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Influence of γ-irradiation (^{60}Co) on the concentration and mobility of carriers in Ge and Si single crystals of n-type

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Abstract. The influence of γ-irradiation (^{60}Co) (within the dose range $1\times10^6 \le D \le 8\times10^7$ R) on the concentration and mobility of major carriers in germanium and silicon has been investigated. In the oxygen-containing samples of $n-\mathrm{Ge}\langle\mathrm{As}\rangle$ and $n-\mathrm{Si}\langle\mathrm{P}\rangle$, and in the compensated crystals of $n-\mathrm{Si}$, the mobility is shown to grow anomalously with the irradiation dose in the region of combined scattering of carriers. Proposed in this paper is the model based on accounting partial neutralization of charge of scattering centers by charge of radiation defects produced mainly around the scattering centers. This model qualitatively explains the experimental data.

Keywords: germanium, silicon, γ-irradiation, Hall effect, carrier mobility, electron concentration, combined scattering.

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1. Introduction

Authors of the monographs [1–5] and sources, cited therein, investigated the influence of γ - radiation and other nuclear radiation on generation of radiation defects (RD) in crystals of *n*-type Si and Ge (as well as in more complex semiconductor compounds) in order to establish the nature of RD, kinetics of their accumulation and annealing under different temperature conditions of environment.

It was found in [6] that in single crystals of $n-\mathrm{Si}\langle\mathrm{P}\rangle$ grown by Czochralski method with a high content of residual oxygen impurities after high temperature annealing (at T = 1200 °C for t = 2 h), the concentration of EPR-active phosphorus N_P sharply (up to ~45%) decreases with almost constant concentration of carriers n_e. According to [6], the conditions of deionization of phosphorus impurity during the lowering of temperature (up to ~20 K) for EPR-measurements were deteriorated as a result of high temperature annealing. This could be conditioned by several factors:

a) some neutralization of the positively charged ionic residues by the negatively charged vacancies diffusing to them in the process of annealing;

b) formation of the traps that can more efficiently (in comparison with ionic residues P^+) capture electrons during lowering the temperature, thereby reducing the ESR-activity of phosphorus atoms (since in order to the phosphorus atoms reveal the EPR-activity, it is required that the electrons with uncompensated spins were captured by these atoms during the temperature decrease). Dislocation loops, for example, can act in the role of such traps. The appearance (as a result of high temperature annealing of Czochralski-grown silicon crystals) of the dislocation loops is proved in [7, 8] using X-ray studies and electron microscopy methods. Capturing the electrons at low temperatures, the traps located near the ion residues will partially neutralize the charge of these residues, reducing the efficiency of electron scattering by the ions. This will lead (as in the case *a*)) to an anomalous increase in the mobility of carriers, which is observed in the experiments with heat-treated Si crystals with a high content of oxygen impurities.

If the assumptions in [6] concerning the mechanism of increasing the mobility due to the partial compensation of positively charged ionic residues by the negatively charged vacancies or their complexes is true, then any damage of the crystal lattice (which occurs, for example, under the radiation treatment of this material), that causes the appearance of point centers with opposite charges with respect to the ionic residues within the range of their Coulomb interaction, must also lead to the increase in the mobility of carriers.

The aim of this work is an attempt to validate this assertion by studying the radiation changes in the basic parameters of $n-\mathrm{Ge}\langle\mathrm{As}\rangle$ and $n-\mathrm{Si}\langle\mathrm{P}\rangle$ single crystals (concentration n_e and mobility μ of carriers) under γ - irradiation (^{60}Co) in the region of the combined scattering of carriers.

2. Results and discussion

The experiments described below were carried out with Si and Ge single crystals of *n*-type conductivity. Changes in the concentration n_e and mobility of carriers $\mu = R \times \sigma$ (R – Hall coefficient, σ – electrical conductivity), depending on the monotonous increase in the γ - irradiation dose within the range $1\times10^6 \le D \le 8\times10^7$ R were investigated. Irradiation was carried out at room temperature. Measurements of the Hall effect at temperatures of 300 and 77 K allowed to overlap the range from conditions of predominant phonon scattering to conditions characterized by significant contribution of the conduction electron scattering by impurity centers.

The Hall effect and resistivity at 300 and 77 K were measured before and after irradiation on the five cruciform samples made from the same crystal of $n-\mathrm{Ge}$ grown by the Czochralski method and weakly doped by As impurity ($\rho_{300\mathrm{K}}$ = 45.7 Ohm·cm, $n_{e,77\mathrm{K}} = 2.48\times10^{13}\,\mathrm{cm}^{-3}$). In the initial crystals the measured values of the mobility are in a good agreement with the values of the mobility, calculated within the framework of the theory of anisotropic scattering [9, 10]. The averaged (by the data for 5 samples) results of carried out experiments for two different temperatures (300 and 77 K) in the range of non-monotonic change in parameters with growth of irradiation doses are summarized in Table 1.

The dose changes of the mobility and concentration in samples of $n-\mathrm{Ge}\langle\mathrm{As}\rangle$ (obtained both for the room temperature and for the temperature of liquid nitrogen) represented in Fig. 1.

In addition to the typical decrease of the carrier mobility under the monotonous increasing of irradiation dose, the most interesting results (for both temperatures, at which these experiments were conducted) were obtained at the minimum and maximum values from the used range of doses: in the first case – some increase and in the second case – a sharp decrease in the mobility μ with increasing the irradiation dose.

Presence of maximum in $\mu = \mu(D)$ (see curves 1 and 3 in Fig. 1), probably, results from two factors:

a) growth of $\mu(D)$ associated with the radiation-induced introduction of the acceptor (negatively charged) centers that, at small doses, are mainly produced in the vicinity of the positively charged ion cores of phosphorus (where the lattice is slightly stressed) and partly neutralize charges of the cores; this effect reduces the efficiency of the Rutherford scattering and enhances the mobility μ.

b) natural decrease in $\mu(D)$ with the further increase in the radiation dose, which is due to the marked growth of the integrated concentration of scattering centers ($N_i = N_d + N_a$) and the further rise (with the dose D) of their compensation factor.

A sharp reduction of carrier mobility at maximum irradiation doses ($\approx7\times10^7$ R) is related, likely, not only with the increased number of crystal lattice regions damaged during irradiation, but also with change in their shape and, perhaps, even with appearance of mutual overlap some of them. It also leads to that the discussed changes in the mobility of irradiated crystals almost equally noticeable both at room temperature and at 77 K, since they relate with changes both of the impurity scattering and the carrier scattering by lattice vibrations.

When $n-\mathrm{Ge}$ crystals were irradiated by γ - quanta (^{60}Co, $D = 10^6$ R), the experimentally measured values of mobility (at 77 K) grew up to 34400 cm^2/V·s (curve 1 in Fig. 1). As shown by the

Table 1. Influence of small doses of γ - irradiation (^{60}Co) on the resistivity ρ, concentration of major carriers n_e and their mobility μ in single crystals of $n-\mathrm{Ge}\langle\mathrm{As}\rangle$.

Dose *D*, R	*T* = 300 K			*T* = 77 K		
	ρ, Ohm·cm	n_e, cm^{-3}	μ, cm^2/V·s	ρ, Ohm·cm	n_e, cm^{-3}	μ, cm^2/V·s
0	45.7	3.86×10^{13}	3560	7.47	2.48×10^{13}	33840
1×10^6	46.8	3.48×10^{13}	3830	7.52	2.42×10^{13}	34400
2×10^6	45.2	3.74×10^{13}	3700	7.59	2.41×10^{13}	34160
3×10^6	46.9	3.67×10^{13}	3690	7.76	2.36×10^{13}	34140

calculations, this change of mobility could be caused by decrease in the concentration of scattering centers from 2.5×10^{13} down to $1\times10^{13}\,\text{cm}^{-3}$. However, the concentration of scattering centers not only did not become lower, but in fact increased slightly. This slight decrease in the carrier concentration $\Delta n_e = (2.48 - 2.42)\times10^{13} = 6\times10^{11}\,\text{cm}^{-3}$ is associated with γ - induced generation of the corresponding number of acceptor centers in the crystal volume, which capture exactly the same number of conduction electrons.

In the case of $n-\text{Ge}$, it is hard to speak about the contribution of specific RD to mobility variations because by now there is no common opinion about the type of the generated RD including oxygen. Nevertheless, it is clear that with increasing the irradiation dose for these crystals, some changes take place there, which restrains mobility lowering in the region of liquid nitrogen temperatures. This assumption gains further support from the fact that the growth of efficiency of carrier removal from the conduction band (in the region of $D > 5\times10^6$ R; see curve 3 in Fig. 1) and, consequently, the increase of concentration of scattering centers do not affect practically on dependence $\mu_{77\text{K}} = \mu(D)$ (Fig. 1, curve 2).

Decrease the carrier mobility related with increasing the irradiation dose from 10^6 up to 7×10^7 R in the region of mainly impurity scattering (i.e., at 77 K – Fig. 1, curve 2) amounts to only 24%, while in the region of preferential scattering by lattice vibrations (i.e., at 300 K – Fig. 1, curve 1) this change of $\mu_{300\text{K}}$ (in the same range of the increasing radiation dose) reaches ~39%. Thus, with respect to γ - irradiation of $n-\text{Ge}$, the change in mobility of major carriers in the direction of its reduction in the region of mainly phonon scattering is more vulnerable than in the region of preferential scattering by impurities. This fact should be taken into account, when the semiconductor devices (which operate in the high radiation fields) are based on high-resistance crystals of $n-\text{Ge}$.

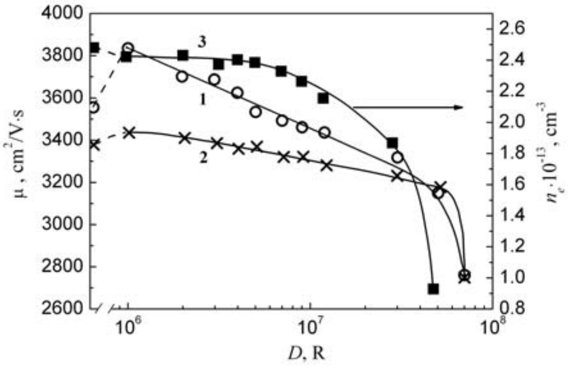

Fig. 1. Dependences of the carrier mobility μ determined at 300 K (*1*) and 77 K (*2*), and their concentration n_e (*3*) determined at 77 K on the γ - irradiation dose *D* in Czochralski-grown crystals of *n* – Ge⟨As⟩.

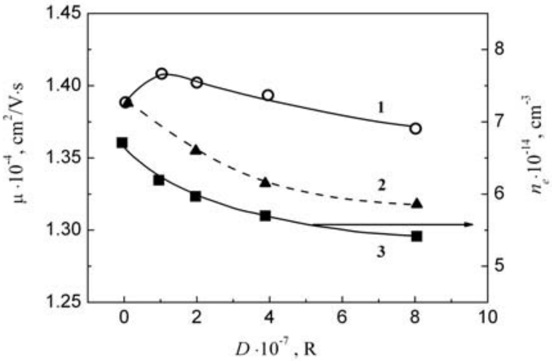

Fig. 2. Dependences of the carrier mobility μ (*1*) and their concentration n_e (*3*) determined at 77 K on the γ - irradiation dose *D* in Czochralski-grown crystals of *n* – Si⟨P⟩. The dashed curve *2* shows the mobility values calculated using the theory of anisotropic scattering.

Similar experiments on the influence of γ - irradiation on the mobility and concentration of major carriers were carried out with silicon crystals. Five samples, intended for subsequent γ - irradiation (^{60}Co), were cut from the ingot of $n-\text{Si}$, doped with phosphorus $N_\text{P} = n_e = 6.6\times10^{14}\,\text{cm}^{-3}$, grown in the ⟨100⟩ direction by the Czochralski method (with residual oxygen impurity of $(1.5-1.7)\times10^{17}\,\text{cm}^{-3}$).

For the initial samples of Si, the concentration of free carriers n_e practically corresponded to the doping level of silicon by phosphorus ($n_e = N_d$). It follows from the coincidence with the error close to ~1.5% for the experimentally determined values of μ with the data calculated according to the theory of anisotropic scattering, in which the concentration of scattering centers is taken equal to the concentration of shallow donors, i.e., for unirradiated samples ($D = 0$) can be considered that the compensation factor is $k = N_a/N_d = 0$. Therefore, using the difference between the initial (n_e^0) and final (n_e^*) values (obtained after irradiation) of carriers concentrations, it was possible to determine the concentration of acceptor centers (N_a) introduced into the crystals during irradiation, namely: $N_a = n_e^0 - n_e^*$. These and other data for $n-\text{Si}$ samples are presented in Table 2.

Fig. 2 presents the changes (obtained at 77 K) in the mobility and carrier concentration in the initial and γ - irradiated samples of $n-\text{Si}\langle\text{P}\rangle$ as a function of the dose within the range $0 \le D \le 8\times10^7$ R.

Fig. 2 shows that the carrier removal from the sample bulk (curve 3) due to radiation introduction of the acceptor centers is characterized by a function that is smoothly decreased with increasing the radiation dose *D*.

Concerning the Hall mobility of carriers, it should be seen that under the monotonical increase in the total concentration of scattering centres $N_i = N_a + N_d$, which is

Table 2. Carrier concentration n_e and concentrations of donor N_d and acceptor N_a centers (at 77 K) in the initial and γ - irradiated using the maximal dose ($D = 8\times10^7$ R) samples of n – Si⟨P⟩.

Samples of $n-\mathrm{Si}\langle\mathrm{P}\rangle$	$n_e\times10^{-14}$, cm^{-3}		$N_a\times10^{-14}$, cm^{-3}	$N_i\times10^{-14}=(N_d+N_a)\times10^{-14}$, cm^{-3}		$k=\frac{N_a}{N_d}$	k (%)
	Before irradiation $n_e\equiv N_d$	After irradiation n_e^*	After irradiation $N_a=n_e^0-n_e^*$	Before irradiation $N_d\equiv n_e$	After irradiation $N_i=N_d+N_a$		
	6.60	5.44	1.16	6.60	7.76	0.176	17.6

accompanied by a continuous decrease in n_e with increasing the irradiation dose, the mobility of carriers should decrease in the region of combined scattering (T = 77 K). However, the experimental dependence of the mobility (as in the case of γ - irradiation of $n-\mathrm{Ge}\langle\mathrm{As}\rangle$ samples) on the dose $\mu = \mu\,(D)$ has a small (but well-marked) maximum (Fig. 2, curve 1) in the range of the minimal irradiation doses (at $D = 1\times10^7$ R). Although the maximum of mobility in its dependence on the irradiation dose in the crystal of $n-\mathrm{Ge}$ (Fig. 1) appears within the range of doses that is an order of magnitude lower than those at which this maximum is observed in $n-\mathrm{Si}$ (Fig. 2).

The radiation-induced lowering the efficiency of scattering by the charged centers probably manifests itself also within the region of the highest doses where the general rise of the concentrations $N_d + N_a$ (and their compensation factor) becomes most pronounced and provides a marked lowering μ as compared with the mobility in the initial crystals. The straightforward substantiation of this interpretation is given by the numerical values of mobility which were calculated in the framework of the theory of anisotropic scattering [9–11] (see Appendix) for the summarized values of $N_i = N_a + N_d$; they appeared to be much smaller than the experimental ones (dashed curve 2 in Fig. 2). At the same time, the testing calculations, performed for the samples before their irradiation, are in a good agreement with the experimental data within the errors less than 1.5%.

The estimations showed that, at the concentrations of phosphorus atoms approximately $10^{14}-10^{15}\,\mathrm{cm}^{-3}$ already at the γ - quanta integral flows of ~10^7 R, the probability of formation of the radiation defects, within the limits of the overlapping of Coulomb cross-sections for scattering by donor and acceptor centers, amounts from 1 up to ~3%. With increasing the irradiation dose, the fraction of neutralized ion residues of phosphorus atoms increases. However, it should be taken into account that the indicated probability of introduction of the neutralized Coulomb charges (1…3%) was obtained for the equiprobable generation of radiation defects in the silicon bulk. At the same time, the probability of appearance of radiation defects in the mechanically strained regions of crystals is substantially higher than in unstrained regions. Therefore, it is naturally to expect that, at low doses of irradiation, the introduction of defects occurs primarily in the locally strained lattice regions, i.e. near the dopant atoms, and that ultimately leads to the increase of μ, which is experimentally observed.

The validity of these estimations is based on the macroscopic homogeneity of the investigated crystals. Our conclusion concerning their homogeneity follows from the fact that during the measurements of magnetoresistance in a transverse magnetic field (under $H\perp J$) the changes of the direction of the magnetic field and current J on the opposite (even at the maximum values of the field H, which amounted to 32 kOe) did not change the value of magnetoresistance within 0.05%. The above-mentioned macroscopic homogeneity of samples is not significantly deteriorated after irradiation, too (Fig. 3).

In Czochralski-grown silicon, the oxygen-vacancy complex (A – center) is the predominant radiation defect responsible for the radiation-induced changes of free carrier concentration. Naturally, one may suppose that the mobility μ is changed in the irradiated crystals just owing to the localization of these negatively charged acceptors near the phosphorus cores. One may expect that in zone-melted silicon, where the vacancy-phosphorus complex (E – center) [12] is a dominant RD, irradiation would not give rise to a mobility increase. The latter suggestion follows from the fact that the phosphorus atom is a component of E – center, thus, during the introduction of E – centers only the sign of charge of scattering centers is changed, but their concentration is not changed.

In order to verify the supposition concerning the mechanism of carrier mobility enhancement in γ - irradiated oxygen-containing crystals of silicon, the comparative experiments with samples cut from the single crystals grown using the floating-zone method, where the concentration of residual oxygen was low ($N_\mathrm{O} \le 10^{16}\,\mathrm{cm}^{-3}$), were carried out. These experiments showed that such crystals did not exhibit the radiation-induced mobility enhancement (Fig. 4), and the observed decrease of μ was caused by additional scattering by RD of other types, such as W – , K – centers, etc.

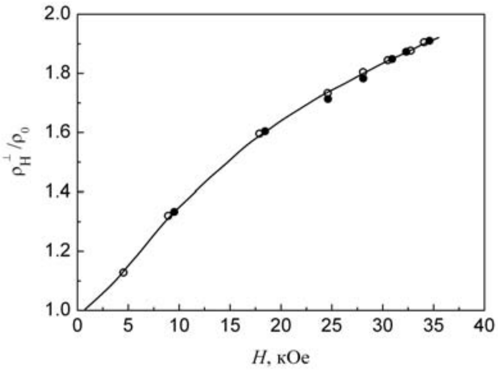

Fig. 3. Dependences $\frac{\rho_H^\perp}{\rho_0} = f(H)$ obtained at 77 K in the experiments with initial (D = 0) crystals of $n-\mathrm{Si}\langle\mathrm{P}\rangle$ (o) and with the samples γ - irradiated using the dose D = 8×10^7 R (●).

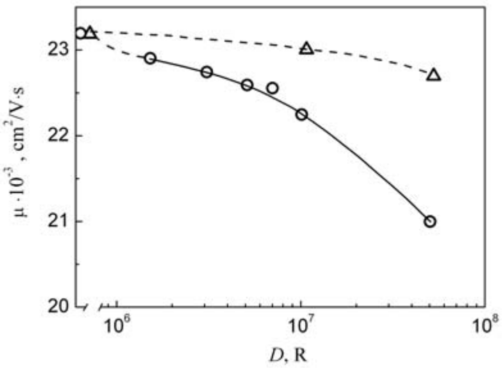

Fig. 4. Dependences of the free electron mobility μ at 77 K on the γ - irradiation dose D in the silicon crystals grown using the floating-zone method. The solid line gives our experimental data; the dashed one presents the calculated results.

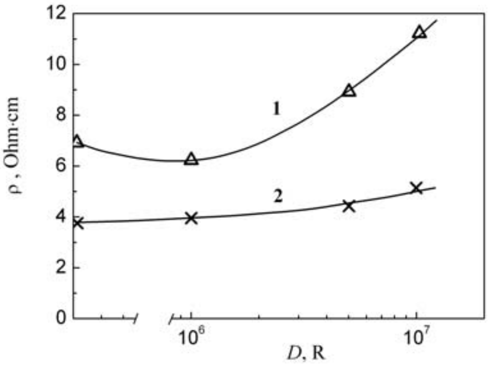

Fig. 5. Dependences of the resistivity ρ on the γ - irradiation dose D measured at 77 K in the samples of $n-\mathrm{Si}$ with different compensation factors: *1* – n_e = $4.73\times10^{13}\,\mathrm{cm}^{-3}$; $k = N_a/N_d$ = 0.83; *2* – n_e = $7.19\times10^{13}\,\mathrm{cm}^{-3}$; $k \approx 0$. Both crystals were γ - irradiated (^{60}Co) at T = 300 K.

The influence of small doses on the resistivity of compensated and of practically uncompensated (in the initial state) crystals of $n-\mathrm{Si}$ was also different, which is illustrated by the data shown in Fig. 5.

Thus, the minimum of resistivity appears in the compensated crystal at the radiation dose $D \cong 10^6$ R, while in the crystal with $k \approx 0$ such minimum does not exist. Since the compensated crystal is quite heterogeneous, in this material there is a high probability of the formation of clusters of dopant impurity in the form of closely spaced positively charged ionic residues of phosphorus (two or more). When such crystal was irradiated by γ - quanta, mobile vacancies may be trapped by one of the ionic residues of phosphorus, which is contained in the cluster structure, with the formation of negatively charged E – center. Scattering efficiency of such a new formation (which is consisted of the located near ionic residue of phosphorus P^+ and E – center) due to the effect of partial screening (or neutralization) will be somewhat reduced. So, we would obtain reduction in the resistivity (and, consequently, respective increase in the carrier mobility) at low γ - irradiation doses in the compensated crystal.

The inhomogeneity is significantly smaller in almost uncompensated material. Therefore, the formation of dopant impurity clusters is unlikely. The γ - quanta irradiation of these crystals introduces additional scattering centers (acceptors), which reduces the mobility, and, consequently, the resistivity is increased, and it is obtained by us in the experiment.

3. Conclusions

The dependences of concentration and mobility of carriers in *n*-type crystals of germanium and silicon on the dose of γ - irradiation (^{60}Co) were investigated. It is shown that under the influence of small doses of γ - irradiation in oxygen-containing single crystals of $n-\mathrm{Ge}\langle\mathrm{As}\rangle$ and $n-\mathrm{Si}\langle\mathrm{P}\rangle$, as well as in $n-\mathrm{Si}$ compensated crystals, increase of the carrier mobility within the range of combined scattering of carriers was experimentally observed. The proposed model that takes into account partial neutralization of the charge of scattering centers by the charge of radiation defects explains the observed features.

Appendix

$$\mu = \frac{3.51\cdot10^7}{T^{3/2}}\,\frac{6.95J_4' + 2J_3'}{J_3' + 13.9J_2'}, \ \mathrm{cm^2/V\cdot s}, \qquad (A1)$$

where

$$J'_1 = \int_0^\infty \frac{x^{3/2}\, e^{-x}\, \mathrm{d}x}{x^2 + 0.662\,\varphi + b_0},$$

$$J'_2 = \int_0^\infty \frac{x^{3/2}\, e^{-x}\, \mathrm{d}x}{x^2 + x^{3/2}\,\varphi + b_1},$$

$$J'_3 = \int_0^\infty \frac{x^{9/2}\, e^{-x}\, \mathrm{d}x}{\left(x^2 + 0.662\, x^{3/2} + b_0\right)\left(x^2 + x^{3/2}\,\varphi + b_1\right)},$$

$$J'_4 = \int_0^\infty \frac{x^{9/2}\, e^{-x}\, \mathrm{d}x}{\left(x^2 + x^{3/2}\,\varphi + b_1\right)^2}, \qquad \text{(A2)}$$

$$\varphi(x, T) = \frac{28.5}{e^{190/T} - 1} \times$$

$$\times \left[\sqrt{x - \frac{190}{T}} + e^{190/T} \sqrt{x - \frac{190}{T}}\; \theta\!\left(x, \frac{190}{T}\right)\right] +$$

$$+ \frac{1260}{e^{630/T} - 1} \left[\sqrt{x + \frac{630}{T}} + e^{630/T}\, \theta\!\left(x, \frac{630}{T}\right) \sqrt{x - \frac{630}{T}}\right] \qquad \text{(A3)}$$

with

$$\theta(x, \alpha) = \begin{cases} 1 & \text{for} \quad x > \alpha, \\ 0 & \text{for} \quad x < \alpha. \end{cases}$$

The values of b_0 and b_1 depending on concentration and temperature are given in detail in [11].

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