# **Results of three years search for the <sup>213</sup>Po half-life variations**

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**Abstract** Description of the TAU-3 installation intended for long-term monitoring of the half-life value  $T_{1/2}$  of the <sup>213</sup>Po is presented. Isotope <sup>229</sup>Th is used as a source of the mother's chain. The methods of measurement and processing of collected data are reported. Solar-daily variation with an amplitude  $A_{So}=(5.3\pm1.1)\times10^{-4}$ , lunar-daily variation with an amplitude  $A_{L}=(4.8\pm2.1)\times10^{-4}$  and sidereal-daily variation with an amplitude  $A_{S}=(4.2\pm1.7)\times10^{-4}$  were found as a results of a processing of the 622 days data series (July 2015 – March 2017). An averaged value of the <sup>213</sup>Po nuclei decay half-life was found to be to  $T_{1/2}=3.705\pm0.001\,\mu$ s. A half-life value data set with the week duration step was constructed for the 1177 days measurement time (July 2015 – September 2018). Features of the half-life time behavior were analyzed. Annular variation with an amplitude  $A=(3.6\pm0.6)\times10^{-4}$  was found.

Keywords: Half-Life, <sup>213</sup>Po Nucleus, Daily and Annual Variations

## **1. Introduction**

Experimental research of the <sup>214</sup>Po half-life ( $\tau$ ) time stabilities is carried out at the Baksan Neutrino Observatory of the INR RAS [1-3] since 2008. A half-life is defined as a result of an analysis of the decay curves constructed from a set of life-time values of separate nuclei of the isotope under consideration. Delays between a birth of the nuclear ( $\beta$ -particle from the <sup>214</sup>Bi decay +  $\gamma$ -quantum) and its decay ( $\alpha$ -particle from the <sup>214</sup>Po decay) are measured to define this parameter.

Half-life value time sequences with different time steps are the objects for a subsequent analysis. The measurements (973 days) are performed at the TAU-2 low background facility placed in the underground low background laboratory DULB-4900 at a depth of 4900 m.w.e.. Further, time series of  $\tau$  with different temporal steps are analyzed. According to data obtained at TAU-2, the averaged value of the <sup>214</sup>Po half-life is  $\tau = 163.47 \pm 0.03 \ \mu$ s. The annual variation with amplitude  $A = (9.8 \pm 0.6) \times 10^{-4}$ , the solar-daily variation with amplitude  $A_{\text{So}} = (7.5 \pm 1.2) \times 10^{-4}$ , the lunar-daily variation with amplitude  $A_{\text{L}} = (6.9 \pm 2.0) \times 10^{-4}$ , and the sidereal daily variation with amplitude  $A_{\text{S}} = (7.2 \pm 1.2) \times 10^{-4}$  are detected in the series of  $\tau$  values. Another pair of radioactive isotopes which have similar decay diagram but much smaller half-life of the daughter isotope was proposed to proof reality of the obtained

variations. The <sup>213</sup>Bi( $T_{1/2} = 46 \text{ min}$ )  $\rightarrow$  <sup>213</sup>Po( $T_{1/2} = 4.2 \text{ µs}$ ) [5] were choose as the pair. These isotopes are the daughter products in the <sup>229</sup>Th( $T_{1/2}=7340$  years) decay chain from the <sup>237</sup>Np series [6]. The results obtained for the data measured during 1177 days with such source are listed in the presented work.

#### 2. The facility description

The construction of the TAU-3 facility with a <sup>229</sup>Th source is similar to that of TAU-2 [1]. It comprises a scintillation detector D1 which is made of two disks d = 18 mm and h = 1 mm of a plastic scintillator (PS) glued together. The radiation source <sup>229</sup>Th ( $T_{1/2} = 7340$  years) positioned between the disks is the parent isotope for <sup>213</sup>Po. The test sample is manufactured at the Khlopin Radium Institute (St. Petersburg).

The source is precipitated from  $Th(NO_3)_4$  salt solution on the surface of a LAVSAN film with  $h = 2.5 \mu m$  and covered by the same film pasted along the edge by the epoxy resin. The assembly is placed at the bottom of a case made of VM-2000 reflecting film open from one end. The case is put inside a stainless-steel rectangular case  $9 \times 23 \times 140$  mm, thickness 0.5 mm. The open end of the case is connected with the bottom of a 2.5-mm stainless-steel cylinder with d =44 mm, and h = 160 mm. Inside the cylinder, there is a high-speed FEU-87 photomultiplier monitoring PS. The signal is taken from the FEU anode load through the matching circuit and is supplied via the cable (50 Ohm) to the first entry of the registering unit. Detector D1 is placed in the 15-cm Pb protective layer in a gap with h = 10 mm between two scintillation detectors NaI(Tl) 150×150 mm (detector D2) in a low-background box of the DULB-4900 underground low-background laboratory [6]. Signals from the anodes of two photomultipliers of the D2 detector are amplified by charge-sensitive preamplifiers, summed, and supplied to the second, starting entry of the registering unit. The registering facility comprises a LA-n10-12 PCI digital oscilloscope (DO) integrated with a PC, which is registering the waveform of pulses arriving from D1 and D2 in the online mode. The frequency of pulse digitization in DO is chosen as 100 MHz. The reading and recording are started by a pulse in the D2 channel. The record frame is 2048 temporal channels (10 ns per channel), including 256 channels of prehistory and 1792 channels of history. In Fig. 1, the decays of <sup>213</sup>Bi and <sup>213</sup>Po isotopes [5] are presented schematically. From Fig1a it follows that 66% of β decays of <sup>213</sup>Bi are transitions to the ground level, and 31% to the excited level with an energy of 440 keV. The decay of this level is accompanied by a  $\gamma$  quantum emission (26% per decay). The isotope <sup>213</sup>Po decays in 100% of cases with emission of an  $\alpha$  particle with an



Fig.1. Decay schemes of <sup>213</sup>Bi (left) and <sup>213</sup>Po (right).

energy of 8537 keV. If the device registers all three particles released by the decay of the pair of isotopes, it is the event with three pulses. In this event, pulses coming from the γ quantum and β-particle coincide instantaneously, and the pulse from the α particle is delayed. In Fig. 2, one of the events (frames) stored by DO in the PC memory is displayed as an example. The pulse on the upper beam (1) is a γ quantum, the first pulse train on the lower beam (2) corresponds to a β particle, and the second one to an α particle. The observed triple coincidences considerably reduce the contribution of background events accompanying decays of the remaining isotopes in the chain of decays of <sup>229</sup>Th to the total counting rate of the facility. The activity of <sup>229</sup>Th is ~ 80 Bq. Alongside the main isotope there are small amounts of extraneous radioactive impurities in the specimen. The DO recording rate of the event started by D2 pulses with amplitudes of 380-500 keV was ~27 s<sup>-1</sup>. The recording rate of useful events with parameters of all pulses corresponding to <sup>213</sup>Po decay was ~ 18 s<sup>-1</sup>. From Fig.2 it follows that signals from β- and α-particles are clusters of short subpulses with total duration of up to ~ 1 µs, decreasing exponentially in frequency and amplitude.



**Fig2.** An example of  ${}^{213}\text{Bi}-{}^{213}\text{Po}$  pair decay event stored by DO in PC memory: (1) upper beam, a pulse from D2 detector ( $\gamma$  quantum), (2) lower beam, pulses from particle (start), and particle (stop) in D1 detector.

**Fig3.** Decay curve for <sup>213</sup>Po plotted by the data from TAU-3 device obtained over 622 days.

The clusters can overlap at small delays between particles; therefore, the processing program should consider the relation between the amplitudes of the first and subsequent subpulses in a cluster to unambiguously separate the delayed ( $\beta \otimes \alpha$ ) coincidences. The delays between pulses in channel D1 are determined as the result of processing the recorded waveforms, and a decay curve of daughter isotope <sup>213</sup>Po is plotted for the chosen time interval. The half-life determination is based on this curve. The sequential time series of this magnitude is plotted.

### 3. Measurement results

Continuous measurements started at TAU-3 on July 9, 2015. The statistics for 622 days (March 2017) was processed at the beginning. In Fig. 3, the decay curve of the <sup>213</sup>Po isotope is given. The value of  $\tau$  was obtained approximating the decay curve by function



**Fig4.** Dependence of <sup>213</sup>Po half-life on the time of solar day obtained by the method of interior moving average (triangles). Approximation by function  $\tau(t) =$  $\tau_0[1+3.4\times10-4\sin\{2\pi/24(t-3)\}]$  (red curve). Restored dependence  $\tau(t) = \tau_0[1+5.3\times10-4\times\times\sin\{2\pi/24(t-9)\}]$ (the blue dot-dashed curve).



**Fig5.** Dependence of <sup>213</sup>Po half-life on the time of sidereal day obtained by the method of interior moving average (triangles). Approximation by function  $\tau(t) = \tau_0 [1+2.7 \times 10-4sin\{(2\pi/24)(t-19)\}]$  (red curve). Restored dependence  $\tau(t) = \tau_0 [1+4.2 \times 10-4 \times sin\{2\pi/24(t-1)\})$  (the blue dot-dashed curve).

 $F(t) = A \times exp[-ln(2)t/\tau] + b$  using the minimum  $\chi^2$  test in the delay interval of 0.5-13.0 µs. It was found that  $\tau = 3.705 \pm 0.001$  µs. The inner moving-average (IMA) method was used to search for a possible time variation of the  $\tau$ -values. A time interval with the duration equal to about 0.5 of the expected periods is chosen to search for any harmonic component and the  $\tau$ -value is determined for this interval. Then, the interval has shifted by one step and the procedure is repeated. In the studies of daily variations of the <sup>213</sup>Po half-life dependence on solar, sidereal, and lunar time, the length of the respective day was divided into 24 hours. The duration of a sidereal and lunar day in the standard solar time is 23 hours 56 minutes 4.09 s and 24 hours 50 minutes 28.2 s, respectively.

A period of 12 hours was chosen as an interval of averaging. The analysis of events was made as follows. We selected the events registered in the interval of 0-12 hours for the entire period study and determined the half-life values. After that, the interval was shifted by one hour and the procedure was repeated. The results of the search of the daily variation in solar time are given in Fig. 4. Here, the result of approximation of the daily half-life dependence by the function  $\tau(t) = \tau_0 [1 + A \times \sin\{\omega(t + \phi)\}]$  (the red curve) is displayed, where  $\tau_0$  is the mean half-life;  $\omega = 2\pi/24 \text{ h}^{-1}$ ;  $A = 3.4 \times 10^{-4}$  is the amplitude;  $\phi = -3$  h is a phase shift of the initial point of the curve relative to 0 hours. The figure shows that the time dependence of the <sup>213</sup>Po half-life is well described by a sinusoidal function. The period found is 24 hours and the relative amplitude is 0.00034 half-lives. It is easy to show that the initial periodic dependence of the shifted by 0.5 of the moving intervals ( $0.25 \times 24 = 6$  h). The amplitude of the initial daily periodic dependence obtained from these data in solar time is  $A_{so} = (5.3 \pm 1.1) \times 10^{-4}$  (the blue dot-dashed curve).

In Fig. 5, the results of the search for a sidereal daily variation of the <sup>213</sup>Po half-life are displayed. The experimental data are approximated by the curve  $\tau(t) = \tau_0[1 + A \times \sin\{\omega(t + \phi)\}]$  (the red curve) with the parameters  $A = 2.7 \times 10^{-4}$  is amplitude;  $\phi = -19$  h is the phase shift of the curve initial point relative to 0 hours. The analysis of the restored initial dependence similar to the analysis made for the solar-daily wave shows the presence of a sidereal-daily wave with the relative amplitude  $A_s = (4.2 \pm 1.7) \times 10^{-4}$  (the blue dot-dashed curve). In Fig. 6, the



results of search for a lunar-daily variation of the <sup>213</sup>Po half-life are given.

**Fig6.** Dependence of <sup>213</sup>Po half-life on the time of lunar day obtained by the method of interior moving average (triangles). Approximation by function  $\tau(t)=\tau 0[1+3.1 \times 10-4sin{2\pi/24(t-14)}]$  (the red curve). Restored dependence  $\tau(t)=\tau 0[1+4.8 \times 10-4sin{2\pi/24(t-20)}]$  (the blue dot-dashed curve).

The analysis of the restored initial dependence like the analysis made for the solar-daily wave shows the presence of a lunar-daily wave with relative amplitude  $A_L = (4.8 \pm 2.1) \times 10^{-4}$  (blue dot-dashed curve). In Fig. 7, the time dependence of  $\tau$  obtained from the decay curve for a weekly data set is presented. It is shown that  $\tau$  increases with time, and that for a data set collected over 127 days,  $\tau = (3.6998 \pm 0.0015) \,\mu$ s; for 320 days,  $\tau = (3.6993 \pm 0.0014) \,\mu$ s; for 422 days  $\tau = (3.7016 \pm 0.0011) \,\mu$ s, and for 622 days  $\tau = (3.7053 \pm 0.0011) \,\mu$ s. The causes of such behavior of the  $\tau$  parameter are not clear yet. It could be both an instrumental effect, for example, equipment ageing, and an unknown real physical effect. The presence of a pulse surge of data within the time interval comparable to a year in the series of weekly data hinders using the method of moving internal average for studies of the half-life annual variation.

The data collected during 1177 days (28 September 2018) allows us to suppose that the obtained trend has a shape of a logistic curve which can be done by an expression of L=A<sub>2</sub>+(A<sub>1</sub>-A<sub>2</sub>)/[1+(x/x<sub>0</sub>)<sup>P</sup>]. The coefficients in the formula fitted the experimental data in the best way were found by the  $\chi^2$ -method as L=3.733+(3.700-3.733)/[1+(x/73.42)<sup>4.94</sup>]. The experimental data was normalized to the values of fitting curve. The results are shown in Fig.8 by points with error bars. This dependence was smoothed by a sliding averaging by the 26 points method at the ORIGIN 8.5 program. The result is shown in Fig.8 by the blue curve which contains the annular variation. This curve was approximated by a sine function  $\tau(t)/\tau_0=1+2.3\times10^{-4}\times\sin((2\pi/365)\times(t-319))$  shown by red color in Fig.8. The amplitude of the functions is A=(2.3\pm0.4)×10<sup>-4</sup>. As mentioned above, this value was integrated over 26 points (the 0.5-year period) in the averaging process. A real value of the annular variation could be found by multiplying of sine amplitude by  $\pi/2$  coefficient. This gives an amplitude of the variation equal to A=(3.6\pm0.6)×10<sup>-4</sup>.

### 4. Discussion

The results of a monitoring of the <sup>213</sup>Po half-life in the period July 2015 – September 2018 presented above show that this parameter undergoes solar-daily, sidereal-daily and lunar-daily variation with the amplitudes  $A_{So}=(5.3\pm1.1)\times10^{-4}$ ,  $A_{S}=(4.2\pm1.7)\times10^{-4}$  and  $A_{L}=(4.8\pm2.1)\times10^{-4}$  respectively. These values coincide within the errors with those obtained in the <sup>214</sup>Po half-life values series. A search for the annular variations in the <sup>213</sup>Po data is complicated because of appearance of a nonperiodic unidirectional deviation of the half-life

values from the averaged one. The process became noticeable in the May - June 2016 data time range. The effect could be caused by the electronics aging



1,008 1,000 0,095 0,995 0,

Fig7. Time dependence of  $\tau$  obtained from the decay curve for the weekly data set (time distance of measurements: July 9, 2015 – September 28, 2018). Trend shape L=3.733+(3.700-3.733)/[1+(x/73.42)<sup>4.94</sup>] – the red curve.

**Fig8.** Time dependence of  $\tau$  normalized on trend – the black dots. Time dependence of smoothed  $\tau$ -values – the blue curve. Approximation by function  $\tau(t)/\tau_0=1+3.6\times10^{-4}\times sin((2\pi/365)\times(t-319))$  – the red curve.

or some unknown physical factors. A length of the half-life series collected to the present moment is enough for estimation of the trend shape. Analysis of the dependence obtained from primary data normalization to the trend shape shows a presence of the annular variation with the amplitude  $A=(3.6\pm0.6)\times10^{-4}$ .

#### 5. Conclusion

Description of the TAU-3 installation intended for long-term monitoring of the half-life value  $T_{1/2}$  of the <sup>213</sup>Po is presented. The isotope <sup>229</sup>Th is used as a source of the mother's chain. The methods of measurement and processing of collected data are reported. Solar-daily variation with amplitude  $A_{So}=(5.3\pm1.1)\times10^{-4}$ , lunar-daily variation with amplitude  $A_{L}=(4.8\pm2.1)\times10^{-4}$  and sidereal-daily variation with amplitude  $A_{S}=(4.2\pm1.7)\times10^{-4}$  were found from treatment of the 622 days data series (July 2015 – March 2017). An averaged value of the <sup>213</sup>Po nuclei decay half-life was found to be equal to  $T_{1/2}=3.705\pm0.001\mu$ s. A half-life value data set with the week duration step was constructed for the 1177 days measurement time (July 2015 – September 2018). Features of the half-life time behavior were analyzed. The annular variation with the amplitude  $A=(3.6\pm0.6)\times10^{-4}$  was found.

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