Researching the possibility to use Fricke dosimeter for measurement of absorbed dose generated by pulse electron beam

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Abstract. The focus of this article is the working range of Fricke dosimeter and modified Fricke dosimeter. This study is motivated by two research questions: (1) Is it possible to use Fricke dosimeter or modified Fricke dosimeter as an instrument for dose measurements when absorbed dose is generated by pulse electron beam? (2) What limitations do these methods have? This article tells in detail about the solutions preparing and chemical analyses. It shows that modified Fricke dosimeter may be used for measurements of absorbed dose generated by pulse electron beam up to 11 kGy. And Fricke dosimeter leads to incorrect results when local dose in solution upper than 500 Gy.

1. Introduction
A successful result of every radiochemical investigation depends, first of all, on right measurement of the absorbed dose as dosimetry represents a quantitative aspect of radiation chemistry. So, every radiochemical experiment starts with dose measurements [1]. An absorbed dose in the irradiated object that corresponds to the optimal yield for this radiation-chemical process, determines its energy capacity and consequently economic efficiency of process realization in industrial construction.

More often the absorbed dose of electron radiation is evaluated by measured beam characteristics in certain operating regime of an accelerator and known characteristics of irradiated material. But accuracy of the computational method is not enough when the beam utilization factor coefficient is less than 1. To increase accuracy of the absorbed dose measurements, standardized dosimetric systems are used.

Absorbed dose measurements by experimental methods have a set of advantages especially when the dosimetric system with its characteristics (state of aggregation, effective atomic number, depth of detecting layer and other) is close to the characteristics of irradiated object.

2. Theoretical background
As a rule, in radiation chemistry for dose measurements chemical methods are used and the ferrosulfate dosimetric system (Fricke dosimeter) that has especially extensive application. It is a water solution of (1-5)·10⁻³ M soured ferrous sulfate FeSO₄·7H₂O in 0.4 M H₂SO₄ saturated by air. Ferrous iron is oxidized to ferric iron when the solution is ionized by radiation [2].

For preparing 200 ml of the Fricke dosimeter solution 4.4 ml of high-concentration sulphuric acid H₂SO₄ were used and 56 mg of iron vitriol FeSO₄·7H₂O. It provides for 56 mg/l ions of Fe²⁺ and
0.4 M H2SO4 in the solution. For preparing 200 ml of the modified Fricke dosimeter solution 0.11 ml of high-concentration sulphuric acid H2SO4 were used, 56.6 mg of iron vitriol FeSO4·7H2O and blue copperas 5 g. It provides for 56 mg/l ions of Fe2+, 0.01 М H2SO4 and 0.1 M Cu2+ in the solution.

For determining the concentration of ferrous iron the standard method is used. Using this method 1 ml of the interested solution was sampled then it was equalized to level 4.5 by adding 5 ml of acetate buffer (pH 4.5). After 1 ml of 0.1 % solution of o-phenanthroline is added and reaches 50 ml by pouring distilled water into the painted solution. 30 minutes later the test portion is exposed to colorimetry with the following parameters of photoelectric colorimeter: wavelength is 490 nm and cuvet depth – 3 cm [3].

Determining the ferric iron in the solution is carried out with the help of the standard method too. To the 1 ml of interested solution 1 ml of ammonium muriate (2 M solution), 1 ml of sulphosalicylic acid (20 % solution) and 0.1 ml of hydrochloric acid (3:2) are poured. 50 ml of the painted solution is obtained by pouring distilled water. Then the sample is exposed to colorimetry: wavelengths 540 nm and cuvet depth 3 cm [3].

Recomputation of optical density was done with the equation (1) [4-6].

\[ C = \frac{D - D_0}{\Sigma} \cdot \frac{V_{retort}}{V_{sample}} \cdot M[Fe], \]  

where \( C \) – ferrum concentration, mg/l; \( D, D_0 \) – optical density of sample and blank corresponding; \( \Sigma \) – extinction coefficient; \( V_{retort} \) – volume of painted solution, ml; \( V_{sample} \) – sample volume, ml; \( M[Fe] \) – ferrum molar weight, mg/mole.

The upper limit of dose measurements is determined by exhaustion of oxygen in the solution. Thus, for the saturated by air solution with \( G(Fe^{3+}) = 16 \text{ ion/}100 \text{ eV} \) the upper limit is 500 Gy. In this paper Fricke dosimeter was used for the dosimetry of pulse high-current electron beam, generated with the accelerator “Astra-M”, with the following parameters: accelerating potential – 450 kV, pulse energy – 1.2 J, half-amplitude duration – 150 ns [2].

The absorbed dose was calculated with the equation (2).

\[ D = \frac{cN \cdot 100}{GP}, \]  

where \( D \) – absorbed dose, Gy; \( c \) –concentration, M; \( N \) – Avogadro constant; \( G(Fe^{3+}) \) – radiation-chemical yield, ion/100 eV; \( P \) – density, kg/l.

The absorbed dose may also be computed with the equation (3), given the mass of a sample and the transferred energy [4-6].

\[ D = \frac{Q}{M}, \]  

where \( M \) – mass of sample, kg; \( Q \) – energy that was transferred to the sample, J.

The sample mass could be calculated if given its density and volume (in this paper we refer to water solution). And the energy that was transferred by an electron beam could be measured with the help of a calorimeter.

3. Experiments

3.1. Fricke dosimeter

The experiment was carried out as follows. Firstly, the cuvet with the diameter 90 mm was filled with Fricke dosimeter solution with the layer depth range of 0.5 mm, 1 mm, 1.5 mm and 2 mm. Then the cuvet was disposed in front of accelerators output window and irradiated. For every depth of Fricke dosimeter solution the series of experiments were carried out with number of pulses varying from 1 to 5. In the experiment the energy in the pulse was measured by the calorimeter and was equal to 1.2 J.
After that chemical analysis was carried out to define the concentration of ferrous and ferric iron in the solution.

![Figure 1](image1.png)

**Figure 1. Concentrations Fe\(^{2+}\) and Fe\(^{3+}\) (solution depth is 0.5 mm)**

In the figure 1 a graph of concentrations ferrous and ferric iron is shown as a function of energy transferred to solution when solution depth was 0.5 mm. Under the influence of pulse electron beam the concentration of ferrous iron decrease depends on the energy transferred to the solution. As the concentration of ferric iron increases the one of ferrous iron decreases by the same value.

Radiochemical yield \(G(\text{Fe}^{3+})\) was computed based on experiment results and equal to \((9 \pm 2) \text{ ion}/100 \text{ eV}\). When radiochemical yield is about this value, the absorbed dose may be measured up to 1000 Gy.

![Figure 2](image2.png)

**Figure 2. Absorbed dose computed with equations (2) and (3) for different depth of irradiated solution**

Absorbed doses were computed with the equations (2) and (3), using the received values of radiochemical yield \(G(\text{Fe}^{3+})\), and measured the amount of beam energy in pulse. In the figure 2 absorbed doses calculated with equations (2) and (3) are shown. This plot confirms the possibility to use Fricke dosimeter for dose measurements of pulse electron beam but to a certain extent. The
amounts of the absorbed dose for 0.5 mm solution depth become really different after the second pulse. It is connected with the conversion of ferrous iron to ferric, as was mentioned above.

3.2. Modified Fricke dosimeter
Then the experiment was carried out with modified Fricke dosimeter. It allows to compare results with literature data and to define the value of the absorbed dose in practice, which will show work limits of this dosimeter.

The experiment with modified Fricke dosimeter was carried out the same way as with Fricke dosimeter. And then by chemical methods the concentration of ferrous and ferric iron in the solution was also measured. But defining of ferric iron in this experiment, using the standard method, shows that its factor is absent in test samples: in the original sample as well as in radiation-exposed samples. It shows that for modified Fricke dosimeter this standard method is not applicable. Probably it is due to copper ions influence in the solution, because it is singular difference between modified Fricke dosimeter and Fricke dosimeter. So, in these experiments only the concentration of ferrous iron was defined.

Figure 4 shows that after the second pulse nearly all ferrous iron in Fricke dosimeter was oxidized to ferric iron. But in modified Fricke dosimeter complete consumption of ferric iron was not achieved.

![Figure 4. Concentrations Fe³⁺ for different dosimeters](image)

The figure 5 demonstrates that in stationary steady-state regime the absorbed dose, calculated by different formulas, has the same values. It allows continuing research with modified Fricke dosimeter in flowing regime in future.

![Figure 5. Absorbed dose calculated with equations (2) and (3) for modified Fricke dosimeter](image)
4. Results

By the experiment values radiation-chemical yield was calculated and for modified Fricke dosimeter it was equal 0.62 ion/100 eV. In the literature [1, 4-5] when the absorbed dose rate is $10^{10}$ Gy/s the radiation-chemical yield for modified Fricke dosimeter equals 0.58 ion/100 eV. So the estimated value is relatively equal to the value from literature [1, 4-5].

For these conditions, as the experiment shows, Fricke dosimeter can not be used. For measurements of these absorbed dose values modified Fricke dosimeters should be used. The absorbed dose was calculated with equations (2) and (3), using calculated early radiation-chemical yield for modified Fricke dosimeter and measured with the calorimeter value of pulse energy. Figure. 5 shows absorbed doses and confirms that modified Fricke dosimeter can be used for dose measurements up to 11 kGy.

Produced results show that ferrosulfate dosimetric system, laced with copper ions, may be used for measurements of the absorbed dose, generated by the pulse electron beam. As well as using the ferrosulfate dosimetric system without copper ions leads to incorrect results when the local dose in solution is higher than 500 Gy. To prevent this, radiation-chemical yield should be controlled all the time. If it varies from 8 to 16 ion/100 eV Fricke dosimeter can be used.

References

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