2) powder to cylindrical pellets (about 1 cm × 1 cm) and sintering them at high temperatures to produce ceramic nuclear fuel pellets with a high density and well defined physical properties and chemical composition. A grinding process is used to achieve a uniform cylindrical geometry with narrow tolerances. Such fuel pellets are then stacked and filled into the metallic tubes (cladding made typically of zircalloy) composing fuel rods. The finished fuel rods are grouped into fuel assemblies that are used to build up the core of a power reactor (Fig. 1). UO_2 pellets are (or have been) produced at least in Argentina, Brazil, Canada, China, France, Germany, India, Japan, Kazakhstan, Korea, Pakistan, Romania, Russia, Spain, Sweden, UK and USA.

2. CHARACTERISTIC PARAMETERS OF UO_2 PELLETS

There are number of characteristic parameters (i.e. signatures) in UO_2 fresh fuel pellets that can give information about their intended use (i.e. reactor type) and, consequently, about their production place. The most prominent “signatures” are:

- Dimensions (e.g. diameter, length, central hole)
- Markings
- Microstructure
- ^{235}U enrichment
- U isotopic composition
- Additives and impurities
- Age

In the following each of the “signatures” and their importance are described shortly.

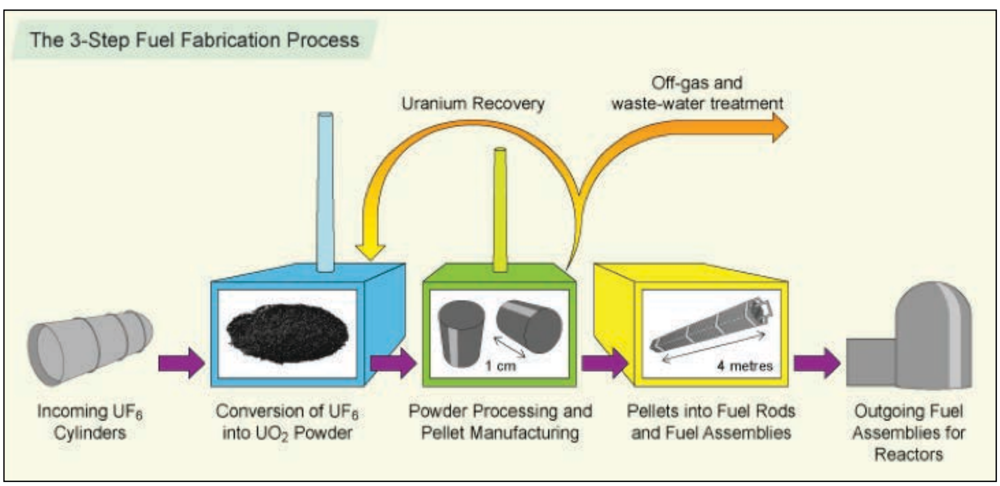

Fig. 1. Fuel fabrication process. (Source: World Nuclear Association)

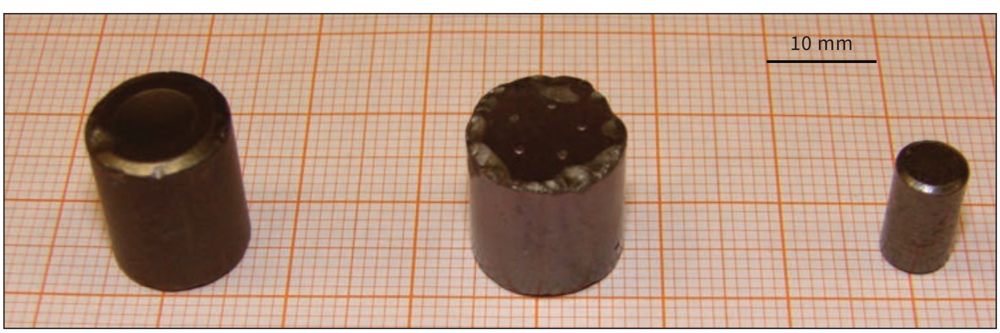

Fig. 2. Various UO_2 fuel pellets. (Source: ITU)

2.1. DIMENSIONS

Typically every reactor type (e.g. HWR, PWR, FBR) requires fuel pellets of tailored dimensions (Fig. 2; Table 1). Pellets are typically cylindrical in shape and may contain well defined geometrical features such as "dishing" and "chamfer" or "central hole" (Fig. 3).
It should be noted, though, that dimensions (encountered in the specifications) are associated with narrow uncertainties, less so for the length.

2.2. MARKINGS

In some cases pellets may bear markings, e.g. dots or numbers, at the ends of the pellets (Figs. 2 and 4). For instance, the numbers on the pellets shown below were intended to represent the nominal ^{235}U enrichment of the pellet (Fig. 4a) and the press number used to press the pellet (Fig. 4b). Markings are comparative signatures and can best be interpreted for forensic purpose with the support of a national nuclear forensic library.

Table 1. Typical dimensions (in mm) of uranium fuel pellets in some common reactor types [1,2]

Reactor type	Model (Name)	Diameter	Length	Central hole
PHWR	Candu 6 (Cernavoda in Romania)	12.16	16	-
PHWR	Candu (Pickering in Canada)	14.3	19.8	-
PHWR	(Rajasthan in India)	12.2/14.34	13.4/17.2	-
PWR	VVER-440 (Loviisa in Finland)	7.6	10	1.2
PWR	VVER-1000 (Kalinin in Russia)	7.55	11-12	2.2-2.4
PWR	(Philippsburg 2 in Germany)	9.11	11	-
PWR	CP2 (Gravelines in France)	8.19	13.3	-
BWR	BWR 5 (Fukushima Daini in Japan)	10.4	10.3	-
BWR	BWR 2 (Nine Mile Point 1 in USA)	9.55	9.6	-
LWGR	RBMK-1000 (Chernobyl in Ukraine)	11.5	15	-
LWGR	RBMK-1500 (Ignalina in Lithuania)	11.5	12-15	2.0
AGR	(Heysham in UK)	14.5	15	6.4

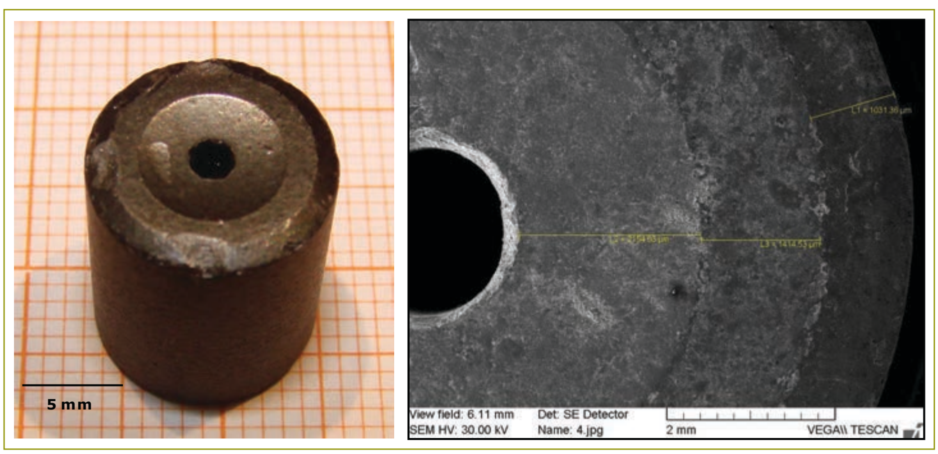

Fig. 3. a) Fuel pellet with a central hole, dishing and chamfer; b) Measurement of the dimensions of the same fuel pellet by Scanning Electron Microscopy (SEM). (Source: ITU)

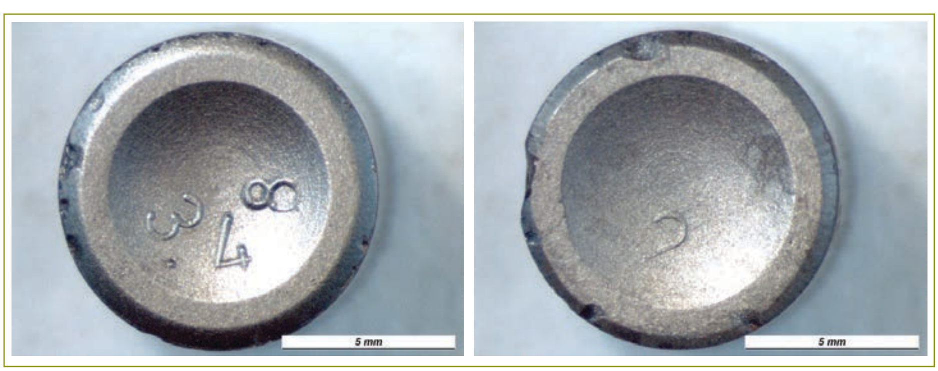

Fig. 4. Markings on pellets. (Source: ITU)

2.3. MICROSTRUCTURE

Microstructure of pellets (e.g. grain size, porosity and surface roughness) can be indicative of the process used for precipitation, solidification, sintering and grinding. For instance, sintering temperature and use of additives can be used to achieve different grain sizes (Fig. 5). Surface roughness, on the other hand, indicates the grinding process used, e.g. the so-called “wet grinding” produces slightly smoother surface compared to the “dry grinding” process [1].

2.4. ^{235}U ENRICHMENT

Depending on the type of reactor and the fuel configuration, the requirement for ^{235}U enrichment may vary. In the first approach we can identify three groups of enrichment level (which may overlap):

- Heavy water moderated reactors such as CANDU use natural uranium as fuel.
- Graphite moderated reactors such as RBMK-1000 and RBMK-1500 use fuel enriched between 2.0% and 2.6%, depending on the reactor model and additives of burnable neutron poison [1].
- Light water moderated reactors such as VVER-440 and VVER-1000 have used typically enrichments of 3.6% and 4.4%, respectively; however the newer models utilise somewhat higher enrichments. Other type light-water reactors (PWR and BWR) have ^{235}U enrichment up to 5% [2].

Within pellet inhomogeneity of the ^{235}U enrichment (e.g. grain to grain variation) may be indicative of the production process (powder blending, co-milling).

2.5. U ISOTOPIC COMPOSITION

Besides the ^{235}U enrichment of the fuel which is a key parameter, also the uranium minor isotopes can constitute to a useful nuclear forensic signature. Firstly, the ^{236}U and ^{232}U in the fuel show that the uranium has been exposed to neutrons, thus being previously in a reactor and afterwards reprocessed (and possibly enriched again). Secondly, as not all fuel fabrication facilities produce fuel from reprocessed uranium, it can limit the number of possible manufacturers.

2.6. ADDITIVES AND IMPURITIES

Several commercial fuels use additives, such as gadolinium, erbium or boron, as burnable poison to control the reactivity of the fuel. The amount of the burnable poisons in fuel is considerably higher than if they were found as impurities.

Impurities are detected in fuel in trace levels and they originate from various sources, including U ore (e.g. REE pattern in natural U fuels), process piping, reagents, etc. Thus, they may be indicative about the process used to produce the fuel. For example, some fuel manufacturers use aluminium stearate as a mold release agent. Use of aluminium stearate can elevate the residual Al in the fired fuel pellet by 30-60 ppmw.

Contaminants on the surface of the pellets may provide information about pellet processing (sintering, grinding, etc.). For example, trace amounts of Mo can be left on pellets after sintering, from the Mo trays used to hold the pellets during the sintering process.

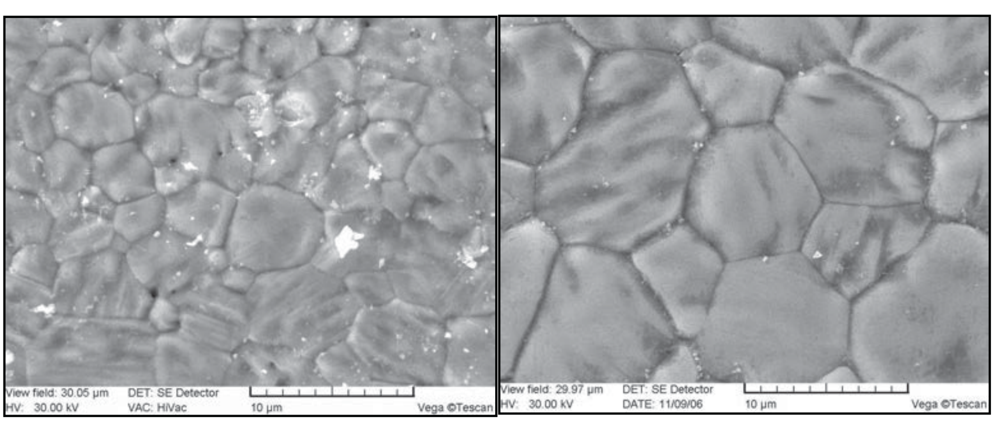

Fig. 5. Surface morphology of two different types of fuel pellets by SEM. (Source: ITU)

2.7. AGE

The radioactive decay of uranium isotopes allows establishing the “age” of the material. As the method is based on certain assumptions (see ITWG guideline INFL-ADPD on age dating), the “age” is referred to as the model age. This is understood to describe the production date of the material. It should also be noted that the final separation of the parent/daughter pair for a UO_2 fuel can take place in various stages depending on the process used before conversion the material to UO_2 powder. Therefore, it can be e.g. during the enrichment, the hydrolysis or the chemical conversion. The model age, even if determined with low uncertainty, should be used thoughtfully when put in the context of a technological production process. One must remember that several sources of material can be combined to make up the final fuel pellet (co-milling, powder blending). This will lead to an “average” age of the material for bulk age dating. In addition, many manufacturers add some U_3O_8 to the powder mix before pressing into pellets, so as to increase the O/U ratio prior to sintering (“U_3O_8 Add-Back”). The U_3O_8 is made from scrap UO_2 and will usually have an age slightly older than the UO_2 used in the pellets. This can bias the model age several months older than the age since conversion of UF_6 to UO_2.

3. DATA INTERPRETATION

Some of the above mentioned signatures are self-explanatory, i.e. the results do not need any comparative data to give an implication. Such a signature is, for instance, the age, because it can be calculated directly from the measured parent/daughter ratio using the common decay equation. However, the elemental impurities are a good example for a comparative signature, where one needs comparison data to elucidate the results. For example, the quality control data from fuel manufacturers could be utilised for this purpose.

4. REFERENCES

1. L. Pajo, Ph.D. thesis 2001, University of Helsinki, http://urn.fi/URN:ISBN:952-10-0122-4.
2. Nuclear Engineering International, World Nuclear Industry Handbook 2012.

DOCUMENT REVISION HISTORY

Document INFL-UOFP			
Version No.	Version Date	Description of Changes	Changes made by
1	2016-09-01	Initial Draft	M. Wallenius (author)