

## **Radiation-Induced Fluidity and Glass-Liquid Transition in Irradiated Amorphous Materials - 9082**

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### **ABSTRACT**

This paper describes the fluidity behaviour of continuously irradiated glasses using the Congruent Bond Lattice model in which broken bonds – configurons – facilitate the flow. Irradiation breaks the bonds creating configurons which at high concentrations provide the transition of material from the glassy to liquid state. An explicit equation of viscosity has been derived which gives results in agreement with experimental data. This equation provides correct viscosity data for non-irradiated materials and shows a significant increase of fluidity in radiation fields. It demonstrates a decrease of activation energy of flow for irradiated glasses. A simple equation for glass-transition temperature was also obtained which shows that irradiated glasses have lower glass transition temperatures and are readily transformed from glassy to liquid state e.g. fluidised in strong radiation fields.

### **INTRODUCTION**

Irradiation behaviour of materials is one of the topics of current interest. New materials and technologies are required to meet the demands of prolonged exposure to ionizing radiation and increased service life for the next generation of reactors including both fission and fusion nuclear reactors as well as to ensure a reliable immobilisation of resulting nuclear waste [1,2]. Ceramic and glassy materials are used in current generation of nuclear reactors. Some ceramics will be used in the strong radiation field of proposed fusion reactors. A number of ceramic formulations were designed to serve as host matrices for nuclear waste. Vitreous materials are used for several decades to immobilise nuclear waste and are planned to be used at an extending scale in the forthcoming years. Vitreous silica is often used as optical fibres operating in strong radiation fields.

The effect of irradiation on properties of ceramic and glasses has been studied in many works. A comprehensive overview of radiation effects in ceramics and glasses used for immobilization of high-level waste and plutonium disposition has been given in [3,4]. Irradiation plays an important role in long term behaviour of vitreous and crystalline materials used as nuclear wastefoms [5,6]. Radiation-induced effects in vitreous and crystalline materials may significantly change their properties. Irradiation of materials for very long times counting for hundreds and thousands of years in case of immobilised nuclear waste creates continuously excitations and long-lived defects, which evolve in a medium which is changing with time. Yet slowly occurring processes being hitherto out of attention may result in drastic consequences for the retention capacities of nuclear wastefoms. For example irradiation of insulators (e.g. glass and ceramics) causes intensification of surface diffusion processes and may result in surface instability [7]. Experimental studies of gamma irradiated clay soils at dose rates 1 kGy/h typical near industrial canisters with vitrified nuclear waste or spent radioactive sources, revealed changes in the microstructure of mineral particles constituting the soil [8]. Amorphisation of crystalline materials is the most evident result of irradiation and currently is intensely investigated for many potential nuclear waste host materials [3,9]. The amorphisation occurs at very high accumulated doses, e.g. critical amorphisation doses expressed in displacements per atom (dpa) are 0.2-0.3 for zirconolite, 0.3-0.4 for zircon, and far

higher (>10) for zirconia and monazite [5]. Studies of ceramics demonstrated that some crystalline structures can resist up to very high doses to swelling and amorphisation, e.g. cubic zirconia with fluorite-type structure [10]. At macro-length scales the highly damage-resistant cubic zirconia ceramic doped with  $^{238}\text{Pu}$  did not show any changes in physico-chemical properties during extended periods of time however, X-ray diffraction studies showed that some damage processes occur at micro-length scales depending on cumulative dose which suggests that the fluorite-type structure of cubic zirconia under self-irradiation demonstrates two competitive processes: defect accumulation and repeated defect self-annealing [10]. Other structures readily amorphise e.g. titanate pyrochlore solid solutions are amorphised with possible phase separation of the solid solution and a degradation of chemical durability [11,12]. There have been many observations of radiation-induced effects in glassy materials such as variations in refractive index, density and mechanical properties [4,6]. E.g. fused silica undergoes extensive densification on prolonged exposure to high-energy neutron, electron, and  $\gamma$ -ray radiation [13]. The densification of fused silica occurs also on exposure to laser radiation [14]. Both high-energy radiation and laser radiation-induced densification involve weakening of interatomic bonds and subsequent relaxation effects so that the densification follows a universal function of the dose. E.g. the amorphous  $\text{SiO}_2$  densification is caused by radiation-induced breaking of bonds and subsequent rearrangements of the  $\text{SiO}_2$  ring network into compacter rings with the density eventually saturated with fluence [13]. Ion irradiation of amorphous solids revealed stress relaxation, surface smoothing and have demonstrated viscous flow below melting temperatures [15,16]. E.g. the radiation-induced viscosity of amorphous silicon was estimated as  $10^{13}$  Pa s, which was approximately four orders-of-magnitude smaller than thermally-activated shear viscosity of non-irradiated amorphous silicon at room temperature. The low viscosity in the presence of the ion beam was explained as due to the creation of broken bonds by the ion beam that otherwise would have to be created by thermal activation [15]. In situ wafer bending measurements demonstrated that radiation-enhanced viscous flow is Newtonian, i.e., the strain rate is proportional to the stress [17]. It was found that the radiation-enhanced fluidity increases with increasing radiation energy loss (or ion mass) and the radiation-induced viscosity is approximately inversely proportional to the nuclear energy loss [16]. This conforms well to viscous flow mediated by flow defects [18] in which the viscosity is inversely proportional to the concentration of defects that contribute to viscous flow. Indeed the shear viscosity expressed in terms of the homogeneous density of broken bonds contributing to flow,  $C$ , can be written as [15,18]:

$$\eta(T) = kT \exp(H / kT) / (\gamma v)^2 v C, \quad (\text{Eq. 1})$$

where  $H$  is the activation enthalpy for flow,  $k$  is Boltzmann's constant,  $\gamma$  is the shear strain produced by the motion of a single defect,  $v$  is the volume of the defect, and  $v$  is an attempt frequency on the order of the Debye frequency. Before any defects have annihilated, the number of flow defects per unit length is proportional to the radiation energy loss, and hence the averaged viscosity is inversely proportional to the number of flow defects created by radiation per unit length [15,16]. The equation (1) was used to assess the activation enthalpy for flow in both irradiated and non-irradiated amorphous silicon [15]. It was found that for irradiated silicon  $H$  was smaller than 0.3 eV whereas the activation enthalpy for thermally activated flow in non-irradiated amorphous silicon is 1.8 eV which is roughly the energy required to form a broken bond. This difference in activation energy demonstrated that the radiation-enhanced flow is not governed by bond breaking but rather by bond motion [15]. Studies of electron-beam-induced sintering of submicrometre silicon particles demonstrated that the viscosity of amorphous silica drastically decreased by many orders of magnitude in a 200 kV TEM electron beam of  $10 \text{ A/cm}^2$  current density [19]. Although the temperature was estimated in those experiments as not higher than a few hundreds degrees, the viscosity was as low as  $10^8 - 10^9$  Pa s, which would correspond to temperatures above 1700 K. Such low viscosity of irradiated amorphous silica was attributed in [19] to the increase of defect concentration associated with the local structure. Molecular dynamics simulations have also demonstrated that point defects (Frenkel pairs) provide an efficient mechanism for radiation-induced viscous flow of solids [20]. By simulation of the injection of interstitial and vacancy like defects it has been demonstrated that point defects induce the same amount of flow as the recoil events, indicating that point-defect-like entities

mediate the flow process in solids even at 10 K. It was concluded that the radiation induced flow does not require thermal spikes (local melting) and that point defects equally, or, in many cases, more efficiently provide the viscous flow, which earlier has been associated with thermal spikes (local melting) [20]. Recent studies [21] have shown that electron microscope beam irradiation of fracture end in form of sharp corners and rough surfaces of a nanoscale glass fibre leads to an *in situ* annealing into a perfectly spherical shape, which evidences on effective electron-beam melting of glasses. One should note that these glasses would otherwise require temperatures above 1400 °C for their conventional melting, temperatures which obviously could not be reached in an electron microscope. This conclusion is in its turn evidenced by crystalline phases not melted in the electron beam. G. Moebus and co-authors [21] have also observed radiation-induced selective alkali accumulation and amorphous phase separation in alkali silicate glasses. Note that the last experimental findings are in a good agreement with the suggested model of radiation-induced unbinding of alkalis from the non-bridging oxygen sites in alkali-silicate glasses, which predicts a radiation-facilitated migration of alkalis with a lower cation-selective activation energy [22].

Although important studies on the effect of radiation on amorphous materials were carried out many aspects of behaviour of vitreous materials under irradiation have not been fully revealed. An important issue that requires additional studies is the viscous flow or fluidity behaviour under irradiation. The fluidity is rate-limiting for many processes and controls ageing and phase transformations in vitreous materials. Changes in the viscous flow behaviour in conditions of long-term irradiation are of a particular interest for components made of silicate glasses designed to be used in nuclear installations [1,2] as well as for nuclear waste immobilisation [3-6]. These changes can also be important in geology in understanding the viscous flow of Earth's deep interior [23,24]. The aim of this paper is to analyse the fluidity behaviour of amorphous materials in conditions of irradiation using the same approach as for non-irradiated materials. We limit our analysis herein to condensed amorphous oxide materials which can be either solid (vitreous) or liquid (melted).

## VISCOSITY OF AMORPHOUS MATERIALS

The viscous flow of materials occurs due to the stress applied. The viscosity of materials e.g. the viscosity coefficient  $\eta(T)$  relates the acting stress  $f$  to the strain rate  $\dot{\epsilon}$  via equation:

$$f = \eta(T) \cdot \dot{\epsilon} \quad (\text{Eq. 2})$$

The fluidity of amorphous materials is defined as the reciprocal of viscosity, e.g.

$$\varphi(T) = 1/\eta(T) \quad (\text{Eq. 3})$$

The fluidity depends on materials composition and external parameters such as pressure and temperature. The fluidity of amorphous condensed matter increases with temperature so that the higher the temperature the higher the fluidity. This behaviour is explained by temperature breaking of chemical bonds between atoms and molecules which constitute the material. The system of chemical bonds which hold the molecules together in a condensed matter defines the congruent bond lattice (CBL) model which is the congruent structure of its chemical bonds [25]. The CBL is a regular structure for crystalline materials and disordered for amorphous materials. A configuron in the CBL is defined as an elementary configurational excitation which involves breaking of a chemical bond and associated strain-releasing local adjustment of centers of atomic vibration [25]. The higher the temperature of an amorphous material the higher the configuron concentration. Configurons weaken the bond system so that the higher the content of configurons the lower the viscosity of an amorphous material. Moreover at very high concentrations configurons can form percolation clusters: this means that the material loses its rigidity as it becomes penetrated by a macroscopic (infinite-size) cluster made of broken bonds. The formation of percolation clusters made of configurons treats the glass transition as percolation-type phase transition in the system of configurons [26,27]. The concentration of broken bonds  $C(T)$  in the CBL in absence of irradiation is given by [25,28]:

$$C(T) = C_0 \exp(-G_d / RT) / [1 + \exp(-G_d / RT)] \quad (\text{Eq. 4})$$

where  $G_d = H_d - TS_d$  is the formation Gibbs free energy,  $H_d$  is the enthalpy,  $S_d$  is the entropy and  $C_0$  is the concentration of unbroken bonds at absolute zero. The viscosity of an amorphous material is related to the diffusion coefficient,  $D$ , of the configurons mediating the viscous flow via Stokes-Einstein equation:

$$\eta(T) = kT / 6\pi r D \quad (\text{Eq. 5})$$

where  $r$  is the radius of configuron. The configuron moving through material will perform jumps between different energy minima in a potential energy landscape. In a crystalline material these minima are associated with lattice or interstitial sites; similarly in an amorphous material these minima are associated with network sites. At each minimum in the energy-distance diagram, the configuron is in an equilibrium position. The energy  $G_m$  which is required to enable the configuron to jump across a barrier equals the difference in energy between the energy associated with the configurons being in equilibrium positions and the energy associated with the diffusing configuron (along with its neighbours, which must move apart to allow the jump) being in a saddle point configuration at a maximum in the energy-distance curve. The probability of the energy gathered is given by the Gibbs distribution:

$$w = \exp(-G_m / RT) / [1 + \exp(-G_m / RT)] \quad (\text{Eq. 6})$$

where  $G_m = H_m - TS_m$  is the Gibbs free energy of motion associated with a jumping configuron,  $H_m$  and  $S_m$  are the corresponding enthalpy and entropy of configuron motion. Assuming that the mean jump time of configurons is short compared to the mean residence time  $\tau(T)$  in network sites, the trajectory of a configuron is composed of a sequence of elementary jumps with average jump length  $\lambda$ . From these microscopic quantities the configuron diffusion coefficient can be defined by:

$$D = fg\lambda^2\nu(T) \quad (\text{Eq. 7})$$

where  $f$  is the correlation factor,  $g$  is a geometrical factor close to 1/6 and  $\nu(T) = 1/\tau(T)$  is the total jump frequency. The correlation factor equals unity for purely random hopping, and in general  $0 < f \leq 1$ . For defect mediated jumps the equation for the total jump frequency is given by:

$$\nu(T) = zp_0 f(T) \nu_0 w \quad (\text{Eq. 8})$$

where  $z$  is the number of nearest neighbours,  $p_0$  is a configuration factor (in simple cases  $p_0 = 1$ ),  $f(T)$  is the relative concentration of configurons given by Eq. (4), and  $\nu_0$  is the configuron vibrational frequency or the frequency with which the configuron attempts to surmount the energy barrier to jump into a neighbouring site which is of the order of the Debye frequency. Hence the viscosity of amorphous materials in absence of irradiation is given by [27-29]:

$$\eta(T) = A_1 T [1 + A_2 \exp(B / RT)] [1 + C \exp(D / RT)] \quad (\text{Eq. 9})$$

where

$$A_1 = k / 6\pi r D_0, \quad D_0 = fg\lambda^2 zp_0 \nu_0, \quad A_2 = \exp(-S_m / R), \quad B = H_m, \quad C = \exp(-S_d / R), \quad D = H_d \quad (\text{Eq. 10})$$

Equation (9) can be fitted to practically all available experimental data on viscosities of amorphous materials [29]. Moreover it can be readily approximated within a narrow temperature interval by known empirical and theoretical models such as VTF or Kohlrausch-type stretch-exponential relationships. The experiments show that is commonly the case, that  $A_2 \exp(B / RT) \gg 1$  so that the viscosity equation can be simplified to the  $\eta(T) = AT \exp(B / RT) [1 + C \exp(D / RT)]$ , where  $A = A_1 A_2$  [29,30]. Note that in contrast to many approximations the equation (9) can be used over wider temperature ranges and gives the correct Arrhenian-type asymptotes at high and low temperatures namely  $\eta(T) \cong AT \exp(B / RT)$  and

$\eta(T) \cong ACT \exp[(B+D)/RT]$  respectively. E.g. the equation (9) correctly gives Arrhenian type asymptotes at high and low temperatures with high activation energy of flow at low temperatures  $Q_H = B + D$  and low activation energy at high temperatures  $Q_L = B$ . Volf gives  $Q_L = 80-300$  kJ/mol for high temperatures and low viscosities when  $\log(\eta/\text{dPa s}) < 3$ , and  $Q_H = 400-800$  kJ/mol for low temperatures and high viscosities when  $\log(\eta/\text{dPa s}) > 3$  [30].

## VISCOSITY OF IRRADIATED AMORPHOUS MATERIALS

Broken bonds weaken the CBL and because of that the fluidity of condensed matter is higher at higher temperatures. Note that in contrast to condensed matter the fluidity of gases is lower at higher temperatures. In addition to temperature fluctuations other processes can cause bond breakages and so increase the fluidity of amorphous materials. E.g. bond breaking can occur under the action of ionizing radiation. The higher the intensity of irradiation the higher the rate of production of broken bonds contributing to flow thus the fluidity of amorphous materials is higher in the radiation fields. To quantify this increase it is necessary to analyse the link between the fluidity (viscosity) and radiation-induced broken bond concentration.

Solids in conditions of irradiation contain radiation-induced defects including broken bonds and are far from thermodynamic equilibrium. They tend to return to the equilibrium states however the relaxation to equilibrium passes through a number of metastable states and can be slow. In glasses this relaxation can be very slow as the glass itself is a metastable product although its lifetime is extremely long if no infinite. We assume that in conditions of continuous irradiation there is established a stationary state (although non-equilibrium) when the concentration of broken bonds does not depend on time. We can therefore describe the viscous flow processes in a steady state approximation assuming that the effects considered have significantly shorter characteristic times than the lifetime of radiation-induced broken bonds and that the fluidity adiabatically follows changes in the concentration of configurons contributing to viscous flow. The concentration of broken bonds in an irradiated amorphous material,  $C_i$ , is the sum of those created by thermal activation and those created by radiation,  $C_R$ , e.g.

$$C_i(T) = C_0 \exp(-G_{di}/RT) / [1 + \exp(-G_{di}/RT)] + C_R \quad (\text{Eq. 11})$$

Due to structural changes both the formation and motion Gibbs free energies of configurons in an irradiated material ( $G_{di}$  and  $G_{mi}$ ) can be different from that for a non-irradiated material particularly at high dose rates of radiation. In a first approximation we can however assume that the irradiated material has the same structure except additional broken bonds and assume that

$$G_{di} \cong G_d, \text{ and } G_{mi} \cong G_m \quad (\text{Eq. 12})$$

In the simplest model with  $C_R \ll C_0$  and one recombination channel the kinetic of radiation-induced bond breaking is described by equation:

$$\frac{dC_R}{dt} = k_b C_0 P_R - \frac{C_R}{\tau_b} \quad (\text{Eq. 13})$$

where  $k_b$  is the rate constant of bond breaking by radiation,  $P_R$  is the intensity of radiation (absorbed dose rate) and  $\tau_b$  is the lifetime of a broken bond created by radiation. In the steady state approximation  $dC_R/dt=0$  and so the concentration of radiation-induced broken bonds is given by:

$$C_R = k_b C_0 P_R \tau_b \quad (\text{Eq. 14})$$

One can see hence that the concentration of broken bonds in the radiation field is directly proportional to the intensity of radiation  $P_R$ . Note that radiation-induced defect concentrations generally depend on parameters of creation-recombination channels available and are often nonlinear with dose [31].

The viscosity of irradiated amorphous materials can thus be obtained accounting for all broken bonds in the matter e.g. for those created by thermal activation and those created by irradiation. This gives for the viscosity coefficient of irradiated amorphous materials the following equation:

$$\eta_R(T) = \eta(T) / \left[ 1 + k_b P_R \tau_b \left[ 1 + C \exp(D / RT) \right] \right] \quad (\text{Eq. 15})$$

The higher the intensity of radiation the higher the increase of fluidity. One can see from (14) that because of  $\left[ 1 + k_b P_R \tau_b \left[ 1 + C \exp(D / RT) \right] \right] > 1$  the viscosity of amorphous materials is lower in conditions of irradiation e.g. that the radiation is always increasing the fluidity. At very low levels of radiation when  $P_R \rightarrow 0$  the viscosity given by (14) is exactly the same as given by (9). However at higher levels of radiation the viscosity is drastically decreased by radiation-induced bond breaking processes.

It is important to note that intensive irradiation changes the activation energy of viscous flow from characteristic high values  $Q_H = B + D$  at low temperatures to low values  $Q_L = B$  which in absence of radiation are characteristic only for high temperatures. As the low activation energy of viscosity is exactly equal to the enthalpy of motion of bonds (see (10)) we conclude that the radiation-enhanced flow is indeed governed by bond motion rather than bond breaking which excellently agrees with experimental data [15] where both the decrease of viscosity and activation energy were observed.

The smaller the temperature the more significant is the radiation-induced decrease of viscosity in the same intensity radiation field which is similarly to the effect of radiation-induced unbinding of alkalis in silicate glasses [22]. In order to see the radiation-induced fluidity the intensity of radiation should be enough high, or the temperature should be enough low.

Consider the temperature of a material is  $T$  and define the critical dose rate  $P_R^*$  above which the radiation-induced increase of fluidity can be observed as

$$P_R^* = \exp(S_d / R) \exp(-H_d / RT) / k_b \tau_b \quad (\text{Eq. 16})$$

The lower the rate constant of radiation-induced bond breaking and the lifetime of broken bonds created by radiation the higher the intensity of radiation field required to observe the radiation-enhanced fluidity. The critical dose rate increases exponentially with temperature with the lowest  $P_R^*$  at lowest temperatures.

Consider the radiation field intensity is  $P_R$  and define the critical temperature  $T^*$  below which radiation-induced increase of fluidity can be observed as

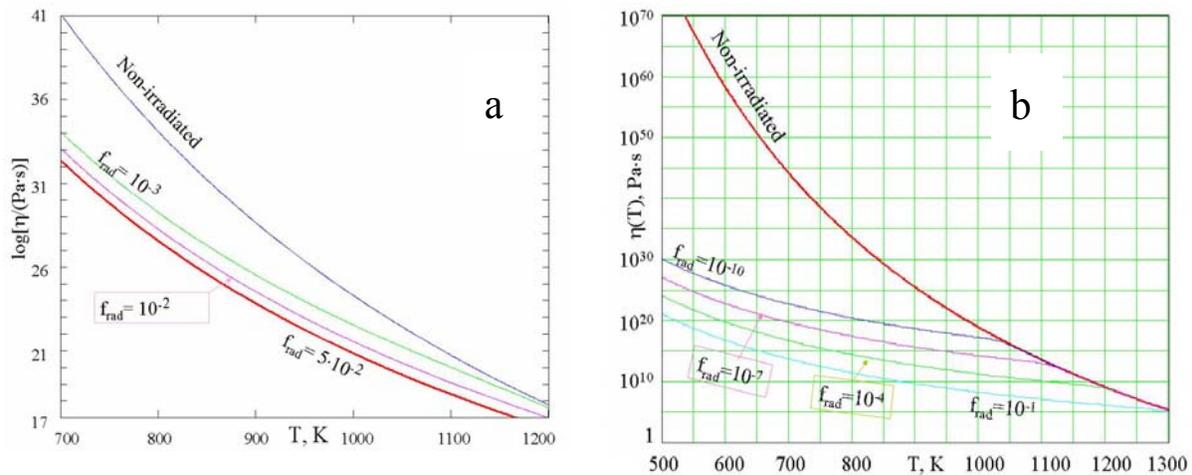
$$T^* = H_d / \left[ S_d - R \ln(k_b P_R \tau_b) \right] \quad (\text{Eq. 17})$$

The radiation-induced fluidity can be observed only if the intensity of radiation is above  $P_R^*$  and the temperatures are below  $T^*$ . The higher the intensity of radiation ( $P_R$ ), the rate constant of radiation-induced bond breaking ( $k_b$ ) and the lifetime of broken bonds created by radiation ( $\tau_b$ ) the higher the critical temperature  $T^*$ . However if the temperature is enough low  $T \ll T^*$  then the viscosity of an irradiated material can be approximately expressed as:

$$\eta_R(T) \cong (AT / k_b P_R \tau_b) \exp(B / RT) \quad (\text{Eq. 18})$$

This equation shows that the activation energy of viscosity of irradiated materials even at low temperatures (e.g. at  $T \ll T^*$ ) has low values  $Q_L = B$ . In absence of radiation such behaviour of viscosity is characteristic only for temperatures much above the glass transition temperature [26-29].

Equation (15) can be used to estimate the viscosity of irradiated amorphous oxide materials. It requires however data on thermodynamic parameters of configurons which in the first approximation (11) can be taken as from non-irradiated materials [29]. Fig. 1 illustrates the results of such calculations for irradiated amorphous silica and anorthite at several irradiation dose rates.



**Fig. 1. Viscosities of non-irradiated and irradiated amorphous (a) silica and (b) anorthite as a function of temperature at several dimensionless irradiation dose rates  $f_{rad} = k_b P_R \tau_b$ .**

One can see that for anorthite the decrease of viscosity is by many orders of magnitude even at relative low dose rates when the radiation creates a relative small fraction of broken bonds (e.g.  $f_{rad} = 10^{-10}$ ). The radiation-induced increase of fluidity is most evident at lower temperatures and is almost not seen at high temperatures. One can also observe that the activation energy of viscous flow has changed at low temperatures ( $T < 1000$  K) from characteristic high values  $Q_H$  to low values  $Q_L$  which are characteristic to non-irradiated materials only at high temperatures. The lower the dimensionless irradiation dose rates  $f_{rad} = k_b P_R \tau_b$  the lower the temperature  $T^*$  below which the radiation-induced fluidity is significant.

### GLASS TRANSITION IN RADIATION FIELDS

Often the liquid-glass transition is regarded as a transition for practical purposes rather than a thermodynamic phase transition [32]. By general agreement it is considered that a liquid on being cooled becomes practically a glass when the viscosity equals  $10^{12}$  Pa·s ( $10^{13}$  poise) or where the relaxation time is  $10^2$  s [32], e.g. the practical purpose or relaxation glass transition temperature  $T_{g,relax}$  is found from the viscosity-temperature relationship  $\eta(T_{g,relax}) = 10^{12} (Pa \cdot s)$ . The irradiation decreases the viscosity thus accordingly to (14) the glass transition temperature of irradiated materials decreases with irradiation dose rate. In an another approach the glass-liquid transition is considered as a percolation in the system of broken bonds [26,27,33]. Indeed at very high configuron concentrations they form percolation clusters so that the material is penetrated by macroscopic clusters made of broken bonds and becomes liquid-like. Although no symmetry changes can be revealed in the atomic distribution at a such change, there will be

a symmetry change expressed by a step-wise variation of Hausdorff dimension of bonds, namely by the reduction of Hausdorff dimension of bonds from the 3 in the glassy state to the fractal  $d_f = 2.55 \pm 0.05$  in the liquid state [26,27,33,34]. The temperature of glass-liquid transition in a such approach is found equalizing the concentration of configurons (3) to the threshold percolation concentration  $f_c$  [26,34]

$$C(T_g)/C_0 = f_c \quad (\text{Eq. 19})$$

This results in an explicit equation for the glass-transition temperature

$$T_g = H_d / [S_d + R \ln[(1 - f_c) / f_c]] \quad (\text{Eq. 20})$$

which is in an excellent agreement with experiment [26,27, 29,34]. The glass-transition temperature  $T_g$  for an irradiated material can be found assuming that the configuron concentration (11) achieves the universal critical density given by the percolation theory

$$C_i(T_g)/C_0 = f_c \quad (\text{Eq. 21})$$

This equation gives a reduced glass-transition temperature of irradiated materials compared non-irradiated ones:

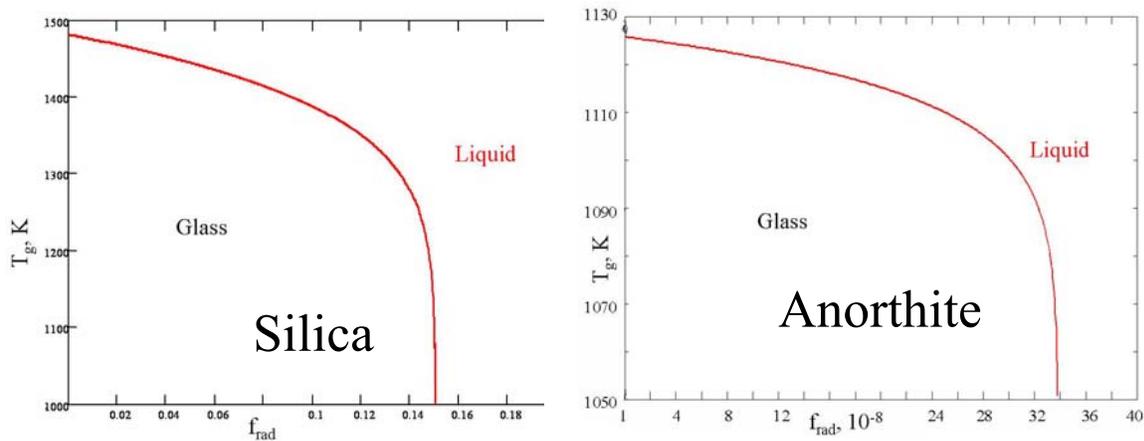
$$T_{ig} = H_{id} / [S_{id} + R \ln[(1 - f_c + k_b P_R \tau_b) / (f_c - k_b P_R \tau_b)]] \quad (\text{Eq. 22})$$

where  $H_{id}$  and  $S_{id}$  are the enthalpy and the entropy of formation of configurons of the irradiated material. The higher the dimensionless irradiation dose rates  $f_{rad} = k_b P_R \tau_b$  the lower the glass-transition temperature. Equation (22) shows that at irradiation dose rates

$$P_R \geq f_c / k_b \tau_b \quad (\text{Eq. 23})$$

the glass transition temperature drops to zero. It means that the amorphous oxide material in a strong radiation field  $P_R \geq f_c / k_b \tau_b$  is liquid even at zero Kelvin. Note that the glassy material transforms to liquid in strong radiation fields e.g. are fluidised not because of thermal melting but because of intensive radiation-induced bond breaking.

To calculate the glass-transition temperatures of irradiated amorphous materials  $H_{id}$  and  $S_{id}$  are required. These in the first approximation can be again taken accordingly to (12) e.g. as from a non-irradiated material (see [29]). Fig. 2 shows the results of such calculations for the irradiated amorphous silica and anorthite.



**Fig. 2. Glass transition temperature of irradiated amorphous silica and anorthite as a function of dimensionless irradiation dose rate  $f_{rad} = k_b P_R \tau_b$ .**

From Fig. 2a one can see that above the intensity of radiation  $f_{rad}=0.15$  the vitreous silica is melt by radiation. Fragile glassy materials have lower percolation thresholds [29,34] hence these materials can be melted at significantly lower intensities of radiation. Note that although fluidised glass melts are very viscous so that in order to experimentally detect the viscous flow very small e.g. sub-micrometer or nano-scale size samples are required such as those studied by Ajayan and Iijima [19] and Moebus et.al. [21]. Fluidisation of amorphous materials in radiation field can be probably used for high intensity dosimeters required for new generation of nuclear reactors and radiation devices [35]. The radiation-induced lowering of glass transition temperature and increase of fluidity can also have important technological implications e.g. in nanopatterning, welding, sintering and surface smoothing of materials [21,36,37]. Note that the radiation used for radiation-induced fluidisation can be both high energetic such as neutron, electron and  $\gamma$ -ray radiation and photonic such as those generated by a laser.

## CONCLUSIONS

We analysed the fluidity of irradiated amorphous oxide materials based on CBL model accounting for the mediating role of broken bonds – configurons – which facilitate the flow and at high concentrations provide the transition of material from the glassy state to liquid. An explicit equation of viscosity (15) was obtained which gives the correct viscosities of non-irradiated amorphous materials and shows the increase of fluidity and decrease of activation energy of flow of irradiated glasses and which is in an agreement with experimental data. The radiation-induced increase of fluidity causes a reduction of glass-transition temperature which can be calculated using both the relaxation approach to glass transition and configuron percolation model of glass transition. In the last case a simple equation for glass-transition temperature (22) was obtained which shows that irradiated glasses have lower glass transition temperatures and are readily transformed from glassy to liquid state (fluidised) in strong radiation fields. The results obtained can be useful both for current applications of glassy materials in conditions of irradiation and for developing radiation technologies such as welding, surface polishing and smoothening.

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