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ДОСЛІДЖЕННЯ РАДІОАКТИВНОГО ГРАФІТУ У ЛІСОВІЙ ПІДСТИЛЦІ

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SEEKING FOR RADIOACTIVE GRAPHITE IN THE FOREST LITTER

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Keywords: ^{14}C, LSC, carbide, benzene, vacuum pyrolysis, forest litter, radioactive graphite.

Radiocarbon analyses have wide application. Sample materials used for study of ^{14}C concentration may have sub-materials with different radiocarbon concentration or sub-materials may have different particular thermo-destruction peculiarities, which may cause radiocarbon variation in corresponding counting media (benzene). Thus, talking about ^{14}C concentration in sample, one take into account, what he want to know: anything, just average and/or sub-components mentioned above. To have measured reproducible any of component, sample mass should be taken in reproducible amount.

Radioactive graphite of Chornobyl NPP's releases

ДОСЛІДЖЕННЯ РАДІОАКТИВНОГО ГРАФІТУ У ЛІСОВІЙ ПІДСТИЛЦІ

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***Метою роботи** є запровадження диференційного методу дослідження ^{14}C у пробах довкілля неоднорідних за вмістом та за особливостями їхньої термодеструкції, оцінка чутливості та відтворюваності методу.*
***Матеріали і методи.** Використовували традиційний метод досліджень ^{14}C на основі рідинно-сцинтиляційного обчислення, застосовували вакуумний піроліз, зокрема для покомпонентної підготовки проб лісової підстилки у пошуках радіоактивного графіту.*
***Результати і висновки.** Ми порівняли результати застосування технології вакуумного піролізу (Skripkin & Kovaliukh, 1997) для отримання зразків бензолу, де фракції одного і того ж зразка були оброблені разом або окремо. Підготовлена окремо друга з двох фракцій зразків систематично дає на 51.5% більше ^{14}C порівняно зі спільним зразком з двох фракцій (R^2=0.9554).*

Ключові слова: ^{14}C, РСЛ, карбід, бензол, вакуумний піроліз, лісова підстилка, радіоактивний графіт.

into environment high ^{14}C activity in small volume. Behavior of those radioactive graphite was studied on forest ecosystem [3, 4].

Spatial distribution of Chornobyl radioactive graphite was estimated on samples of forest litter and upper soil layer collected during summer of 1997 [2].

Table

Radiocarbon (pMC)[a] in the samples of various layers of forest litter for observation sites. Coordinates of sampling sites for studying ^{14}C in the Chornobyl NPP vicinity (E-longitude, N-latitude) [1, 2]

Site	Longitude (E)	Latitude (N)	Layer	^{14}C, pMC		
				«Two fractions»	«Pyrolysis»	«Charcoal»
96/04	30°5'15''	51°22'40''	Total	385.3	193.9	451.8
96/13	30°4'0''	51°23'25''	A	132.2[c]	121.3	130.2
96/13	30°4'0''	51°23'25''	B	127.1[c]	124.8	126.1
96/13	30°4'0''	51°23'25''	C	152.4[c]	134.9	156.2
96/17	30°7'25''	51°21'50''	A	125.0[c]	126.6	126.1
96/17	30°7'25''	51°21'50''	B	135.8[c]	131.7	132.4
96/17	30°7'25''	51°21'50''	C	155.8[c]	137.0	154.0
96/33	30°3'38''	51°23'25''	A	122.5	119.6	120.3
96/33	30°3'38''	51°23'25''	B	126.9	121.5	120.7
96/33	30°3'38 ''	51°23'25 ''	C	132.7	123.9	130.6
96/34	30°4'30''	51°23'26 ''	Total	135.5	122.8	132.1
96/36	30°4'30''	51°24'0''	Total	188.1		
96/36	30°4'30 ''	51°24'0''	A	125.0[c]	124.8	127.0
96/36	30°4'30 ''	51°24'0''	B	134.2[c]	124.4	130.5
96/36	30°4'30''	51°24'0''	C	207.1[c]	142.3	226.0
97/06	30°6'20''	51°25'10''	A	123.0	119.2	122.9
97/06	30°6'20''	51°25'10''	B	123.8	127.1	129.5
97/06	30°6'20''	51°25'10''	C	149.1	126.1	132.1
97/08	30°0'55''	51°29'5''	A	116.1	115.9	116.7
97/08	30°0'55''	51°29'5''	B	121.3	115.5	120.7
97/08	30°0'55''	51°29'5''	C	126.8	118.1	129.6
97/24	30°1'52''	51°23'3''	A	126.8	121.8	121.2
97/24	30°1'52''	51°23'3''	B	147.8	126.0	136.2
97/24	30°1'52''	51°23'3''	C	561.2		742.1
TL[b]	—	—	A	130.0	129.0	133.3
TL[b]	—	—	B	272.0	161.0	332.1
TL[b]	—	—	C	378.0	191.6	655.5
97/25			Total		116.4	117.0

[a] pMC is the percent of «modern» ^{14}C, a relative unit that expresses the ^{14}C activity in the carbon of the Oakwood of 1890 growth (100 pMC = 226 Bq/kgC).

[b] The sampling site is outside our network.

[c] Old data, which are presented first.

In this case, the forest litter samples were collected from the ground surface and were sampled from an area of 2500 cm^2 (50 x 50 cm). The samples were stratified by 3 layers according to their morphological state:

A – upper layer: fresh sagged needles;

B – middle layer: old needles without structural changes;

C – lower layer, adjacent with upper soil layer: destroyed needles and other humus-like substances.

The relative average distribution of the mass of forest litter samples for 38 sites was found to be A = (13 ± 5)%; B = (29 ± 10)%; and C = (58 ± 11)% [2].

Samples of total litter were collected in 3 sites where we could not stratify the forest litter by layers.Results of above cited paper were obtained using of two stages vacuum pyrolysis [5] for processing of corresponding organic material of layer. We found that application of those approach is quite sensitive for discovering of graphite deposition in environment even 10 years after accidental release [2].

Aim of the study is to introduce a differential method for investigating ^{14}C in environmental samples that are heterogeneous in content and in terms of their thermal degradation, estimation of sensitivity and reproducibility of the method.

Methods. When studying radioactive graphite dispersion in forest environment [2] we had used vacuum pyrolysis method [5], i.e. sample carbide was prepared in two stages: one – while pyrolysis occur all gaseous carbonaceous materials were absorbed into lithium alloy and second all resulting charred material of forest litter was melted directly with lithium in the same reaction vessel. Thus we had missed ^{14}C details of sample material

fractions. Again, coming back to the same sample's material, now (2018) we used vacuum pyrolysis method [5] considering two above mentioned components of each sample prepared separately and comparing them with data we had got earlier [2].

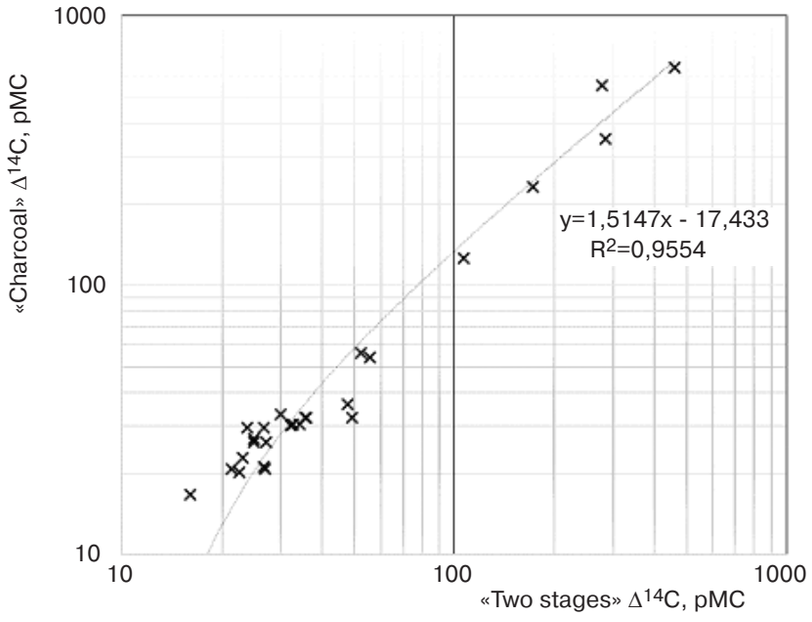
Two sub-samples were producedone by one by separate carbide formation. Now we use some residual material of about 5 g for each sample, which we still have at the moment a long time after initial work. Benzene samples were measured using of Perkin Elmer LS spectrometer Quantulus 1220™ and corresponding optimized small volume Teflon vials [6].

Results and discussion. Thus, all available data we put together in table, where, we include already published data of table [2], called as «two stages», and we enclose (in the same column) some our earlier similar data, which was not shown at [2], i.e. sampling sites: 96/13, 96/17 and 96/36. Besides of that, in table, we had enclosed separate data of two fractions, called as («pyrolysis» and («charcoal»), which were obtained now using our two separate fractions approach.

Comparing all ^{14}C data enclosed in table, which we had got on the same sample set using of different approaches of vacuum pyrolysis: total two stage and separate stages («two stages» vs «pyrolysis» and «charcoal») we find that in the same sample value of «pyrolysis» is, in most cases, lower than «charcoal». So as it was concluded in [2], most of activity associated with graphite has being preserved while sample processing by charring or vacuum pyrolysis procedure during of sample preparation. Compared data for total two stages method «two stages» and new – «charcoal» correlate well ($R^2 = 0.9554$), when new data «charcoal» are systematically 51.5% higher, see figure. This is simple to explain as «two stages» data have total ^{14}C of those two stages when «charcoal» is higher, as it is not diluted by «pyrolysis» fraction, which is lower in most cases as it was mentioned above.

Figure

Comparison of ^{14}C concentration in charred material «charred» samples and charcoal samples obtained by vacuum pyrolysis «charcoal»



Conclusions

Vacuum pyrolysis technology allows simple production of benzene sample for most kind of sample materials. It allows utilize gaseous carbon-containing material for production of carbide, which releasewhile pyrolysis.

Sample materials, which have different ^{14}C concentrations and/or different thermodestruction properties can be processed together or separate aiming joint or separate analyze of sub-materials fractions.

Use about 5 g of dry forest litter material allow us preparing two fraction samples in most cases.

Comparison of ^{14}C concentration in forest litter samples, which have being contaminated due to Chornobyl accident by radioactive graphite shows significant difference depending on kind of vacuum pyrolysis technology used: two stage total or two separate fractions. Those ^{14}C data sets, obtained for total «two stages» sample and for sample based on charcoal resulting after pyrolysis, give strong correlation, when «charcoal» are systematically 51.5% higher.

INVESTIGATION OF RADIOACTIVE GRAPHITE IN THE FOREST LITTER

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***Objective**: Our goal was to introduce the differential method for the investigation of ^{14}C in the samples of the environment that were inhomogeneous by the content of ^{14}C and by the inhomogeneity of thermo-destruction of their components. We estimated sensitivity and reproduction of the method.*
***Materials and methods:** For the investigation of ^{14}C we used traditional method on the basis of liquid scintillation count and applied vacuum pyrolysis for the component preparation of the forest litter in the search of radioactive graphite.*
***Results and conclusions:** We compared the results of the application of vacuum pyrolysis technology (Skripkin & Kovaliukh, 1997) for the production of benzene samples where the fractions of one and the same sample were processed together or separately. ^{14}C is indicated systematically more by 51.5% in the prepared separate second fraction of two ones of the samples in comparison with the joint sample of two fractions ($R^2 = 0.9554$).Systematically, there was ^{14}C more by 51.5% in the prepared separate second of two fractions of the samples in comparison with the joint sample of two fractions ($R^2 = 0.9554$).*

Keywords: ^{14}C, LSC, carbide, benzene, vacuum pyrolysis, forest litter, radioactive graphite.

ИССЛЕДОВАНИЕ РАДИОАКТИВНОГО ГРАФИТА В ЛЕСНОЙ ПОДСТИЛКЕ

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***Целью** работы являются введение дифференциального метода исследования ^{14}C в пробах окружающей среды неоднородных по содержанию ^{14}C и по неоднородности термодеструкции их компонент, оценка чувствительности и воспроизводимости метода.*

***Материалы и методы.** Использовали традиционный метод исследований ^{14}C на основе жидкостносцинтилляционного счета. Применяли вакуумный пиролиз, в частности для покомпонентной подготовки проб лесной подстилки в поисках радиоактивного графита.*
***Результаты и выводы**.*
Мы сравнили результаты применения технологии вакуумного пиролиза (Skripkin&Kovaliukh, 1997) для получения образцов бензола, где фракции одного и того же образца были обработаны вместе или по отдельности. Подготовленная, отдельная вторая из двух фракций образцов систематически дает на 51.5% больше ^{14}C по сравнению с общим образцом из двух фракций ($R^2 = 0.9554$).

Ключевые слова: ^{14}C, ЖСС, карбид, бензол, вакуумный пиролиз, лесная подстилка, радиоактивный графит.

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