- Radioactivity in Education -

Proof of Radon Exhalation from Building Materials and Natural Stones
A Completely Hazard-free, Instructive and Simple Experiment on Radioactivity

Bernd Laquai, Sept. 4, 2015

Description of the experiment

Goal of the experiment is the measurement of the time dependent radioactivity of a sample and the determination of the half-life from the time dependent decay. The sample may be a natural stone or a building material that contains small amounts of natural uranium. The sample may also be a phosphorus fertilizer containing uranium in small quantities as it is used frequently in farming (superphosphate). From its half-life of 3.82 days the noble gas radon as a decay product of the natural uranium is determined unambiguously.

The measurement is executed in two steps. First, activated charcoal is added to the sample in terms of a charcoal tablet. The activated charcoal binds the radon escaping from the sample in the course of the uranium decay. Second, the charcoal tablet is put in a fixture and sealed gas-tight with an adhesive foil. Then its activity is measured through the foil with a Geiger counter. The Geiger counter may be a commercial instrument or a self-made measurement instrument for radioactivity (e.g. the "Tino"-kit as decribed on the website opengeiger.de).

The radiation emitted from the natural sample (about 50-250ml sample volume) can hardly be detected by direct measurement. The concentration of radon when absorbed by activated charcoal in presence of only a small remaining air volume is increased and therefore it becomes easily measurable. As a stone sample porphyry from a quarry near Ellweiler in South Germany (Saarland) is used for demonstration. Porphyry is a frequently used building material as a natural stone.

From this experiment the decay of uranium along the uranium-radium decay chain is demonstrated. The radioactive equilibrium of the decay products plays an important role, in particular the equilibrium between radium and radon or the equilibrium between the radon decay products and their mother nuclide radon. The experiment is easily transferred to other building materials or uranium / radium containing materials from every day's life such as old watches with radium pointers or uranium minerals. The directly visible exponential characteristic of the measured activity versus time impressively depicts the radioactive decay law. The variation of the measured values also highlights the statistical properties of the decay process very clearly.

The experiment however may also be used to pinpoint to the hazards of naturally occurring radon in radon-prone areas. Radon not only exhales from building materials but also from soil in specific geographic areas and therefore may enter into dwellings and living rooms when no respective prevention measures are taken (key word: "radon safe construction") . Radon imposes a risk to health when it becomes a permanent part of the breathing air in significant activity concetrations (e.g. more than 300Bq/m^3). The deposition of the radon decay products on the sensitive lung tissue may lead to lung cancer with a significantly increased risk.

Preparation of the experiment

The crushed building material is put into a glass with a tight twist-off cap. The glass is filled completely to minimize the remaining air volume. One or more activated charcoal tablets as available from pharmacies are added to the sample. Then the glass is tightly closed. The pile-up of the radon activity concentration follows an exponential law and reaches its saturation value after about 3 half-lives of the radon (ca. 12 days). During this time the charcoal tablets need to remain in the tightly closed glass. Right after closing the glass, the Geiger counter should already be prepared for measurement. The data recording feature should be set for storing a count rate measurement value every hour. With this initial measurements the background rate without the sample is obtained afterwards.

Fig. 1: Jam jar with a porphyry stone sample and an activated charcoal tablet

As a fixture for the charcoal tablet a 100mm acrylic glass (Plexiglass) is used. A blind hole is drilled into the acrylic glass such that the tablet can be inserted flush-fitting with a minimum of remaining air volume. After “loading” the charcoal tablet with radon over 12 days it is taken out of the glass and it is inserted into the fixture. An radon tight adhesive foil (or tape) is used to quickly seal the tablet in the fixture without bubbles from the center to the outer edge of the fixture. The adhesive foil should overlap the tablet about 15mm in each direction for the sealing to remain gas tight. Note that not each foil is radon tight.

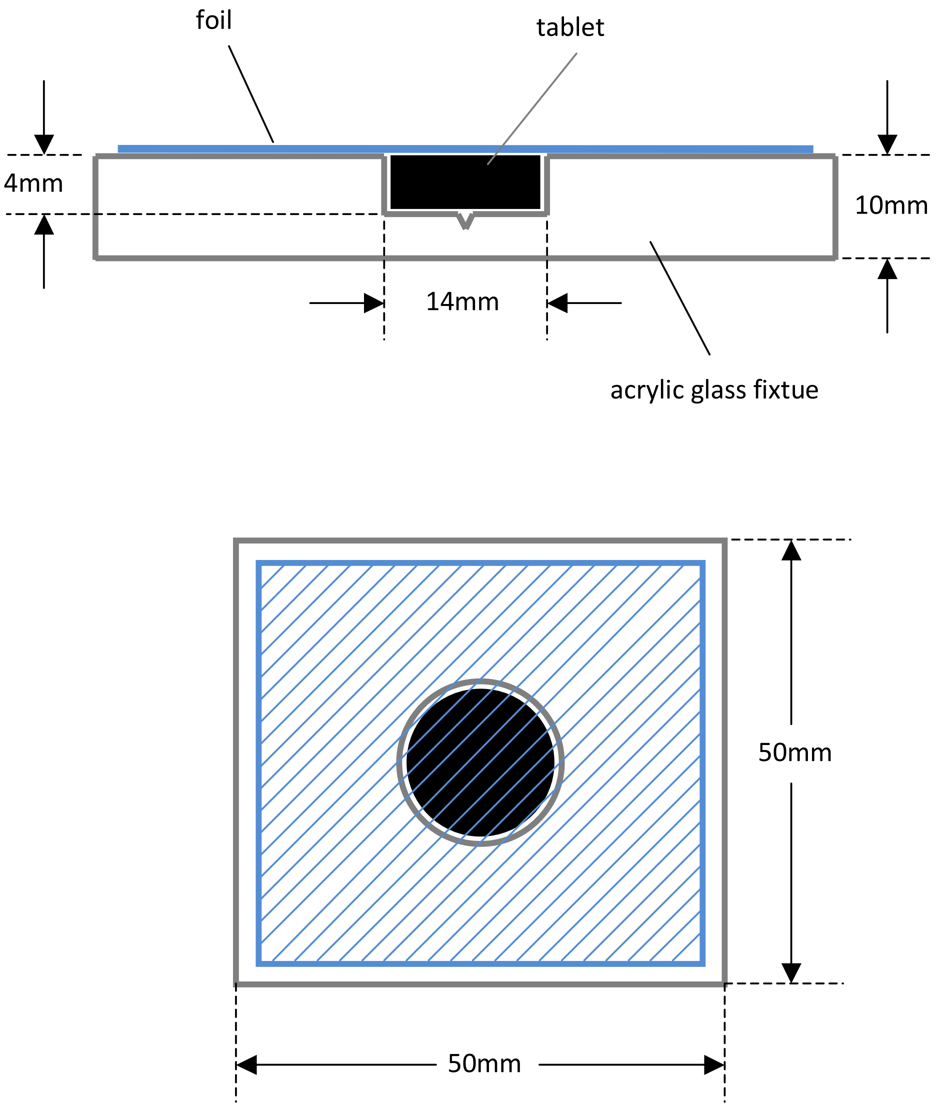

Fig. 2a,b: Example construction of the tablet fixture, cross section and top view. Dimensions match the size of the German brand “Kohle-Compretten, Merck”

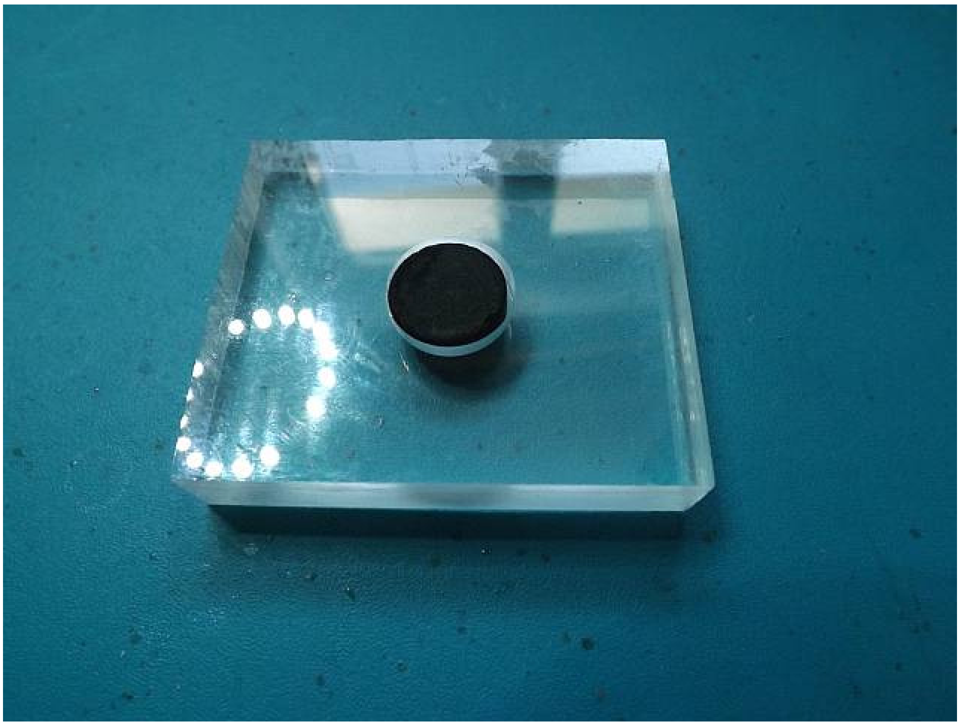

Fig. 3a: Charcoal tablet inserted into the fixture

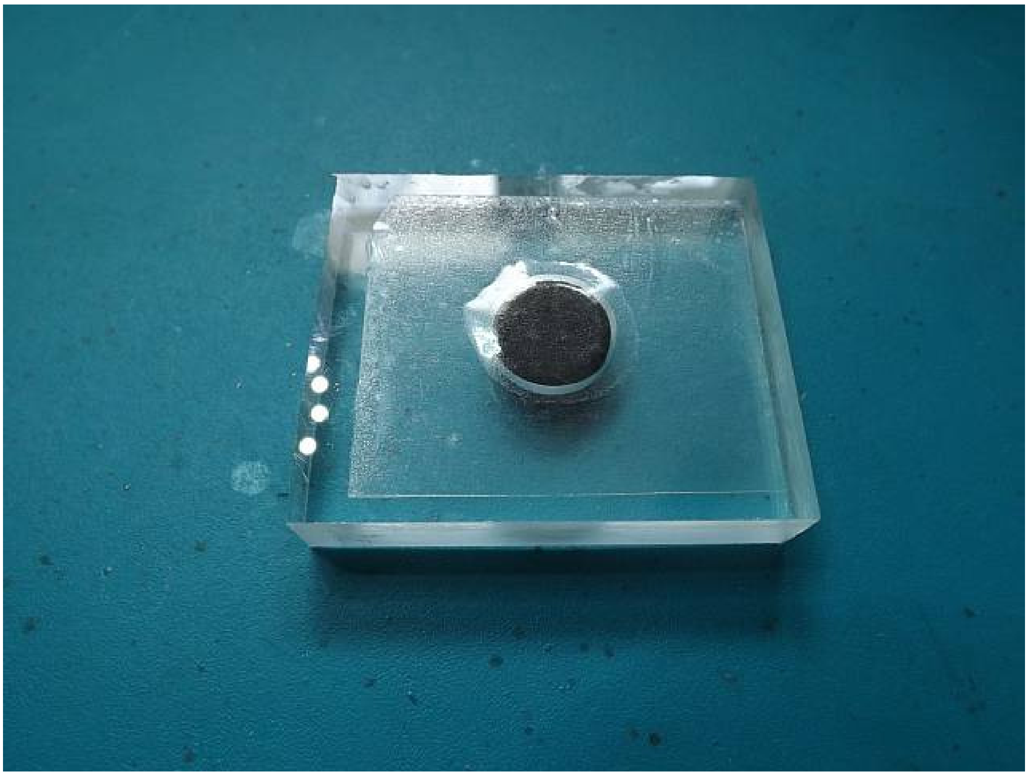

Fig . 3b: Charcoal tablet sealed with radon tight adhesive foil (e.g. HAWE 8000 foil)

Fig. 4: Through-the-foil measurement using the fixture and a Gammascout

Right after sealing the fixture is positioned in front of the active window or the most sensitive portion of the Geiger counter with minimum distance. After measuring the background the pulse counting for each hour is continued. Now the radioactivity of the radon decay takes place through the foil (or better the radioactivity of the beta- and gamma emitting decay products that are in equilibrium with the radon). The emitted alpha radiation of the radon can't penetrate the foil and will not be measured.

The measurement should also last about 3 half-lifes yielding about 100 measurement values. After the measurement time has passed the obtained values (100 values background and 100 values with sample) are read out from the Geiger counter instrument and are post-processed in a spreadsheet such as MS Excel. The background rate is calculated by averaging the first 100 values taken without the sample. The net rate of the sample activity is then obtained by subtracting the averaged background rate from the second 100 values taken with the sample. The net rate is finally plotted versus the time. The resulting graph is showing the exponential decay and the half-life can directly be read out at the 50% point of the initial activity (this is reached after about 91 hours). When a logarithmic scale is used for the activity plot a straight line is visible in the graph and the half-life may be determined more accurately from the slope.

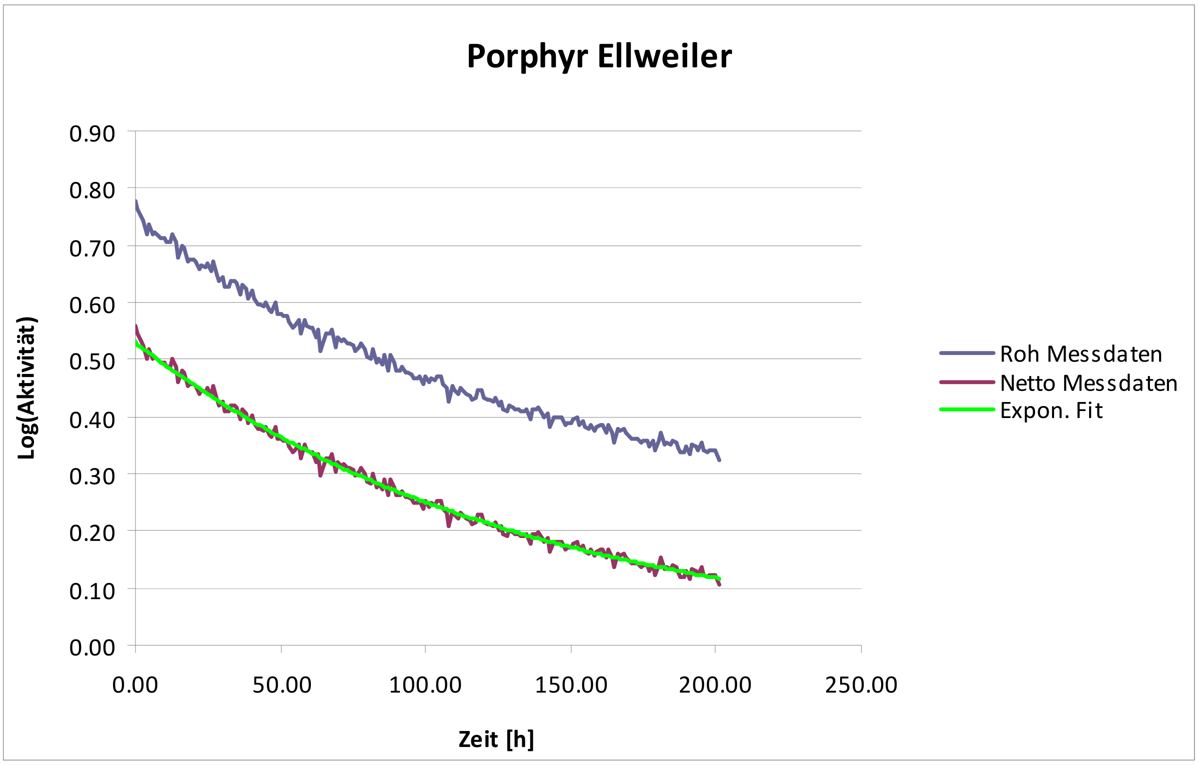

Fig. 5a: Analysis of the Gammascout measurement data with a MS Excel spreadsheat

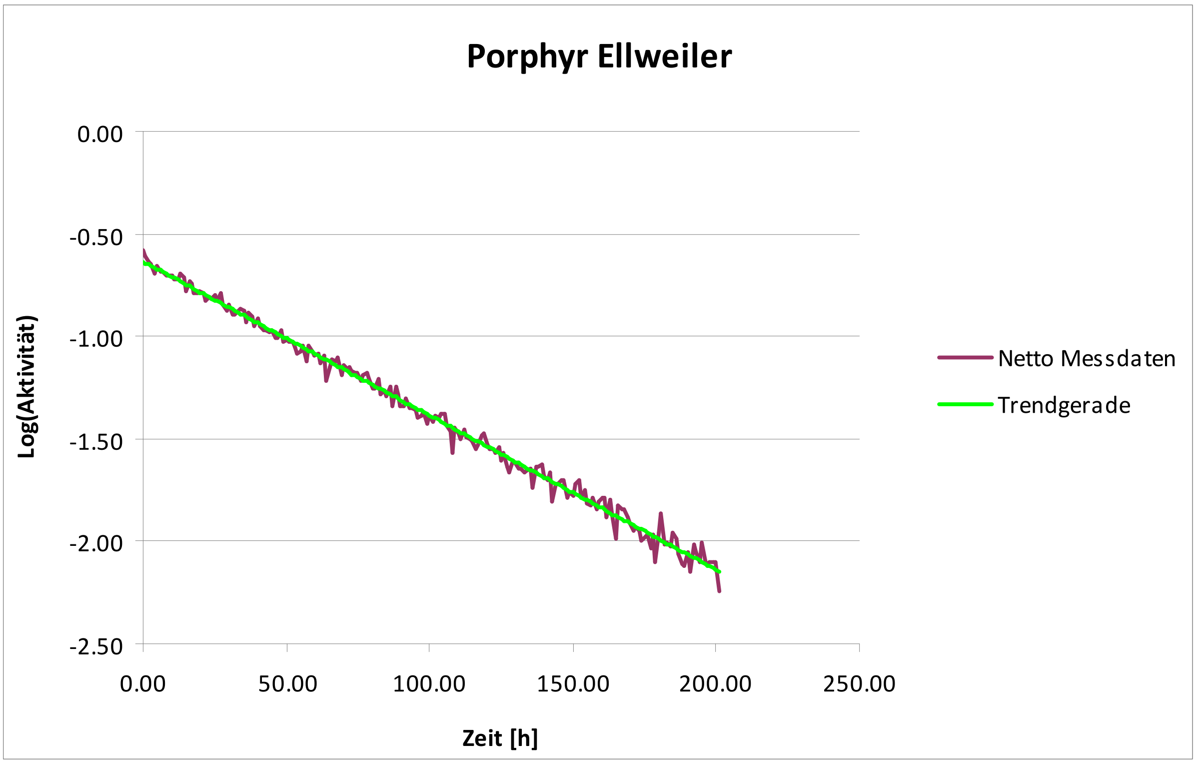

Fig. 5b: Semi-logarithmic plot of the measuremen data

The straight line may be fitted as a trend line into the data using a regression analysis in a least-mean square sense. In case of the Porphyry stone from Ellweiler, a measurement with the Gammascout Geiger counter and regression analysis resulted in a half-life of 91.15h (3.84days). The value is impressively close to the expected 3.82 days and additionally shows that the used sealing foil was indeed radon-tight.

<u>Measurement with the self-made instrument “Tino”</u>

The experiment described above is even more instructive and fascinating when the instrument used for measuring the radioactivity is self-made. This is pretty simple with the “Tino Kit” from the opengeiger.de website and some minimum programming knowledge for an Arduino micro-controller. The Tino (Teviso Sensor + Arduino) uses a PIN-diode detector module supplied by the Swiss company Teviso. It is a small module that only requires a 5V supply and produces digital counting pulses at its digital signal output. The digital pulses may be counted and displayed using the Arduino. The fact, that the sensor module doesn’t require a high voltage such as Geiger-Muller tubes makes it particularly suitable for education purposes.

The Teviso Sensor module has a pretty small active window area that fits perfectly to the size of a charcoal tablet and therefore gets irradiated completely. As a result the dose rates are much higher as it would normally be expected, especially when the tablet is positioned in close vicinity of the sensor window. The reason is the extreme radiation geometry for the tablet in front of the Teviso sensor window that hardly can be achieved with a regular Geiger-Muller tube.

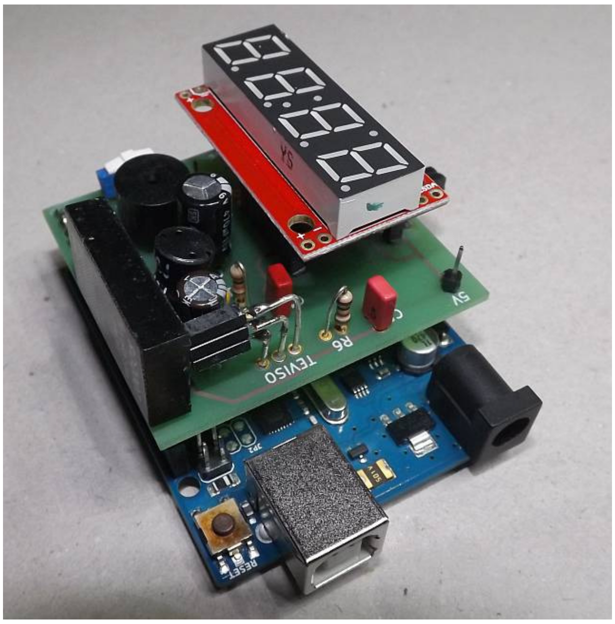

Fig. 6: The DIY measurement instrument „Tino“ with the Teviso radioactivity sensor module and an Arduino microcontroller

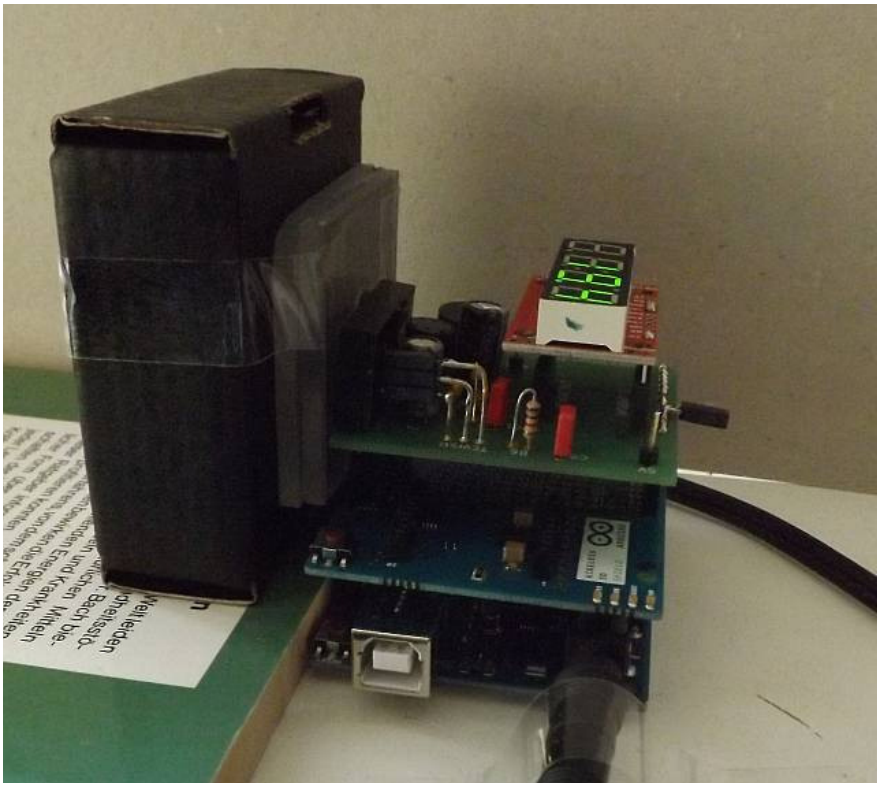

Fig. 7a, b: Measurement with the Tino instrument and recording of the data to a SD-card (located on the middle PCB behind the instrument)

What is recommended as an add-on to the Tino kit is a SD-card shield (piggy-back PCB) that is put between Arduino and Tino-PCB. Using such a SD-card as a non-volatile storage, the measurement values produced every hour for 12 days can easily be stored in a file. A readily available library for creating, writing and reading to and from files on a SD-card are included in the Arduino software that comes for free. After storage of data the card is removed and inserted into a PC card-slot, from where the data are transferred into a spreadsheet for post-processing on the PC.

The most cheap and simple type of Arduino named “Uno” and a SD-card shield with female headers on the top side (e.g. ITead Studio Stackable SD card Shield or Arduino Wireless SD card shield without wireless module) are fully sufficient for the Tino radioactivity measurement instrument. The only thing that is important for programming is to check at what digital pin the chip select (CS) of the SD card appears. For the newer cards this is the pin 4 (for the older cards it was pin 10). In any case it is required to declare pin 10 as output, even though pin 4 is used to initialize the SD-card inside the sketch with the SD.begin(4) statement. All other programming is relatively easy going. The pins 2, 5, 6 and 7 will be used by the Tino.

In the following an example for an Arduino sketch (program) is given. It records the time in milliseconds since program start and the respectively measured dose rate for a Teviso RD2007 sensor and writes the values into a file on the SD-card separated by a semicolon. This program is a simple variation of the Tino sketch on the opengeiger.de website with just the output to the SD-card added. Since there is plenty of time for calculating a stable count rate, the parameter MAXCNT is set to 100 in this application. This way the sketch measures the time that passes until 100 pulses are registered and calculates the rate from these numbers. This also causes the standard deviation of the statistics (Poisson-type) to be always 10% of the respective activity .

```
#include <SD.h>
#include <SPI.h>
#include <SoftwareSerial.h>

#define MAXCNT 100
#define CalFactor 3.4

SoftwareSerial Serial7Segment(7, 6);
char tempString[10];

char fileName[15] = "datalog.txt";
File myFile;
volatile int counter = 0;
unsigned long oldTime = 0;
unsigned long oldRepTime = 0;
float rate = 0.0;

int speaker = 5;
```

```
void setup()
{
  pinMode(speaker, OUTPUT);
  Serial7Segment.begin(9600);
  Serial7Segment.write('v');
  Serial7Segment.write(0x77);
  Serial7Segment.write(0b00000100);
  Serial.begin(9600);
  pinMode(10, OUTPUT); //SD card CS
  digitalWrite(speaker, HIGH);
  if (!SD.begin(4)) {
    Serial.println("SDcard not ready\n");
    return;
  }
  if (!SD.exists(fileName)) {
    myFile = SD.open(fileName, FILE_WRITE);
    myFile.println("###");
    myFile.flush();
  }
  else {
    myFile = SD.open(fileName, FILE_WRITE);
    myFile.println("-----");
    myFile.flush();
  }
  digitalWrite(speaker, LOW); // status ok
  int i = (int)(rate*10.0);
  sprintf(tempString, "%4d", i);
  Serial7Segment.print(tempString);
  attachInterrupt(0, count, RISING);
}

void loop() {
  unsigned long time;
  unsigned long dt;
  unsigned long repTime;

  detachInterrupt(0);
  time = millis();
  if (counter >= MAXCNT) {
    dt = time-oldTime;
    oldTime = time;
    counter = 0;
    oldRepTime = millis();
    rate = (float)MAXCNT*60.0*1000.0/(float)dt/CalFactor;
    Serial.print(time);
    Serial.print(";");
    Serial.println(rate);
    myFile.print(time);
    myFile.print(";");
```

```
    myFile.println(rate);
    myFile.flush();
    int i = (int)(rate*10.0);
    char tempString[10];
    sprintf(tempString, "%4d", i);
    Serial7Segment.print(tempString);
  }
  attachInterrupt(0, count, RISING);
}

void count()
{
  counter++;
  digitalWrite(speaker, HIGH);
  delayMicroseconds(50000);
  digitalWrite(speaker, LOW);
}
```

Listing of the data logging and display sketch for the Arduino based measurement instrument “Tino“

A special additional feature of the program is, that the piezo-speaker of the Tino creates a short beep when the power is turned on or a reset is executed. In case the SD-card is not ready for writing the tone will not stop and thus indicates an error. As long as the file with the name “datalog.txt” will not be deleted, the sketch simply appends further measurement data after inserting a dash line as a separator. Otherwise the file is simply created from scratch on the SDC card.

Post-processing and data analysis

The recorded measurement values describe the course of the radon decay activity over time. This function has the form:

$A(t)= A_0*\exp(-\lambda*t)$

The constant λ is the decay constant. The half-life $T_{1/2}$ results from the decay constant as follows:

$T_{1/2} = Ln(2)/\lambda$

The analysis of the data and the extraction of the half-life may be significantly simplified when the logarithm of the net count rate is computed (the net-count rate data are the measurement data subtracted by the background). For the natural logarithm of the net count rate we obtain:

$Ln(A(t)) = Ln(A_0) -\lambda*t$

Putting y(t) = Ln(A(t)) and b = Ln(A_0), then y(t) gets the form of a straight line, with the slope representing the decay constant λ and b = Ln(A_0) representing the intercept with the axis. When now the measurement values are superimposed by noise of the statistical nature of the decay process, a regression analysis may be used to fit a trend line into the logarithmized noisy data reducing the mean square error to a minimum. To do this, spreadsheets like MS Excel provide ready to use functions like slope() and intercept() . These functions can be applied to the array of the logarithmized net count data as y-data and the time data since start of the measurement as x-data. The functions yield the slope and intercept of the trend line that best fits into the logarithmized data. The slope directly represents the decay constant (which is $0.0076 \cdot h^{-1}$ for radon) and the intercept represents the logarithm of the initial activity as estimated with the regression. This initial activity is a rough measure for the exhalation potential of the sample.

Results from the Tino measurements

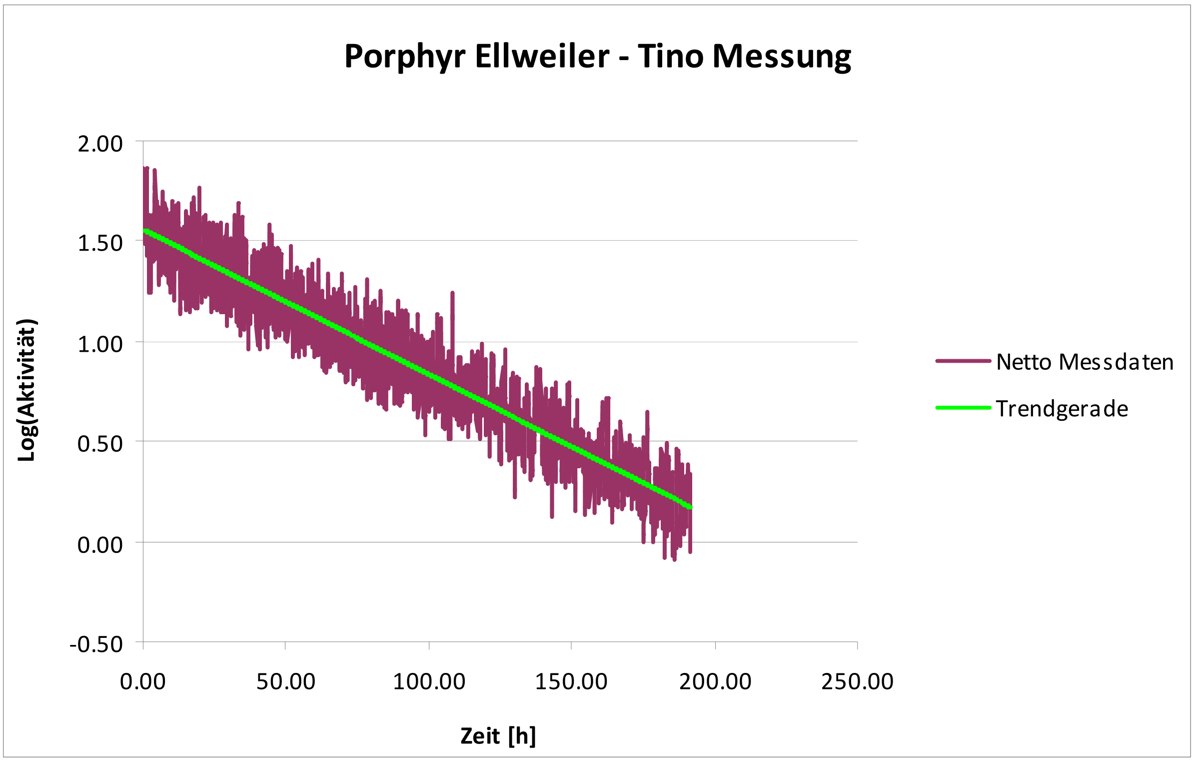

Fig. 8: Measurement result with the self-made Tino instrument

A measurement on the porphyry stones from Ellweiler with the Tino instrument and the commercial Gammascout instrument in parallel yields a completely comparable result. It therefore becomes obvious, that even with a DIY instrument the radon exhalation from building material can be proven. The only difference to the measurement with the Gammascout is the higher noise caused by the lower count rate of the Teviso sensor module. Since less pulses per measurement value are used to obtain a similar temporal resolution of 1h, the variance is respectively higher. By calculation of the trend line however, this noise is completely eliminated.

<u>Background information on the experiment</u>

The method of using activated charcoal that binds the noble gas radon by physisorption and to seal the charcoal during measurement to proof the exhalation from bulding materials with the half-life was originally developped by G. Just, H v. Phillipsborn and R. Geipel. This method was just improved using charcoal tablets and a dedicated fixture for the tablets. This helps the handling of the activated charcoal that is a bit tedious when it is powdered.

Most important is the understanding of the equilibrium within the uranium-radium decay chain. Primarily the uranium U-238 contained dominantly in the natural uranium has a half-life of 4.5 billion years.

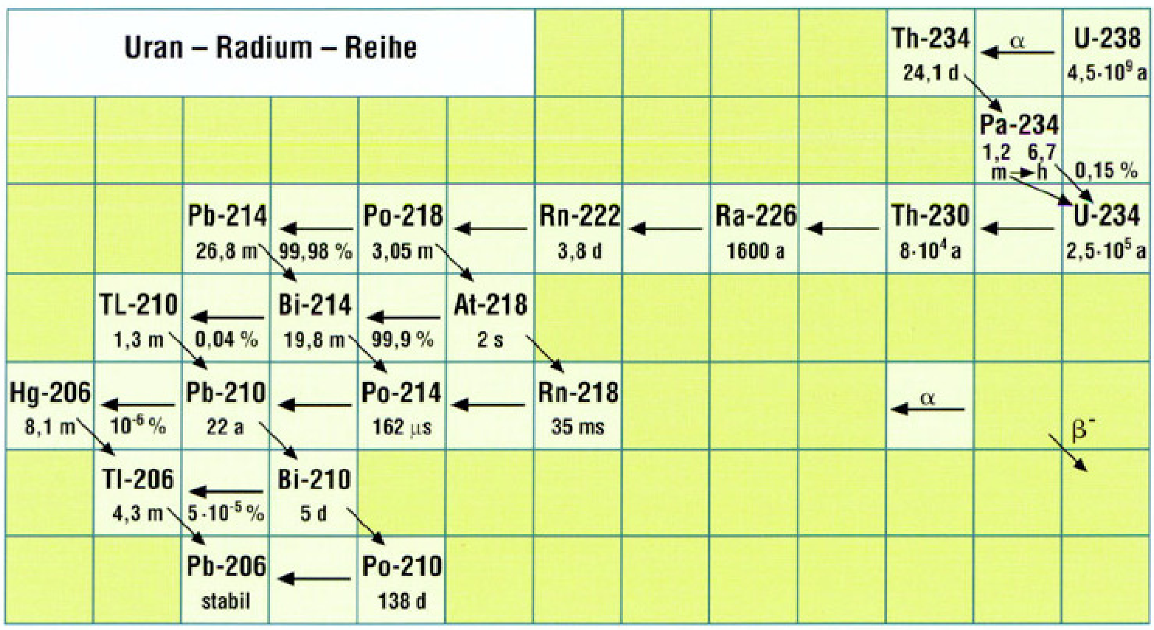

Fig. 9: Uran-Radium decay chain

In the decay chain all other decay products have half-lives smaller by orders of magnitude down to the radium. Therefore it can be assumed that uranium found in the undisturbed nature contains all decay products of the decay chain in parallel with a similar activity as the U-238 itself (a so-called secular equilibrium). This happens because all decay products are generated sequentially one after each other and decay again in average long before the next uranium nucleus decays. This means additionally that the activity of the natural uranium is higher than that of the pure U-238 contained in the mixture when it would be present alone.

Now, there is one abnormality in the decay chain. The radium does not decay into a solid material but into the noble gas radon (Rn-222) which is highly mobile. When this radon is not enclosed gas-tight until it decays again into a solid decay product, it escapes (exhales) and mixes itself with air. It then decays further in the air and its decay products get attached to aerosols and they decay further. These radioactive aerosols may be inhaled with the breathing air and are finally deposited in the respiratory tract where they become dangerous.

Starting point for the radon is always the radium. Radon may only be generated from radium, therefore the method is also a good proof for the existence of radium. Even when all uranium gets removed from a sample, when there is still some radium radon will always be generated too.

Radium has a half-life of 1600 years that also is by far longer than all half-lives of the following decay products including radon with its 3.82 days (91 hours). This again means that when radium is present in a sample more than 3 half-lives of one of its decay products, then these decay products have the same activity due to the secular equilibrium to the radium.

From the U-238 down to the radium all decays accur with alpha radiation that are difficult to measure with simple means. Among the radon decay product however, mainly the Pb-214 and the Bi-214 are beta emitters that in parallel also emit gamma radiation in significant quantities and therefore are easily measurable. As a consequence, as long as the the mother nuclide radon is in equilibrium with its decay products (the so-called radon-daughters) it is possible to determine the radon activity from the daughter activities. This is also the case when the radon has escaped from the original sample and got bound on the activated charcoal.

The binding of radon to the activated charcoal however is a process, that also is following a thermodynamic equilibrium. Depending on temperature the absorption is competing with a desorption. When measuring a charcoal tablet “loaded” in a radon-rich atmosphere and afterwards is not enclosed tightly, then it loses its activity within hours mainly due to desorption. However, when the tablet gets sealed in a minimum remaining air volume, the radon decays completely in this volume. When a radon tight foil is used the radon can’t escape and its beta and gamma emitting decay products can be measured easily through the foil. Since the radon activity decays with the half-life of 3.82 days the decay products also vanish with the same half-life. Actually, what is measured is not the radiation generated by the radon but the radiation of its decay products Pb-214 and Bi-214 that just map the half-life of the radon through the foil. However this process would not happen without the presence of the radon inside, therefore this proof is unambigous.

From a quantitative point of view, this proof of radon exhalation is not really fully exact. It would be possible to calibrate the measurement with a reference sample but since the radon exhalation strongly depends on the way the sample was crushed and its surface condition, there are strong differences in the activity to the mother nuclide radium contained in the sample and the radon in the surrounding air volume. Additionally the radon in the air is not in a perfect equilibrium with its daughters. Humidity, aerosol density and plate-out effects on the walls of the sample container play a certain role. Therefore the presented method only gives a gut feeling what material exhales a lot and what material exhales less radon.

Literature

/1/ H. von Phillipsborn, R. Geipel, G. Just
Radon Exhalation from Building Materials – A contribution to Metrology in Radiation Protection
Proceedings of the 5. international conference on high levels of natural radiation and radon areas: Radiation dose and health effects; Munich (Germany); 4-7 Sep 2000

/2/ Bundesamt für Strahlenschutz (BfS)
What is radon?
http://www.bfs.de/EN/topics/ion/environment/radon/introduction/introduction.html
/3/ WHO Handbook on Indoor Radon
http://whqlibdoc.who.int/publications/2009/9789241547673_eng.pdf

/4/ Radiological Protection Principles concerning the Natural Radioactivity of Building Materials, European Commission, Radiation protection 112, 1999
http://ec.europa.eu/energy/sites/ener/files/documents/112.pdf

/5/ D. LAURIER
Progress in understanding radon risk
Institute for Radiological Protection and Nuclear Safety (IRSN)
EU Scientific Seminar 2010 "Issues with internal emitters“
Luxembourg, 23 November 2010
http://ec.europa.eu/energy/sites/ener/files/documents/laurier_understanding_radon_risk.pdf

/6/ TEVISO Sensor Technologies Ltd.
http://www.teviso.com

/7/ The Radiation Measurement Shield „Tino“ for the Arduino
and Arduino-Compatible Microcontrollers
http://opengeiger.de/TinoKitDescription.pdf

/8/ Hawe Hugentobler AG
Adhesive foil Type HAWE 8000
http://www.hawe-folien.ch/

Appendix

Fig. 10: Wrist watch with radium pointers and an arctivated charcoal tablet in a jam jar

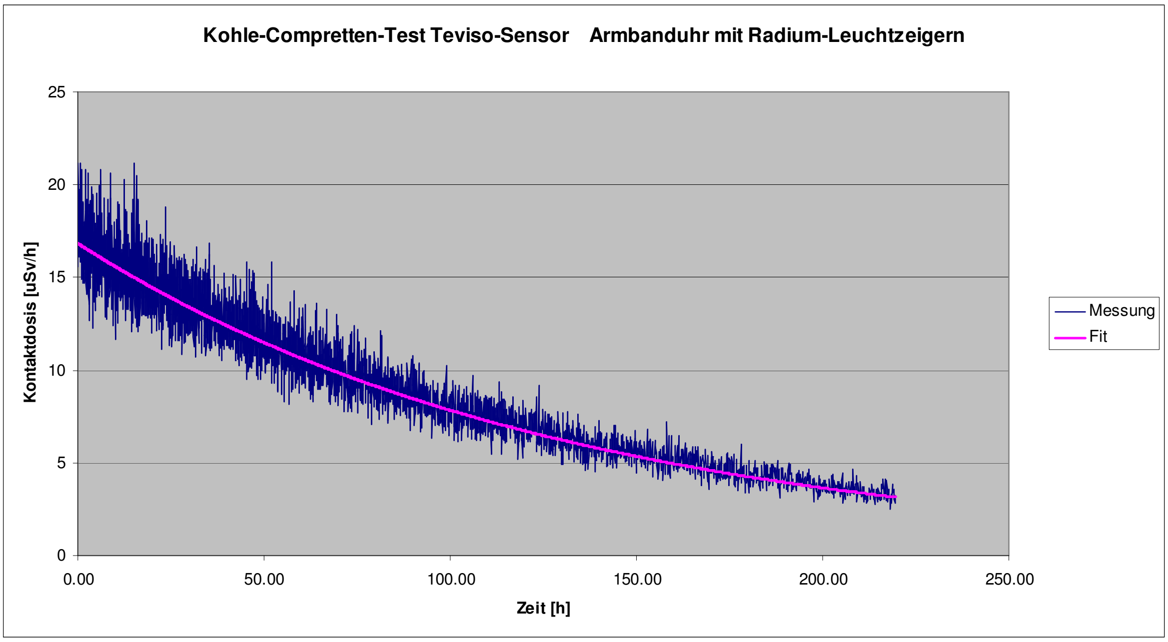

Fig. 11: Measurement of a charcoal tablet, loaded from the wrist-watch with radium pointers using the Tino with SD-card for data logging (measurement of the count rate with 100 pulses and following conversion into a dose rate according to the factor given by Teviso)