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 analysed 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, α-Al_2O_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 post-irradiation 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.

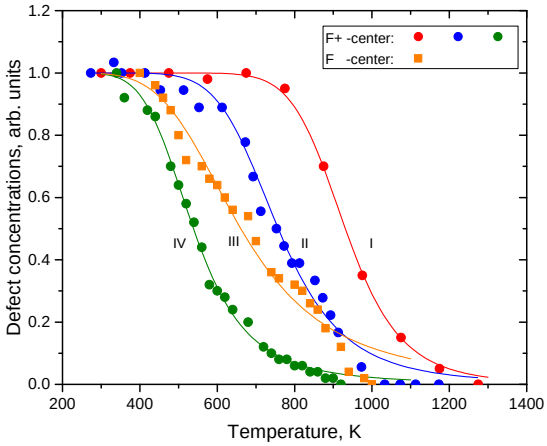

Figure 1: The kinetics of the F or F^+ center annealing in Al_2O_3 (see Table 1 for details, only four representative kinetics are plotted here for illustration).

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 Oi 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) Post-irradiation 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 $D = D_0 \exp(-E_a/k_BT)$ and R recombination radius. The non-isothermal kinetics (heating)[19,20] is considered in this paper.

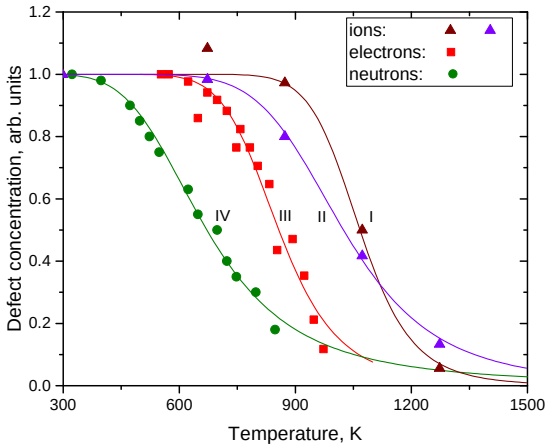

Figure 2: The kinetics of the F^+ center annealing in MgO for different types of radiation (see Table 2 for details).

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 $N_0 = 10^{17}\text{cm}^{-3}$, $R = 10\mathring{A}$, $D_0 = 10^{-3}$ cm^2 s^{-1} (typical estimate for solid state diffusion), $\beta = 10$ K/min, one gets the estimate $A = 10^8$ 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, sapphire (α-Al_2O_3) has hexagonal structure and partly covalent.

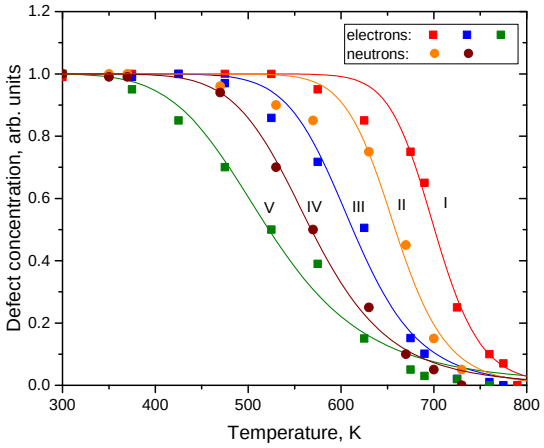

Figure 3: The kinetics of the F^+ center annealing in MgF_2 for different types of radiation (see Table 3 for details).

Results

We demonstrate in this paper that the first recent observation for neutron irradiated sapphire[19] 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.[19,20] are presented in Fig.1 and Table 1. One can see a trend in 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 interstitials[22] (to be published). The relevant pre-factor is the largest and close to above-mentioned theoretical limit for a perfect solid.

Similar data for MgO crystals irradiated with electrons, neutrons and heavy swift ions are presented in Fig.2 and Table 2. Table 2 shows the same trend in the decrease of the migration energy and pre-factor A with the fluence, irrespective the type of radiation. The largest migration energy of 1.7 eV is close to theoretical calculations (1.5 eV, ref.[23]).

Lastly, Fig.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 neutrons[24] is presented in Fig. 3, respectively. All data agree very well each with other, indicating that the effect is not restricuted by a particular irradiation type. This is also not result of the increase of initial defect concentration with dose (with longer irradiation or large local concentration in narrow incident particle tracks): this would lead to 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 pre-factor 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 Fig.3). The relevant migration energy of 1.6 eV corresponds to the intersti-

Table 1: The explanation of curves I-IV in Fig.1 and the obtained migration energy of interstitial ions E_a and pre-exponential factor A in sapphire under different condition of radiation and different doses (Nr.1-10). The information is ordered with respect to the activation energy increase

Nr.	Type	E_a (eV)	A (K^{-1})	Legend
1 (I)	F^+	0.89	$7.0 \cdot 10^1$	Neutron fluence $2 \cdot 10^{17}$ n/cm^2, Ref.[36]
2	F	0.79	$2.1 \cdot 10^1$	Same as 1, Ref.[36]
3 (II)	F^+	0.47	$1.2 \cdot 10^0$	Fast neutron (En > 1.2 MeV), fluence $9.1 \cdot 10^{17}$ n/cm^2, Ref.[37]
4	F^+	0.40	$2.3 \cdot 10^{-1}$	Fluences from $5 \cdot 10^{18}$ to $2 \cdot 10^{21}$ n/cm^2, Ref.[38]
5	F^+	0.39	$5.3 \cdot 10^{-1}$	Fast neutron, fluence > 10^{17} n/cm^2, Ref.[39]
6	F^+	0.35	$1.4 \cdot 10^0$	Isochronal thermal anneal of F band in Al_2O_3: fission-spectrum neutrons, Ref.[40]
7 (IV)	F^+	0.27	$4.0 \cdot 10^{-1}$	14-MeV and fission-neutrons, fluence of 10^{17} n/cm^2, Ref.[41]
8 (III)	F	0.22	$3.3 \cdot 10^{-2}$	Same as 7, Ref.[41]
9	F	0.17	$1.3 \cdot 10^{-2}$	Fast neutron, fluence $4 \cdot 10^{16}$ n/cm^2, Ref.[39]
10	F	0.14	$1.9 \cdot 10^{-3}$	Fast neutron (En > 1.2 MeV), fluence $4 \cdot 10^{16}$ n/cm^2, Ref.[42]

tials 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 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, 7 (Table 3) performed on the same sample allow us to estimate dependence of the migration energy on the fluence. The insert in Graphical TOC Entry shows the exponential dependence $E_a(d) = E_a(0)\exp(-d/d_0)$, where d is dose. The fitting gives $Ea(0) = 1.75$ eV and $d_0 = 1.25 \cdot 10^{17}$ e/cm^2. This energy is close to our estimate above for the 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 pre-factor A (Tables 1-3) fits quite well to the relation known as the Meyer-Neldel rule:[28,29]

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

where A_0 is a constant and T_0 some characteristic temperature. Eq.(1) shows how reduction of the activation energy with growing disorder is compensated by orders of magnitude decrease of the pre-factor A. Fig.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. Eq.(1) could be also interpreted as the diffusion coefficient with exponentially dependent pre-exponent

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

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

Table 2: The explanation of curves I-V in Fig.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 (Nr.1-12)

Nr.	Type	E_a (eV)	A (K^{-1})	Legend
1	ion	1.74	$1.4 \cdot 10^5$	Fluence $5 \cdot 10^{14}$ Kr^+/cm^2 with energy 150 keV, Ref.[43]
2 (I)	ion	1.28	$1.3 \cdot 10^3$	Fluence $1 \cdot 10^{15}$ Kr^+/cm^2, Ref.[43]
3	ion	0.86	$5.4 \cdot 10^0$	Fluence $1 \cdot 10^{15}$ Ar^+/cm^2 with energy 100 keV, Ref.[44]
4	electron	0.71	$1.5 \cdot 10^2$	1.7 MeV, F centers $1.6 \cdot 10^{18}$ cm^{-3}, Ref.[45]
5 (III)	electron	0.68	$1.1 \cdot 10^1$	Optical absorption, 1.8 MeV, fluence similar to $8 \cdot 10^{19}$ n/cm^2, Ref.[47]
6	electron	0.65	$1.5 \cdot 10^2$	Optical absorption, 3 MeV, F centers $5 \cdot 10^{18}$ cm^{-3}, Ref.[46]
7 (II)	ion	0.60	$6.0 \cdot 10^{-1}$	Fluence $5 \cdot 10^{15}$ Kr^+/cm^2 with energy 150 keV, Ref.[43]
8	neutron	0.54	$1.0 \cdot 10^1$	Fluence $1 \cdot 10^{17}$ n/cm^2, Ref.[48]
9	ion	0.41	$1.7 \cdot 10^{-2}$	Same as 3, Ref.[44]
10	ion	0.39	$2.2 \cdot 10^{-2}$	Same as 3, Ref.[44]
11	Ne ions	0.24	$2.5 \cdot 10^{-3}$	Fluence $1 \cdot 10^{15}$ Ne/cm^2, Ref.[49]
12 (IV)	neutron	0.23	$3.6 \cdot 10^{-2}$	Fluence $6 \cdot 10^{17}$ n/cm^2 with energy > 1 MeV, Ref.[45]

Table 3: The explanation of curves I-V in Fig.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 (Nr.1-7)

Nr.	Type	E_a (eV)	A (K^{-1})	Legend
1	electron	1.87	$4.8 \cdot 10^{10}$	Optical absorption, 2 MeV electron irradiation, dose $1 \cdot 10^{16}$ e/cm^2, Ref.[24]
2 (I)	electron	1.60	$1.0 \cdot 10^8$	EPR, 2 MeV electron irradiation, dose $1 \cdot 10^{16}$ e/cm^2, Ref.[24]
3 (II)	neutron	1.24	$9.8 \cdot 10^6$	EPR, E $>$ 1 MeV, irradiation 15 min, flux $3 \cdot 10^{13} n/cm^2 s$, Ref.[24]
4 (III)	electron	0.80	$8.4 \cdot 10^3$	EPR, 2 MeV electron irradiation, dose $1 \cdot 10^{17}$ e/cm^2, Ref.[24]
5 (IV)	neutron	0.56	$1.8 \cdot 10^2$	EPR, E $>$ 1 MeV, irradiation 10 h, flux $3 \cdot 10^{13} n/cm^2 s$, Ref.[24]
6	ions	0.46	$9.6 \cdot 10^0$	Optical absorption, U ions (11.1 MeV/u), fluence $6 \cdot 10^{11}$ cm^{-2}, Ref.[50]
7	electron	0.35	$3.5 \cdot 10^0$	EPR, 2 MeV electron irradiation, dose $2 \cdot 10^{17}$ e/cm^2, Ref.[24]

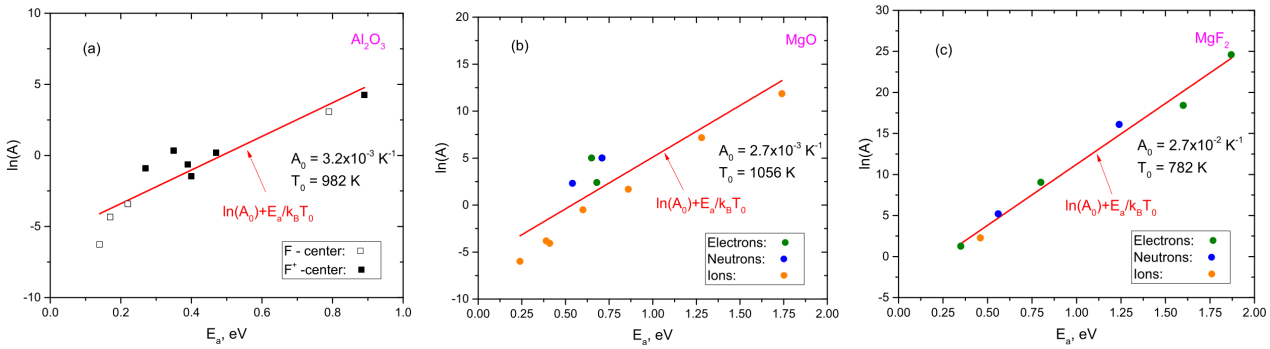

Figure 4: Correlation of the effective energies and pre-exponents for sapphire (a), MgO (b) and MgF_2 (c). The high quality of this correlation is characterized by the standard Pearson correlation coefficient: (a) $r = 0.92819$, (b) $r = 0.87847$ and (c) $r = 0.99291$, very close to the perfect case ($r = 1$). In all three cases the positive linear relationship takes place between the two variables.

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 complicates 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 semi-conductor physics.[29–32] The foundations of this empirical rule are still not fully understood. One of possible phenomenological models[33] 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 slags[34] 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 glass[35] and metallic glass.[30]

One of possible logical explanations 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, experiments[52] show that oxygen diffusion in vitrous silica (and probably, many other disorder solids) is slower than in the crystalline phase, whereas computer modeling of a series of binary oxides[51] indicated that activation barriers for radiation damage recovery increses with the material disordering. Another explanation could be related with the simulations of the radiation damage of MgO[53] 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 the understanding the observed effect.

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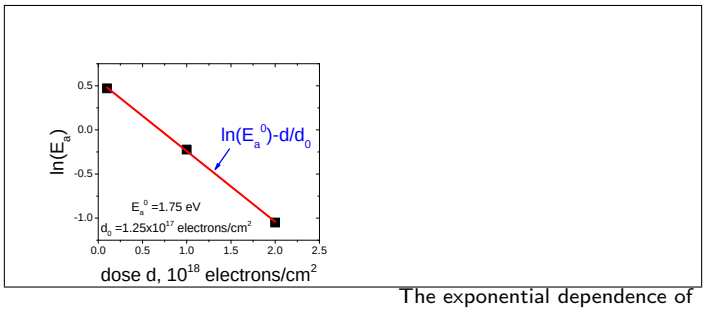
Graphical TOC Entry



The exponential dependence of the migration energy on the radiation fluence (estimate for MgF_2)

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