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DINA, method for measuring natural specific acitivities in ores, minerals and waste streams in the non-nuclear industry

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DINA, METHOD FOR MEASURING NATURAL SPECIFIC ACTIVITIES IN ORES, MINERALS AND WASTE STREAMS IN THE NON-NUCLEAR INDUSTRY

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ABSTRACT

The specific natural radioactivity in ores, minerals and waste streams varies in a great amount. Therefore the Non-Nuclear Industry (NNI) has in principle to perform continuously control measurements in order to detect in time, if the specific activity in the different materials is not exceeding limits set by authorities. However, the throughput of a NNI is mostly hundreds of tons a year. These huge quantities make it almost impossible to measure all ores, minerals and waste streams. When the increase of radioactivity, caused by decay of ^{226}Ra, has to be taken also into account, it becomes really impossible. This is due to the fact that for reaching equilibrium of the ingrowth a waiting time of 25 days is necessary. Most NNI's have no space to store the materials for a maximum of 25 days.

Therefore, a special method for checking quikly the specific natural radioactivity in ores, minerals and waste streams is developed. This method is called "DINA", which stands for "Data Interpretation of Natural Activity". The aim of DINA is to check the specific natural activity against exemption and clearance levels. This means in practice, that a NNI has not to wait 25 days, but only some days before they are sure, that a material will superceede a limit set by authorities.

BASICS OF DINA

The secular equilibrium of the uranium decay-chain is disturbed when a material has been treated. The noble gas radon can be escaped. For a good measurement the equilibrium must be restored, but reaching equilibrium needs waiting for 20-25 days.

The aim of this special for the NNI developed non-destructive assay method (DINA) is to detect and calculate by means of emitted gamma photons the natural specific radioactivity of ores, minerals and waste streams etc. as fast as possible with a high accuracy and for a low price. The calculations are performed in such a way that all uncertainties of all important parameters are taken into account. Assessments can be made about the chance that the specific activity will superceding a limit set by the companies health physics manager or set by authorities.

The advantage of such a method is evident. The NNI is able to make quick assessments if materials entering or leaving the company will cause problems. Further by using a simple method the NNI can use own personnel and specialists will only be needed in special cases.

The basic philosophy of DINA can be summarized in four topics:

- data will always be measured by equipment, which is free from failures and correctly calibrated. In practice this means, that values of measured data are considered as really true and can be trusted. It is evident, that this is only possible with correct qualified procedures
- each uncertainty about the natural specific activity of a sample caused by a lack of knowledge about the equilibrium of ^{226}Ra and daughters will not influence in any way the reported measured/calculated natural specific activity of this sample. A lack of knowledge will only influence the standard deviation of this reported specific activity

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- all gathered knowledge of a sample may and will be combined to calculate the specific activity. This means, that depending on the gathered knowledge different calculation methods will be used (DINA accepts a maximum of three data inputs per sample). All used methods have to be physically correct
- results of different calculation methods may be compared and selection on bases of the results may be applied, so that the NNI can use the most appropriate calculation method for each individual sample. In practice this will mean, that the final selected method will be the method, which estimates the smallest change for superceding limits set by the companies health physics manager or set by authorities.

DESCRIPTIONS OF PARTS OF DINA

DINA consists of five main parts:
- measuring device
- characterization of materials
- calibration
- sample
- calculation methods and data base

and will be discussed in more detail. The relation of these parts are shown schematically in figure 1.

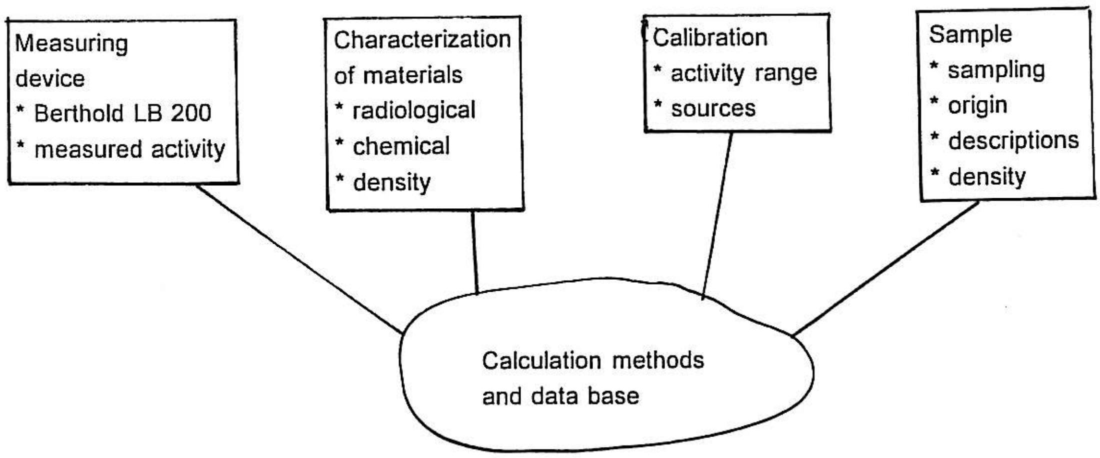

Figure 1 Schematic presentation of parts of DINA

Measuring device
The LB 200 monitor is developed after the Tsjernobyl accident to investigate the specific activity of food and other materials, which could be contaminated. The food monitor LB 200 has a small NaI(Tl) detector and integrating electronics. This equipment is very easy to operate and an acoustic signal is given when the statistical uncertainty becomes below 5%. The sample holder is a 0.5 litre marinelli beaker. Important is, that the sample is homogeneous distributed in the marinelli beaker. The beaker must have the possibility to be sealed air tight. In the case, that the radon exhalation is not negligible, an equilibrium between ^{226}Ra and daughters will be established in 25 days.

Characterization of materials
The characterization of bulk material has to be performed on the following aspects:
- radiological. The radiological characterization is done by defining a nuclide vector including uncertainties for the bulk material

- chemical. For an outsider it looks. if an industry can use in his plant all raw materials of a certain kind. However in practice this is seldom true. This is also one of the main reasons for checking incoming materials. It means also, that by a steady industrial process and by a restricted chemical composition of incoming materials, produced semi-manufactured articles, waste streams etc. have also a limited chemical composition
- density.

The chemical characterization and density data will be used by the calculation of the unique material calibration factor and its standard deviation. Both characterizations influence the self absorbtion of emitted gamma photons and for this absorbtion corrections have to be performed.

The radiological characterization data will be used for calculating the specific radioactivity of the sample.

Calibration
The calibration has to be performed according qualified procedures, so that the first statement of the basic philosophy will be true and can be proven. The specific radioactivity of bulk materials will cover in general a certain range. Therefore it is necessarily to have different calibration standards, which are able to cover this range.

The best chemical and physical characterization of a calibration standard is a standard which is identical to samples to be measured. When a NNI is dealing with more than one ore, semi-manufactured article, waste stream etc. it become not practical to have for each material a separate set of calibration standards. Therefore methods are developed which are able to calculate within 7% unique material calibration efficiency curve. It is evident, that this method has to be verified and validated. KEMA has developed its own simulation method (called DENSITY) and validated this method with 42 (30 theoretical and 12 experimental) different validation runs. Therefore DENSITY may be used in for our ISO 9001 certified analysis.

Sample
Sampling must be performed according to international or national standards. If these standards do not exist, than the industry has to write its own procedures according the current state of the art. An sample has to be classified for DINA at:
- unique sample identification (needed for traceability of the results and eventually for ISO 9000 certification)
- date and time of sampling
- description of sample and material stream. This data define the calibration factor
- density. The density is used for correction of the specific activity due to differences of this parameter between the calibration source and the sample
- date and time of measuring. The time difference between sampling and measuring is used for making an assessment of the ingrowth of the ^{226}Ra daughters.

The radon emanation is at this moment not taken into account, because the samples are supposed to be sealed air tight and thus ^{222}Rn can not escape.

Calculation methods
As stated in the basics of DINA all gathered data of a sample may and will be used by the calculation of the specific activity and its uncertainty. This is performed by storing all data in a data base. In this data base is space available for the data storage of three measurements.

The number of possible different calculation methods depends of coarse on the number of individual measurements. At this moment is implemented in the software code (runs on a standard PC) for 1, 2 and 3 measurements respectively 1, 3 and 4 different calculation methods. The methods differ in the calculation of the uncertainty of the measured specific activity.

The most important parameter for making the assessment of this uncertainty is the "equilibrium correction factor". This factor describes the difference between a total equilibrium between ^{226}Ra daughters and the possible existing equilibrium level based on data filled in in the data base. In table 1 values of this correction factor C is given as a function of the equilibrium level. This factor will have in practice a value which lays between 1.00 and about 22.5.

Table 1 The equilibrium level of the ^{226}Ra chain after a certain time interval. The decay time of ^{222}Rn is expressed as λ (2.1E-06 s)

Equilibrium period [days]	Equilibrium fraction $I_{fract} = (1-e^{-\lambda^* t})$	Correction factor $C = I_{fract}/I_{fract\ 100\%}$
0.25	0.04	22.55
0.5	0.09	11.53
1	0.17	6.03
4	0.52	1.94
8	0.77	1.31
12	0.89	1.13
16	0.95	1.06
20	0.97	1.03
25	0.99	1.01

When the correction factor is larger than 1, it means that there is no steady equilibrium level between ^{226}Ra and daughters and that there can be a possibility that the specific activity of the sample has to be increased with this correction factor. This possible increase must be taken into account by reporting of the measured activity value. This is done by an assessment of the increase of the positive standard deviation. In the basic philosophy paragraph stands that "a lack of knowledge will only influence the standard deviation of the reported specific activity and not the measured data itself"

A large increase of the standard deviation makes that the chance on superceding a limit set by the company or set by authorities become very realistic.

VALIDATION

The validation is done experimentally. A sample was heated up to 1400 °C to drive away the present radon. The sample was measured almost every day during a period of 25 days. The activity measured by the Berthold LB 200 increased in this period from 176 Bq/kg up to 250 Bq/kg. This increase shows clearly that at the beginning there was no steady equilibrium between ^{226}Ra and daughters (see table 2).

Table 2 Validation data. The sample was heated up to 1400°C and then cooled down without a force cooling, before the first measurement was performed.

Day	Activity [Bq/kg]
1	176
2	187
3	202
4	210
5	211
8	233
9	236
10	237

Day	Activity [Bq/kg]
11	243
12	240
15	246
16	249
17	250
18	246
23	245

The measured data and all characterization data were filled in the data base with all possible combinations (data of respectively one, two and three measurements). In figures 2 and 3 are given two examples of the validation. Figure 2 is based on the input of one measurement and figure 3 on two measurements - combination of the data of the first day with the data of every other day-.
In the figures are shown:

- the expected end value of the activity
- the measured activity
- an arced area representing the range in which the activity can be
- the change on superceding a limit (the limit was set at 400 Bq/kg for these validations).

By comparing figure 2 and 3 one can see the advantage of combining all knowledge of a sample. In figure 2 (one measurement) the chance on superceding the limit of 400 Bq/kg is at the 4th day about 20%, while in figure 3 (combination of two measurements) this chance is decreased till about 1% at a 99.5% confidence level.

CONCLUSION

We have developed a method called DINA "Data Interpretation of Natural Activity" which is a useful tool for the NNI. The largest benefit of this tool is, that the chance of not superceding an activity limit can be assessed with an uncertainty of less than 1% within 4 days instead of up to 20 days. Other advantages are that the analysis can be performed by own analysts and are relative cheap.

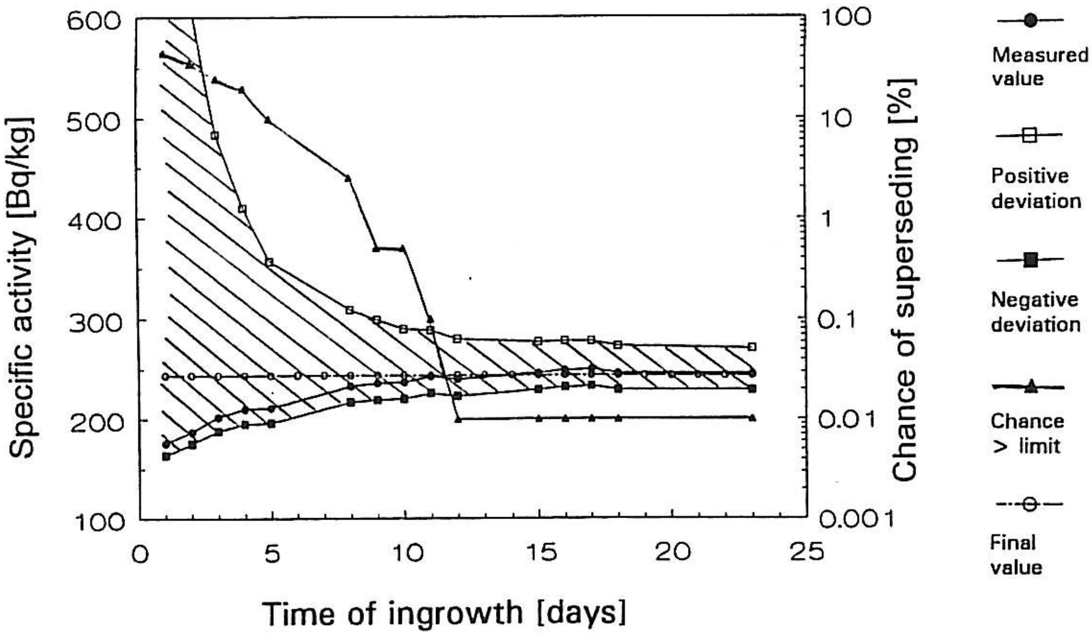

Figure 2 Data presentation of the validation results of DINA with one measurement

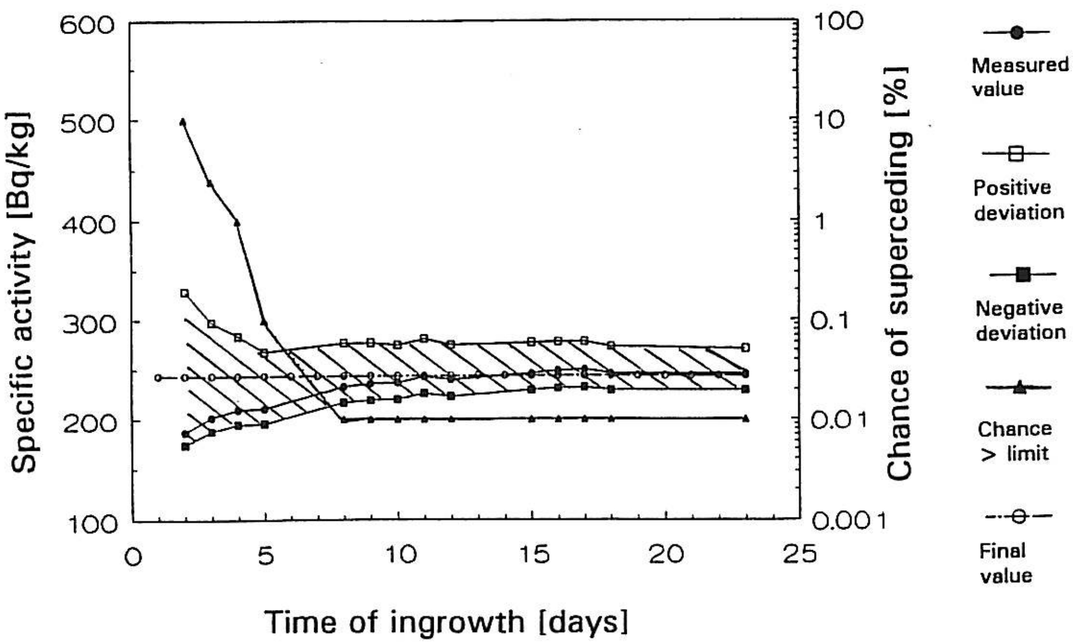

Figure 3 Data presentation of the validation of DINA with a combination of two measurements