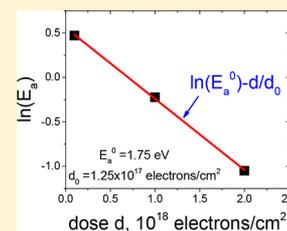


Anomalous Kinetics of Diffusion-Controlled Defect Annealing in Irradiated Ionic Solids

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ABSTRACT: The annealing kinetics of the primary electronic *F*-type color centers (oxygen vacancies with trapped one or two electrons) is analyzed for three ionic materials (Al_2O_3 , MgO , and MgF_2) exposed to intensive irradiation by electrons, neutrons, and heavy swift ions. Phenomenological theory of diffusion-controlled recombination of the *F*-type centers with much more mobile interstitial ions (complementary hole centers) allows us to extract from experimental data the migration energy of interstitials and pre-exponential factor of diffusion. The obtained migration energies are compared with available first-principles calculations. It is demonstrated that with the increase of radiation fluence both the migration energy and pre-exponent are decreasing in all three materials, irrespective of the type of irradiation. Their correlation satisfies the Meyer–Neldel rule observed earlier in glasses, liquids, and disordered materials. The origin of this effect is discussed. This study demonstrates that in the quantitative analysis of the radiation damage of real materials the dependence of the defect migration parameters on the radiation fluence plays an important role and cannot be neglected.



INTRODUCTION

Irradiation resistance is important material property, vital for many applications, including nuclear fuels,¹ reactor materials, and nuclear waste storage.^{2,3} MgO , $\alpha\text{-Al}_2\text{O}_3$ (sapphire, corundum), and MgF_2 are three wide gap insulating materials with different crystalline structures and chemical bonding. All three materials are radiation resistant and have many important applications, e.g., sapphire, is a promising material for fusion reactors, mainly for diagnostics as a general insulator or optical components,⁴ whereas optical lenses and devices from MgF_2 are transparent in an extremely wide range of photon energies, from IR to UV.⁵ To control radiation stability, it is very important to predict/simulate the kinetics of defect accumulation and thus radiation damage in these materials as well as long-time defect structure evolution.^{6–11}

There were numerous experimental measurements of the primary defect accumulation kinetics (first of all, *F*- and *F*⁺ color centers, oxygen vacancy with two and one trapped electrons, respectively) as a function of radiation dose rate and temperature with subsequent postirradiation annealing.^{12–14} Defect kinetics in solids are controlled by defect diffusion. Unlike liquids, where diffusion coefficients of particles and impurities are similar,^{15,16} in solids diffusion coefficients of different defects could vary by many orders of magnitude.^{6,13} As is well-known, the *F* center mobility is much smaller than that of the complementary Frenkel defects, interstitial oxygen ions.^{17,18} Thus, at moderate radiation fluences and temperatures, the kinetics of the *F*-type center annealing is governed by their diffusion-controlled recombination with mobile oxygen interstitials. Despite numerous experimental data on defect kinetics, very few theoretical efforts were devoted so far to the quantitative analysis of available data, in order to extract main

kinetic parameters, interstitial migration energy E_a and diffusion pre-exponent D_0 , necessary for further prediction of the secondary defect kinetics and radiation stability of sapphire and related materials.

Recently, a simple phenomenological theory of diffusion-controlled bimolecular recombination of Frenkel defects was developed and applied to irradiated ionic solids.^{19,20} We developed the alternative approach based on the formalism of joint correlation functions^{6,7} of spatial distribution of similar (*F* – *F* centers) and dissimilar (Frenkel pair of defects: the *F* center – interstitial *O_i* ions) which is well suited for the study of radiation defect kinetics and aggregation.

The atomistic model of radiation damage takes into account the following steps: (i) Frenkel defect production (*F* center, interstitial). (ii) Defect migration with the diffusion coefficient determined by the activation energy E_a and pre-exponent D_0 . (iii) Dissimilar defect recombination upon mutual approach within the critical radius *R*. (iv) Postirradiation annealing with linear increase of temperature.

One could estimate the diffusion coefficients of defects in solids, from measurements of the defect concentration changes (by optical absorption and ESR) under different conditions, e.g., sample heating (annealing) which stimulates defect reactions and recombination. The reaction rate *K* is related to a mutual diffusion coefficient *D* via simple relation $K = 4\pi DR$,²¹ where $D = D_0 \exp(-E_a/k_B T)$ and *R* recombination radius. The nonisothermal kinetics (heating)^{19,20} is considered in this paper.

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As it was shown therein, the kinetics of bimolecular recombination of the primary radiation defects, F type centers and interstitial ions, is controlled by the two parameters: the activation energy E_a for migration (diffusion) of more mobile component (interstitial) and the pre-exponential factor $A = N_0RD_0/\beta$, where N_0 is initial defect concentration and β heating rate.

Assuming standard parameters $N_0 = 10^{17} \text{ cm}^{-3}$, $R = 10 \text{ \AA}$, $D_0 = 10^{-3} \text{ cm}^2 \text{ s}^{-1}$ (typical estimate for solid state diffusion), $\beta = 10 \text{ K/min}$, one gets the estimate $A = 10^8 \text{ K}^{-1}$ (normal solid state diffusion). First results of this theory were discussed for the neutron irradiated sapphire.¹⁹ The analysis has demonstrated that the diffusion energy of oxygen interstitials varied considerably from one experiment to another, very likely due to different radiation fluences. To led more light to the problem, in this paper we analyze available experimental data of F -type center annealing in three above-mentioned materials, Al_2O_3 , MgO , and MgF_2 irradiated by electrons, neutrons, and heavy ions with very different fluences (and respectively, defect concentrations). As mentioned, these three materials represent a wide class of radiation resistant solids and have quite different crystalline structure and chemical bonding: MgO is cubic and ionic, MgF_2 has rutile structure and ionic, and sapphire ($\alpha\text{-Al}_2\text{O}_3$) has hexagonal structure and partly covalent.

RESULTS

We demonstrate in this paper that the first recent observation for neutron irradiated sapphire¹⁹ could be a quite general phenomenon: diffusion of interstitial ions in many heavily irradiated ionic materials depends strongly on the radiation fluence and is characterized by unusually high mobility and low migration barriers.

The experimental kinetics of the F - and F^+ center annealing in neutron irradiated sapphire obtained in several studies and their theoretical analysis described in refs 19 and 20 are presented in Figure 1 and Table 1. One can see a trend in

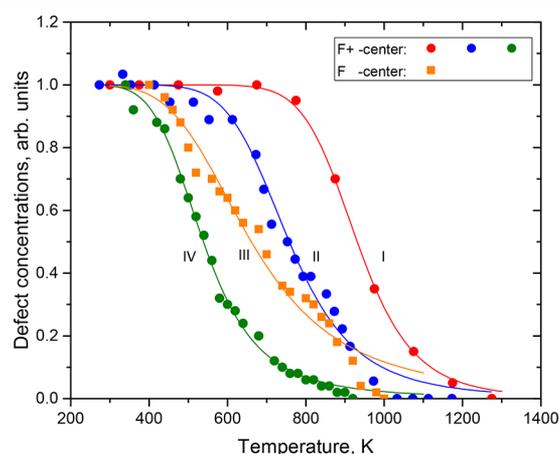


Figure 1. Kinetics of the F or F^+ center annealing in Al_2O_3 (see Table 1 for details, only four representative kinetics are plotted here for illustration).

decrease of both the migration energy and pre-exponential A with fluence (defect concentration). Note that the F - and F^+ centers show very similar annealing energies. The migration energy of 0.8–0.9 eV obtained at low fluences is close to the calculated activation energy for charged oxygen interstitials²² (to be published). The relevant prefactor is the largest and

Table 1. Explanation of Curves I–IV in Figure 1 and the Obtained Migration Energy of Interstitial Ions E_a and Pre-Exponential Factor A in Sapphire under Different Conditions of Radiation and Different Doses (Nos. 1–10)^a

no.	type	E_a (eV)	A (K^{-1})	legend
1 (I)	F^+	0.89	7.0×10^1	neutron fluence $2 \times 10^{17} \text{ n/cm}^2$, ref 36
2	F	0.79	2.1×10^1	same as 1, ref 36
3 (II)	F^+	0.47	1.2×10^0	fast neutron ($E_n > 1.2 \text{ MeV}$), fluence $9.1 \times 10^{17} \text{ n/cm}^2$, ref 37
4	F^+	0.40	2.3×10^{-1}	fluences from 5×10^{18} to $2 \times 10^{21} \text{ n/cm}^2$, ref 38
5	F^+	0.39	5.3×10^{-1}	fast neutron, fluence $> 10^{17} \text{ n/cm}^2$, ref 39
6	F^+	0.35	1.4×10^0	isochronal thermal anneal of F band in Al_2O_3 : fission-spectrum neutrons, ref 40
7 (IV)	F^+	0.27	4.0×10^{-1}	14-MeV and fission-neutrons, fluence of 10^{17} n/cm^2 , ref 41
8 (III)	F	0.22	3.3×10^{-2}	same as 7, ref 41
9	F	0.17	1.3×10^{-2}	fast neutron, fluence $4 \times 10^{16} \text{ n/cm}^2$, ref 39
10	F	0.14	1.9×10^{-3}	fast neutron ($E_n > 1.2 \text{ MeV}$), fluence $4 \times 10^{16} \text{ n/cm}^2$, ref 42

^aThe information is ordered with respect to the activation energy increase.

close to the above-mentioned theoretical limit for a perfect solid.

Similar data for MgO crystals irradiated with electrons, neutrons, and heavy swift ions are presented in Figure 2 and

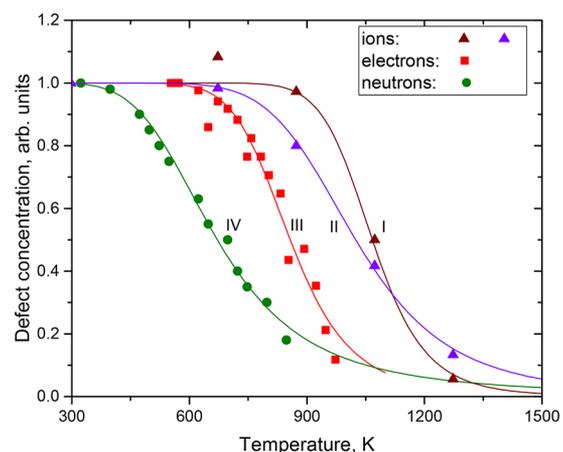


Figure 2. Kinetics of the F^+ center annealing in MgO for different types of radiation (see Table 2 for details).

Table 2. Table 2 shows the same trend in the decrease of the migration energy and prefactor A with the fluence, irrespective of the type of radiation. The largest migration energy of 1.7 eV is close to theoretical calculations (1.5 eV, ref 23).

Lastly, Figure 3 and Table 3 present similar results for MgF_2 crystals. The annealing kinetics for the F centers in MgF_2 monitored by means of the two different methods, optical absorption and EPR, under electron irradiation and neutrons²⁴ is presented in Figure 3, respectively. All data agree very well each with other, indicating that the effect is not restricted by a particular irradiation type. This is also not a result of the increase of initial defect concentration with dose (with longer irradiation or large local concentration in narrow incident

Table 2. Explanation of Curves I–V in Figure 2 and the Obtained Migration Energy of Interstitial Ions E_a and Pre-Exponential Factor A in MgO under Different Types of Radiation and Different Doses (nos. 1–12)

no.	type	E_a (eV)	A (K^{-1})	legend
1	ion	1.74	1.4×10^5	fluence 5×10^{14} Kr^+ /cm ² with energy 150 keV, ref 43
2 (I)	ion	1.28	1.3×10^3	fluence 1×10^{15} Kr^+ /cm ² , ref 43
3	ion	0.86	5.4×10^0	fluence 1×10^{15} Ar^+ /cm ² with energy 100 keV, ref 44
4	electron	0.71	1.5×10^2	1.7 MeV, F centers 1.6×10^{18} cm ⁻³ , ref 45
5 (III)	electron	0.68	1.1×10^1	optical absorption, 1.8 MeV, fluence similar to 8×10^{15} n/cm ² , ref 47
6	electron	0.65	1.5×10^2	optical absorption, 3 MeV, F centers 5×10^{18} cm ⁻³ , ref 46
7 (II)	ion	0.60	6.0×10^{-1}	fluence 5×10^{15} Kr^+ /cm ² with energy 150 keV, ref 43
8	neutron	0.54	1.0×10^1	fluence 1×10^{17} n/cm ² , ref 48
9	ion	0.41	1.7×10^{-2}	same as 3, ref 44
10	ion	0.39	2.2×10^{-2}	same as 3, ref 44
11	Ne ions	0.24	2.5×10^{-3}	fluence 1×10^{15} Ne/cm ² , ref 49
12 (IV)	neutron	0.23	3.6×10^{-2}	fluence 6×10^{17} n/cm ² with energy >1 MeV, ref 45

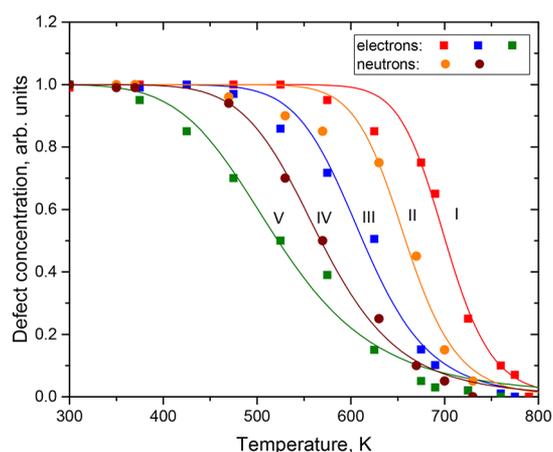


Figure 3. Kinetics of the F^+ center annealing in MgF_2 for different types of radiation (see Table 3 for details).

particle tracks): this would lead to an increase of N_0 and thus parameter A , which, in fact, decreases with dose by orders of magnitude.

As was mentioned above, in a crystalline structure the prefactor is expected to be of the order of $A = 10^8$ – 10^9 K^{-1} . This is indeed close to values obtained for the lowest radiation doses (curve I in Figure 3). The relevant migration energy of 1.6 eV corresponds to the interstitials in almost perfect MgF_2 crystals. However, there are strong arguments^{25,26} that indicates that fluoride interstitials in MgF_2 form neutral immobile F_2 molecules and the observed energy could be related not entirely to the migration of a single interstitial but includes also the energy necessary to break a bond and to release it.

Measurements 2, 4, and 7 (Table 3) performed on the same sample allow us to estimate dependence of the migration energy on the fluence. The insert in the graphical TOC entry shows the exponential dependence $E_a(d) = E_a(0) \exp(-d/d_0)$, where d is dose. The fitting gives $E_a(0) = 1.75$ eV and $d_0 = 1.25 \times 10^{17}$ e/cm². This energy is close to our estimate above for the

Table 3. Explanation of Curves I–V in Figure 3 and the Obtained Migration Energy of Interstitial Ions E_a and Pre-Exponential Factor A in MgF_2 under Different Types of Radiation and Different Doses (nos. 1–7)

no.	type	E_a (eV)	A (K^{-1})	legend
1	electron	1.87	4.8×10^{10}	optical absorption, 2 MeV electron irradiation, dose 1×10^{16} e/cm ² , ref 24
2 (I)	electron	1.60	1.0×10^8	EPR, 2 MeV electron irradiation, dose 1×10^{16} e/cm ² , ref 24
3 (II)	neutron	1.24	9.8×10^6	EPR, $E > 1$ MeV, irradiation 15 min, flux 3×10^{13} n/cm ² s, ref 24
4 (III)	electron	0.80	8.4×10^3	EPR, 2 MeV electron irradiation, dose 1×10^{17} e/cm ² , ref 24
5 (IV)	neutron	0.56	1.8×10^2	EPR, $E > 1$ MeV, irradiation 10 h, flux 3×10^{13} n/cm ² s, ref 24
6	ions	0.46	9.6×10^0	optical absorption, U ions (11.1 MeV/u), fluence 6×10^{11} cm ⁻² , ref 50
7	electron	0.35	3.5×10^0	EPR, 2 MeV electron irradiation, dose 2×10^{17} e/cm ² , ref 24

limit of low doses, as well as theoretical calculations.^{14,23} As the fluence increases, the migration energy strives asymptotically to zero.

The above observed correlation of the migration energy E_a and prefactor A (Tables 1–3) fits quite well to the relation known as the Meyer–Neldel rule:^{28,29}

$$\ln(A) = \ln(A_0) + E_a/k_B T_0 \quad (1)$$

where A_0 is a constant and T_0 some characteristic temperature. Equation 1 shows how reduction of the activation energy with growing disorder is compensated by orders of magnitude decrease of the prefactor A . Figure 4 demonstrates that this relation indeed is well satisfied for all three materials and, more importantly, for different types of irradiations (and initial defect spatial distributions). Note that all experimental points lie below the characteristic temperature T_0 . Equation 1 could be also interpreted as the diffusion coefficient with exponentially dependent pre-exponent

$$D \propto \exp(E_a/k_B T_0 - E_a/k_B T), \quad T < T_0 \quad (2)$$

The decrease of both the migration energy and pre-exponential A with radiation fluence (defect concentration) is very well documented for the MgF_2 case discussed above where experiments were performed on the same samples. In the case of sapphire this effects is partly hidden by use of different samples with different history, pre-existing defects, and irradiation by neutrons of different energies and at different temperatures. It would be of great interest to perform focused experiments on the same sapphire samples varying only the neutron fluences, similarly as it was performed for MgF_2 .²⁴

DISCUSSION AND CONCLUSIONS

Summing up, it is demonstrated, for the first time, that in three types of strongly irradiated ionic solids of different crystalline structure and chemical nature, the pre-exponential factor of diffusion is strongly correlated with the migration energy. In other words, the defect recombination kinetics is not characterized uniquely by the activation energy for diffusion with a constant pre-exponent (as generally accepted in solid state physics and chemistry) but instead these parameters depend on the radiation fluence which considerably compli-

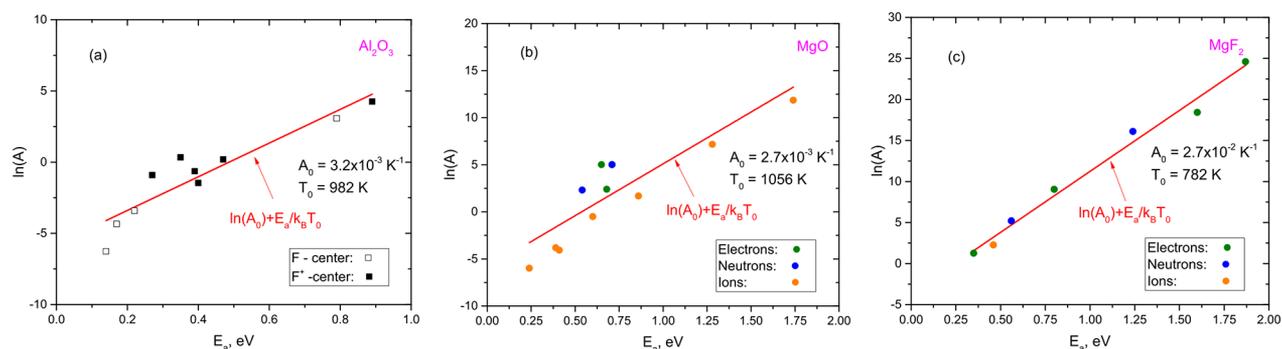


Figure 4. Correlation of the effective energies and pre-exponents for sapphire (a), MgO (b), and MgF₂ (c). The high quality of this correlation is characterized by the standard Pearson correlation coefficient: (a) $r = 0.92819$, (b) $r = 0.87847$, and (c) $r = 0.99291$, very close to the perfect case ($r = 1$). In all three cases the positive linear relationship takes place between the two variables.

ates analysis of the radiation-induced kinetic processes. Note that this correlation ($E_a - A$) effect, known as the Meyer–Neldel rule,²⁸ is well-known in chemistry, biology, even semiconductor physics.^{29–32} The foundations of this empirical rule are still not fully understood. One of possible phenomenological models³³ claims that eq 1 holds in disordered systems with exponential probability distribution of energy barriers of localized quasi-particles. The relation 1 is observed usually in multicomponent systems (for example, metallurgical slags³⁴ containing tens of oxides). Our observation shows that this is possible in simple systems and without changes of composition. Similar observations were also done in glasses under weak neutron or ion irradiation characterized by small variation of activation energies: chalcogenide glass³⁵ and metallic glass.³⁰

One possible logical explanation of the observed effect could be a growing disordering of materials under irradiation with a continuous transition from a perfect crystalline structure to the amorphous-like one.²⁷ However, three studied materials do not reveal considerable amorphization and strong disordering under irradiation.⁵¹ Moreover, experiments⁵² show that oxygen diffusion in vitreous silica (and probably, many other disorder solids) is slower than in the crystalline phase, whereas computer modeling of a series of binary oxides⁵¹ indicated that activation barriers for radiation damage recovery increases with the material disordering. Another explanation could be related with the simulations of the radiation damage of MgO⁵³ which suggested extremely high mobility of small interstitial clusters (and thus, average interstitial mobility). A number of such clusters could increase with the fluence growth and explain our observation. More detailed experimental and theoretical studies for a broader class of materials are necessary for understanding the observed effect.

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Notes

The authors declare no competing financial interest.

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