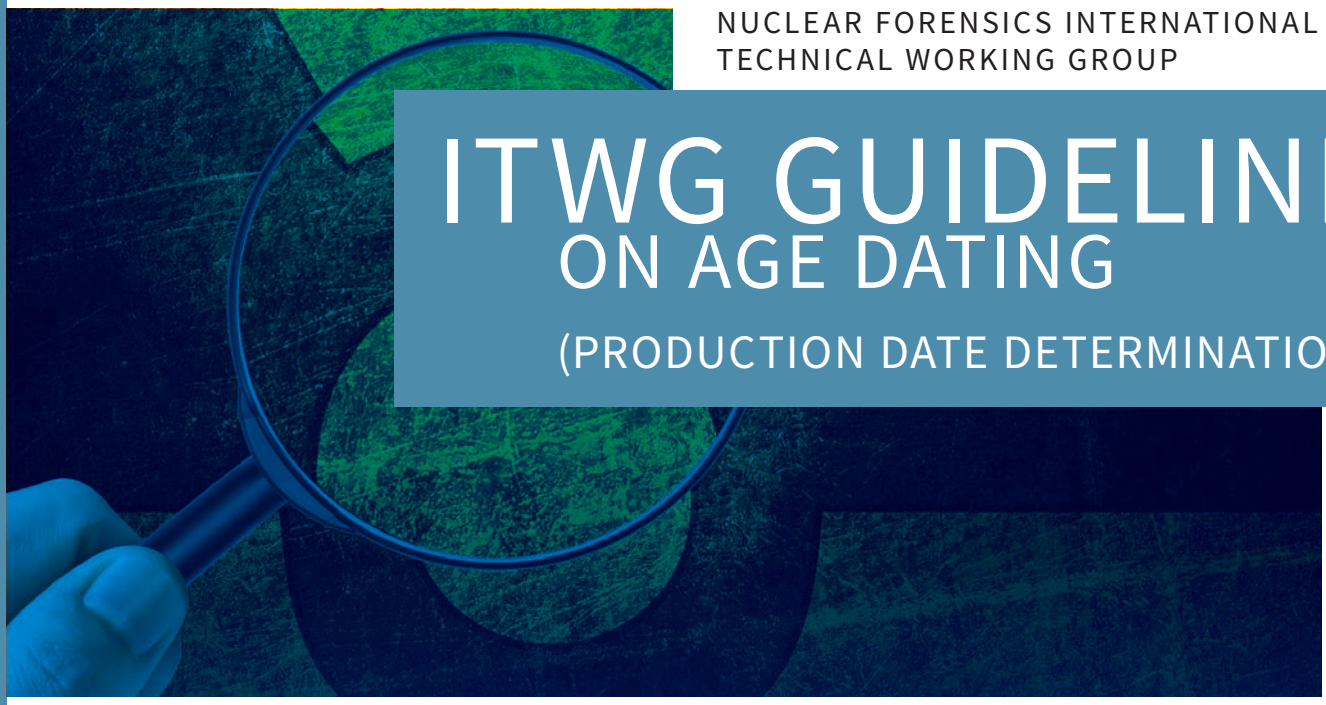
NUCLEAR FORENSICS INTERNATIONAL
TECHNICAL WORKING GROUP



ITWG GUIDELINE
ON AGE DATING
(PRODUCTION DATE DETERMINATION)

EXECUTIVE SUMMARY

Age dating (production date measurement or radiochronometry) of a radioactive/nuclear material in nuclear forensics refers to the time when the material was last chemically or physically purified or modified. The principle is based on the measurement of the radioactive progenies (decay products) relative to the parent nuclides, which serves as a chronometer for age dating. After the measurement of a progeny-to-parent ratio, the time of their last separation can be calculated based on the radioactive decay equations. Important theoretical conditions have to be fulfilled in order to obtain accurate and meaningful age results, such as completeness of separation or closed system behavior after production. Ideally, the obtained age results will give predictively the time of a certain chemical or physical process related to the considered progeny-to-parent ratio.

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1. INTRODUCTION

Nuclear materials, just like other radioactive materials, contain radioactive nuclides, which decay over time. This unique property can be exploited to measure the time elapsed since the last chemical or physical processing of the material. This time is generally referred to as the production date or (model) age of the nuclear material in nuclear forensics. The age determination technique (radiochronometry) is based on the fact that progenies (decay products or daughters) of the nuclear materials are continuously growing-in within the nuclear material after the last chemical separation of the base material (e.g. uranium or plutonium). The amounts of decay products are proportional to the amount of parent nuclide and the time elapsed since the last chemical separation. In the simplest case, if a radioactive nuclide decays to a stable daughter nuclide, the amount of the produced daughter nuclide as a function time can be calculated based on the Bateman equations as follows [1]:

$$N_B(t) = N_A^0 \times \left(1 - e^{-\lambda t}\right) \qquad \text{(Eq.1)}$$

where $N_B(t)$ is the amount of daughter nuclide as a function of time, N_A^0 is the amount of parent nuclide at $t = 0$ (time of production), λ is the decay constant of the parent nuclide (λ is equal to $\ln(2)/T_{1/2}$, where $T_{1/2}$ is the half-life of the parent nuclide) and t is the elapsed time since $t = 0$. The elapsed time is often referred to as the (model) age of the material. Rearranging Eq. 1. the elapsed time since production (t) can be calculated then as follows:

$$t = \frac{1}{\lambda} \ln\left(1 + \frac{N_B(t)}{N_A(t)}\right) \qquad \text{(Eq.2)}$$

A similar equation can be applied using the activities of the radioactive nuclides instead of the amounts, if radiometric technique is used for the measurement.

For more complex decay schemes, such as successive decays in the case of nuclear materials, the following decay equation can be used to calculate the production date of the material assuming that no daughter nuclide was present at $t = 0$:

$$t = \frac{1}{\lambda_A - \lambda_B} \ln\left(1 + \frac{N_B(t)}{N_A(t)} \cdot \frac{(\lambda_A - \lambda_B)}{\lambda_A}\right) \qquad \text{(Eq.3)}$$

where $N_A(t)$ and $N_B(t)$ are the amount of parent and daughter nuclides as a function of time, respectively, λ_A and λ_B are the decay constants of the parent and daughter nuclide, respectively, and t is the elapsed time

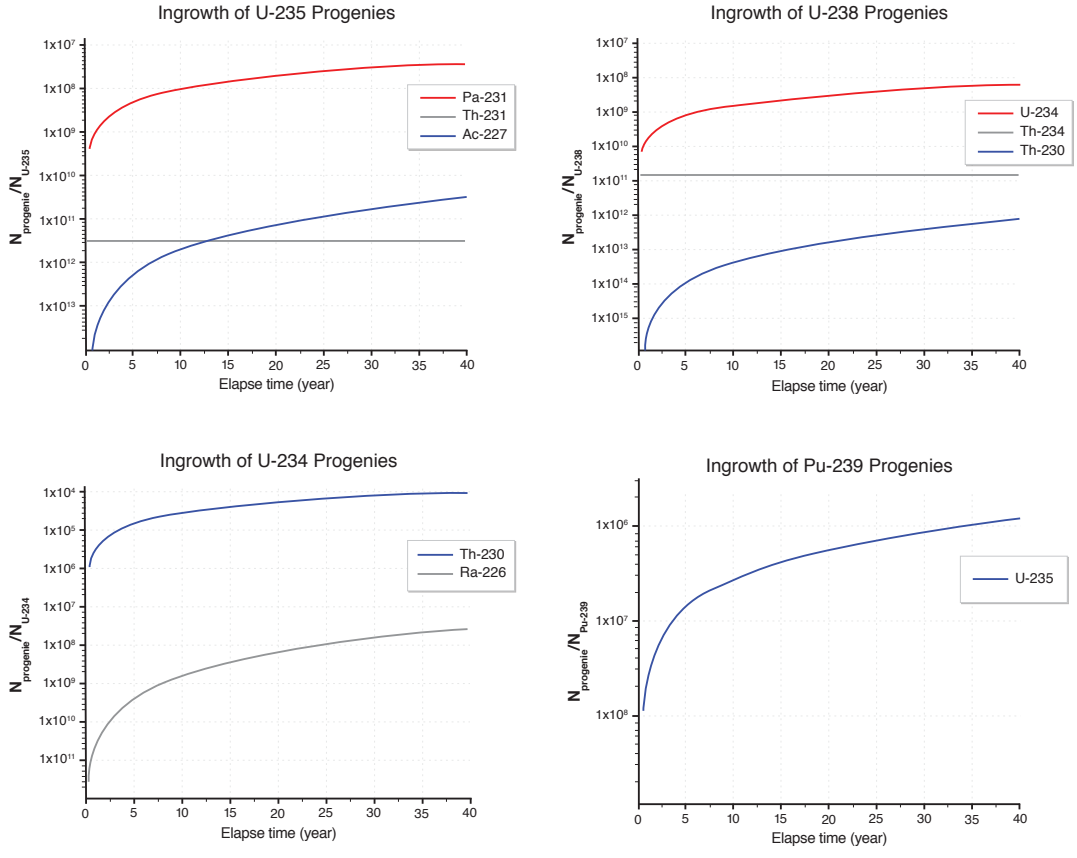

Fig. 1. Variations of the daughter-to-parent amount ratios in nuclear materials as a function of time [2].

since $t = 0$ (age of the material). The decay equations can also be written equivalently using the activities of the respective nuclides if the radiometric technique is used for the measurement:

$$t = \frac{1}{\lambda_A - \lambda_B} \ln\left(1 + \frac{A_B(t)}{A_A(t)} \cdot \frac{\lambda_A - \lambda_B}{\lambda_B}\right) \quad \text{(Eq.4)}$$

Note that in any case the age is the function of the daughter/parent ratio; therefore, the ratio is often referred to as the *chronometer* of the age measurement. The variations of a few daughter-to-parent amount ratios in nuclear materials as a function of time are shown in Fig. 1.

It is important to emphasize that the measured age corresponds to the time when the considered parent and daughter pair was last separated: this implies that the time refers to a certain process step (e.g. uranium enrichment) of the material production for a given chronometer. Different chronometers may give the time of a different production step.

As age dating is based on the radioactive decay equations above, some assumptions or simplifications are applied so that the unknown (the time elapsed since separation) could be mathematically calculated. Important assumptions are:

- Complete removal of the progenies from the parent nuclide at the time of production (t = 0).
- Material has to behave as a closed system after production, i.e. neither parent nor daughter nuclide are added or removed.
- The parent/daughter ratio should not be in secular equilibrium.

The chronometer used for the age calculation usually also defines which analytical techniques can best be used for the analyses. Various mass spectrometric or radiometric techniques and different spike isotopes have already been used successfully for age dating studies, a collection of the methods can be found in the references.

2. USE FOR NUCLEAR FORENSICS

Age dating was found to be highly valuable in nuclear forensic investigations in the recent decades as being a predictive characteristic (i.e., no comparison sample or information is necessary). Besides, age dating is also very useful as a comparative signature to eliminate potential production facilities or to discriminate multiple materials, which was also demonstrated during the 3rd Round Robin exercise and CMX-4 of the ITWG [3, 4].

Age dating can theoretically be used for any radioactive materials, however, technical limitations or restrictions from the measurement capabilities can encumber the application.

3. SAMPLE AND REPORTING REQUIREMENTS

The amount and form of the sample needed for the analysis depends on the applicable chronometer used for age dating, which, in turn, also defines the analytical technique applied for their measurement. Therefore, the amount and form of sample can be highly variable. However, care has to be taken to use a representative sample amount and not to change the parent/daughter ratio by sampling; otherwise an incorrect age will be obtained.

The reported age should be given either as the time of production together with the combined uncertainty or the (model) age of the material together with its combined uncertainty relative to a specified reference date can be reported. It is also advisable to indicate the half-lives used for the calculation. The chronometer used for age dating also has to be given, since the time refers to a specific process step of the material production. Use of multiple chronometers from the same material is advantageous as they will increase the confidence in the obtained result (in case of consistent age values) or it may provide hints on different process steps affecting the various parent/daughter pairs in different ways (in case of inconsistent age values).

4. PRO'S AND CON'S OF THE TECHNIQUE

+ Predictive signature, no need for comparison samples.

+ Very precise production date may be given.

+ Highly useful to eliminate potential origins.

– Measurement often requires expertise and advanced or expensive instrumentations.

– Age dating fails for samples, where the theoretical prerequisites (completeness of separation, material has to behave as closed system after production) are not fulfilled. In such cases multiple chronometers can be helpful. Further examples are available in the references.

– Expert knowledge is required to interpret the result and its relation to the specific production stage.

5. FAQ

- Which half-life values are the best to be used? Values from metrological institutions have higher level of confidence, as they are often reevaluated taking into account several reported values from different laboratories.

- Are there certified reference materials available to validate the results? The CRM 125-A (New Brunswick Laboratory, USA) and the provisional IRMM-1000a and IRMM-1000b (European Commission JRC Institute for Reference Materials and Measurement) uranium materials are certified for the $^{230}Th/^{234}U$ chronometer production date.

6. REFERENCES

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DOCUMENT REVISION HISTORY

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