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IR Spectra of Aqueous Disperse Systems Adsorbing Atmospheric Gases: 2. Argon

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Abstract—The absorption, reflection, and scattering of IR radiation by aqueous ultradisperse systems that absorb argon are studied with the molecular dynamics method on the basis of a flexible molecule model. Both the real and imaginary parts of dielectric permittivity are substantially increased at frequencies corresponding to intramolecular vibrations of atoms when each cluster of aqueous system adsorbs one argon atom. This effect disappears for the real part and becomes weak for the imaginary part of dielectric permittivity when there are two argon atoms added per cluster. In the region of intramolecular frequencies, the absorption coefficient of aqueous disperse systems containing argon increases as well. The reflection coefficient of the systems of water clusters that absorbed argon decreases in the frequency region of vibrations of molecules and increases in a frequency range corresponding to intramolecular vibrations. The power of radiation generated by cluster systems at the expense of thermal energy increases considerably when there is one adsorbed argon atom per cluster and decreases with a twofold increase in the number of argon atoms in clusters.

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INTRODUCTION

The abundance of argon on Earth is greater than all other elements of its group. The content of argon in the atmosphere is 0.934 vol %. The solubility of argon in water at 273 K and pressure of 0.1 MPa is equal to 57.8 mg/l. In water, argon is dissolved better than nitrogen. Argon does not contribute directly to the Earth's greenhouse effect because its atoms have no dipole moments. Its indirect influence can be realized only through the incorporation into aqueous disperse system. By the calculation of the surface of pair-additive potential energy, it was shown that the most advantageous structure of argon atoms captured one water molecule is the icosahedron containing H_2O molecule in its center [1]. Quantum-chemical calculations indicate that, upon the capture of argon atoms by grouping water molecules, pentagonal dodecahedron composed of water molecules and containing argon atom in its interior is formed with higher probability [2]. The presence of argon atoms in a water cluster should lead to changes in vibration frequencies of water molecules and their intramolecular vibrations that is reflected in the IR spectra. The effect of absorption of rare gases on the spectral characteristics of water clusters has not so far been studied.

The account of the polarizability in the model makes it possible to investigate nonuniform and isotropic properties of water, for example, near the surface separating the liquid from the solid body or gas. When studying the behavior of water molecules near the molecules of another kind, the important role is assigned to the degrees of freedom that are responsible for the fast response of electron subsystem. It could be expected that, upon the interaction with water clusters, argon atoms tend to penetrate inside the polar medium, because argon dissolves in water, although in small quantities. However, the motion of argon atom to the depth of the $(H_2O)_i$ cluster is related with the large work done on the rearrangement of cluster structure. In this case, the hydrophobic effect is manifested that hampers the construction of the cell from water molecules around the argon atom but stabilizes such cell in the bulk water.

The use of intramolecular flexibility allows us to more precisely describe some vibrational properties of water molecules and their deformation at perturbations. Many of these effects are related with the influence of molecules of another kind or the presence of interfaces. However, substantial changes in the dipole moments of molecules and, hence, in dielectric properties of molecules under the action of temperature and pressure can also occur in pure water. The account of intramolecular flexibility and electronic polarizability in the model enables us to better understand changes in some special properties of water, but, in general, this does not improve the accuracy of determining its thermodynamic and structural characteristics, as compared with traditional models of rigid molecules. In particular, there is no improvement in describing the maximum of density of water and its thermodynamic properties at high temperatures [3]. However, the use of the flexible molecule model leads to more accurate calculation of

Table 1. Parameters of atom–atom interaction potential of Ar–H_2O system

System	Exponent 6			Repulsion
	ε/k, K	r_m, nm	β	B, $\mathrm{nm^{12}kcal/mol}$
Ar–OH_2	156.8	0.3556	12.5	
Ar–HOH	–	–	–	1.259×10^{-7}

spectra of reflection of IR radiation by aqueous disperse media [4]. This study is based on the construction of flexible molecule model by the procedure developed in [5–7]. The calculation of spectral characteristics in such model was performed in [8]. Here, we used analogous procedure for this purpose.

The goal of this work is to study the effect of the absorption of argon by aqueous disperse systems on their ability to absorption, reflection, and scattering of IR radiation.

ARGON–WATER INTERACTION POTENTIAL

Argon–water pair potential obtained by ab initio calculations can be presented in the form [2]

$$\Phi(r) = \frac{\varepsilon}{\beta - 6}\left[6\exp\left[\beta\left(1 - \frac{r}{r_m}\right)\right] - \left(\frac{r}{r_m}\right)^6\right], \quad (1)$$

where r is the distance between argon and oxygen atoms. The values of potential parameters ε, β, and r_m are listed in Table 1. However, the calculation of interaction energy between water molecules and argon atom leads to some discrepancy between the results obtained using formula (1) and by direct ab initio calculations. The delocalization of electrons along the hydrogen bond creates the effect of additional force acting on atom (molecule) occupying the guest cell. In order to account for the effect of the hydrogen bond in the argon–water system, pair atom–atom potential (1) should be corrected for the changes in Ar–HOH interaction. Additional Ar–HOH interaction was taken into account by the inclusion of interaction potential characterizing pure repulsion [9]

$$\Phi_{\mathrm{repul}}(r) = \frac{B}{r^{12}}. \quad (2)$$

The value of parameter B is given in Table 1. The use of additional interaction potential in the form of Eq. (2) makes it possible to minimize the errors in describing the interaction between guest argon atom and water molecules creating the cell for this atom.

EMISSION OF ELECTROMAGNETIC WAVES BY OSCILLATOR

In the model of emission of electromagnetic waves by the oscillator, the distribution of radiation intensity over frequencies has the following form [10]:

$$I_\omega = I_0 \frac{\gamma}{2\pi} \frac{1}{(\omega - \omega_0)^2 + \gamma^2/4}, \quad (3)$$

where total intensity I_0 is expressed as

$$I_0 = \int_0^{\omega_{\max}} I_\omega d\omega. \quad (4)$$

The $\gamma = \frac{2q^2\omega_0^2}{3mc^3}$ value in Eq. (3) represents the natural width of Lorentz contour determining the lifetime of the excited state of oscillator as $\tau_{\mathrm{rel}} = 1/\gamma$. Here, q is the charge of atom oscillating with characteristic frequency ω_0, m is its mass, and c is the speed of light. The frequency corresponding to the major maximum of ω_0 spectrum was used as the magnitude of $P(\omega)$ and the value of limiting operating frequency was $\omega_{\max} = 3500\ \mathrm{cm}^{-1}$. Vibrations of hydrogen atoms in water molecules and nitrogen atoms in the N_2 molecule were considered. Radiant friction was absent ($\gamma = 0$) for argon atoms having no electric charge.

DIELECTRIC PERMITTIVITY OF AQUEOUS DISPERSE SYSTEMS

The character of argon absorption by the $(H_2O)_{50}$ cluster is illustrated by Fig. 1, where the cluster configuration is shown 20 ps after the appearance of one (*a*) or two (*b*) Ar atoms near the cluster. During this time, Ar atoms came closer to the cluster so that they became its integral part. However, even in the case of the absorption of one Ar atom, its penetration inside the cluster appeared to be hindered and, as a result, the atom was accommodated on the surface. The shape of a cluster changed due to the absorption of two Ar atoms and its size along the line connecting Ar atoms becomes shorter than the size in perpendicular direction. The solubility of argon in water is 2.37-fold higher than that of nitrogen [11]. In the case of dispersion medium, this difference is exhibited as the closer contact between Ar atoms and water clusters than between N_2 molecules and the same cluster for the identical period of time.

Let us classify ultradisperse systems using statistical weights for constituting clusters in accordance with the procedure described in [4, 8]:

(I) the set of $(H_2O)_i$ clusters for systems $10 \le i \le 50$ with the step $\Delta i = 5$;

(II) the region filled with $Ar(H_2O)_i$ clusters;

(III) the space occupied by $(Ar)_2(H_2O)_i$ aggregates;

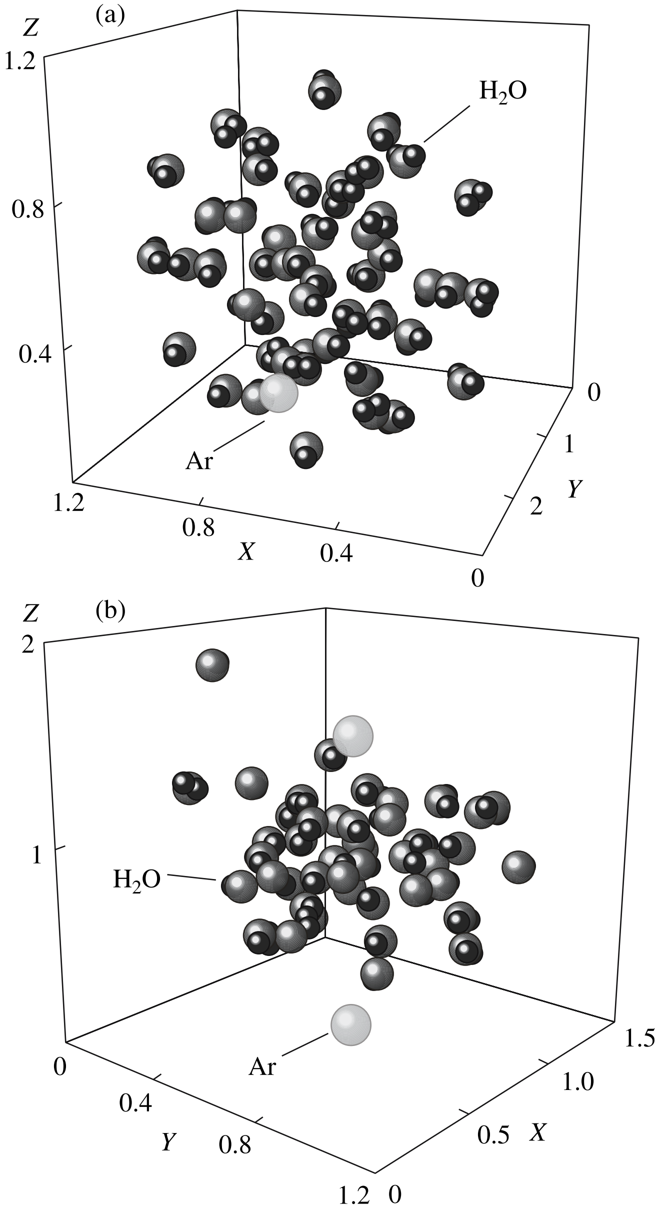

Fig. 1. Configurations of (a) $Ar(H_2O)_{50}$ and (b) $(Ar)_2(H_2O)_{50}$ clusters corresponding to time of 20 ps after the addition of Ar atoms to a system. Values of distances on coordinate axes are presented in nanometers.

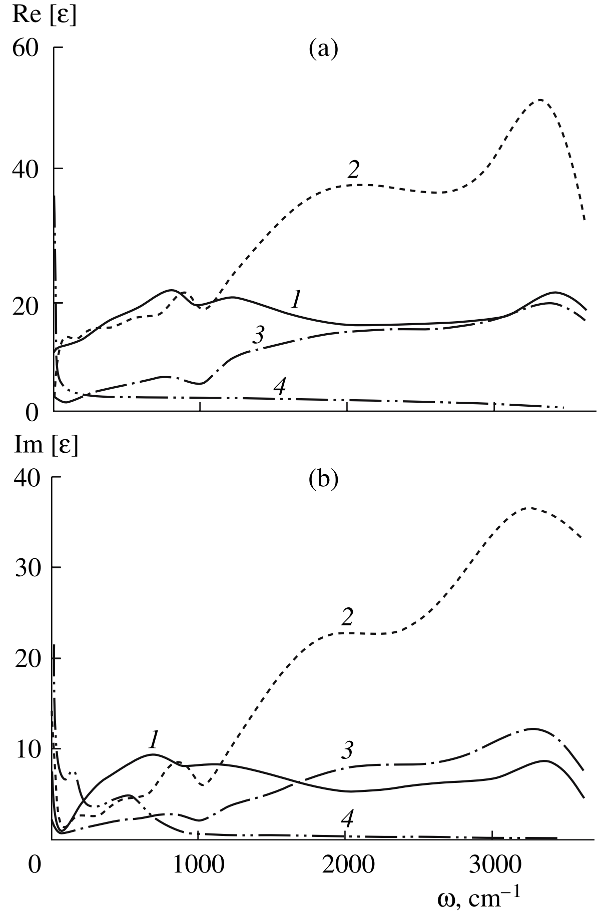

Fig. 2. (a) Real and (b) imaginary parts of the dielectric permittivity of disperse systems (*1*) I, (*2*), II, and (*3*) III, and (*4*) bulk water. Panel (a): calculation, TIP4P model [12]. Panel (b): experiment [13].

(IV) the medium composed of $(H_2O)_i$ clusters that absorb one N_2 molecule;

(V) the system composed of $(N_2)_2(H_2O)_i$ aggregates.

The dielectric permittivity of aqueous disperse system changes substantially after the capture of argon atoms (Fig. 2). In this case, both the real and imaginary parts of ε increases appreciably at the frequencies $\omega > 1050$ cm^{-1} upon the absorption of one argon atom by each water cluster. The addition of the second argon atom to each water cluster decreases the ε' value throughout the studied frequency range. At the same time, the ε'' value decreases only at $\omega < 1670$ cm^{-1}; at the opposite sign of the inequality, the ε'' values of the dispersion medium that has captured two argon atoms per cluster are larger than frequency-correspondent ε'' value for pure dispersed water. At $\omega > 260$ cm^{-1}, the ε' values and, at $\omega > 680$ cm^{-1}, the ε'' value for all considered disperse systems becomes larger that corresponding characteristic for the bulk water [12, 13].

IR ABSORPTION SPECTRUM

The absorption of argon leads to a significant change in the absorption coefficient α of aqueous disperse system beginning with the frequency 1200 cm^{-1}, i.e., with the region, where intramolecular vibrations in water contribute mainly to the IR spectrum (Fig. 3). Note that a twofold increase in the Ar concentration does not result in such strong changes in the α(ω) spectrum as took place when passing from the pure dispersed water to the aqueous disperse system containing argon (system II). This also manifests that the change in

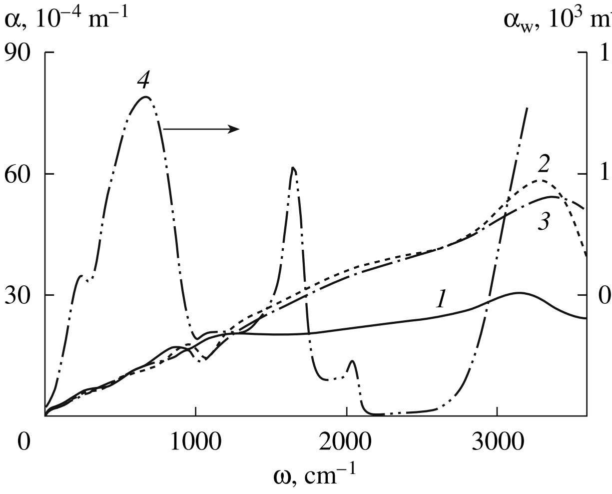

Fig. 3. Absorption coefficients of IR radiation of aqueous disperse systems (*1*) I, (*2*) II, and (*3*) III, and (*4*) bulk water, experiment [13]. Curve *4* refers to the right-hand coordinate axis.

the absorption spectrum is related with the changes in frequencies of intramolecular vibrations in water after the absorption of Ar atoms by the clusters. The position of the major maximum of α(ω) spectrum shifts from 3160 cm^{-1} for pure dispersed water (system I) to 3280 and 3390 cm^{-1}, respectively, for disperse systems II and III that absorbed argon. In the region $\omega < 1200$ cm^{-1} of α(ω) spectrum determined by vibrations of molecules, the major maximum also shifts toward higher frequencies after the absorption of Ar atoms by the clusters. The major maximum of this part of IR spectrum for the bulk water falls on 700 cm^{-1}, that is, corresponds to lower frequency than the localization of corresponding maximum for pure dispersed water (840 cm^{-1}).

In general, the absorption of IR radiation by the systems formed by monodisperse clusters constituting systems II and III proceeds in the same manner beginning with the cluster size $i = 10$ and ending with the size $i = 50$ (Fig. 4). Here, the intensity of absorption rises with an increase in frequency passing through a number of extreme points. For many clusters, the largest α value fits frequency 3300 cm^{-1}; however, at $i = 10$, 25, 30, and 35, the major maximum is positioned beyond the region limited by the frequency 3500 cm^{-1}. As a rule, the part of α spectrum corresponding to $\omega < 1200$ cm^{-1} determined by the vibrations of the centers of molecule masses is separated from the spectrum ($\omega > 1200$ cm^{-1}) (that is created by the vibrations of atoms in a molecule) by some extreme point, most often, by the minimum. Integral intensity of the absorption of IR radiation by monodisperse systems, most likely, is determined by the structure of constituting cluster rather than by its size.

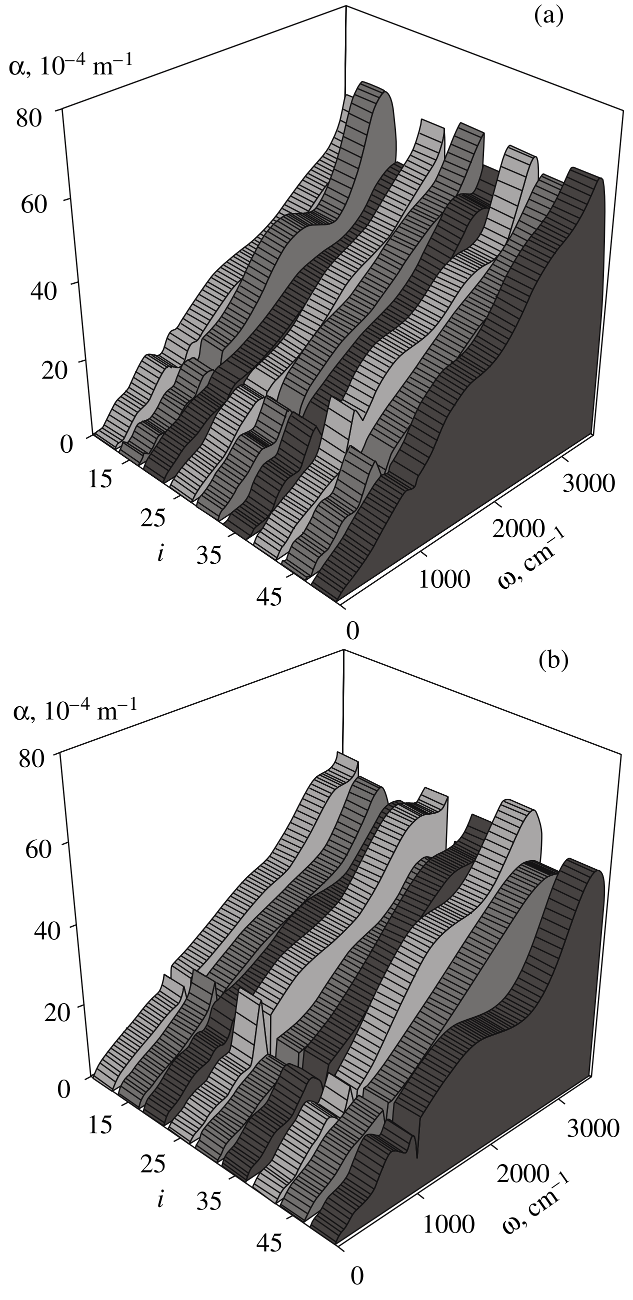

Fig. 4. Absorption coefficients of IR radiation for monodisperse systems of (a) Ar$(H_2O)_i$ and (b) $(Ar)_2(H_2O)_i$ clusters containing *i* water molecules.

SPECTRA OF REFLECTION AND POWER OF SCATTERED ENERGY AND THE NUMBER OF ACTIVE ELECTRONS

The pattern of the spectrum of reflection, *R*(ω), of plane monochromatic wave by disperse systems changes with the absorption of Ar atoms by the clusters (Fig. 5). Pure dispersed water is characterized by the *R*(ω) spectrum with the major maximum in the vicinity of 780 cm^{-1} and extended minimum with the lowest *R* value at 2625 cm^{-1}. When one Ar atom is added to each

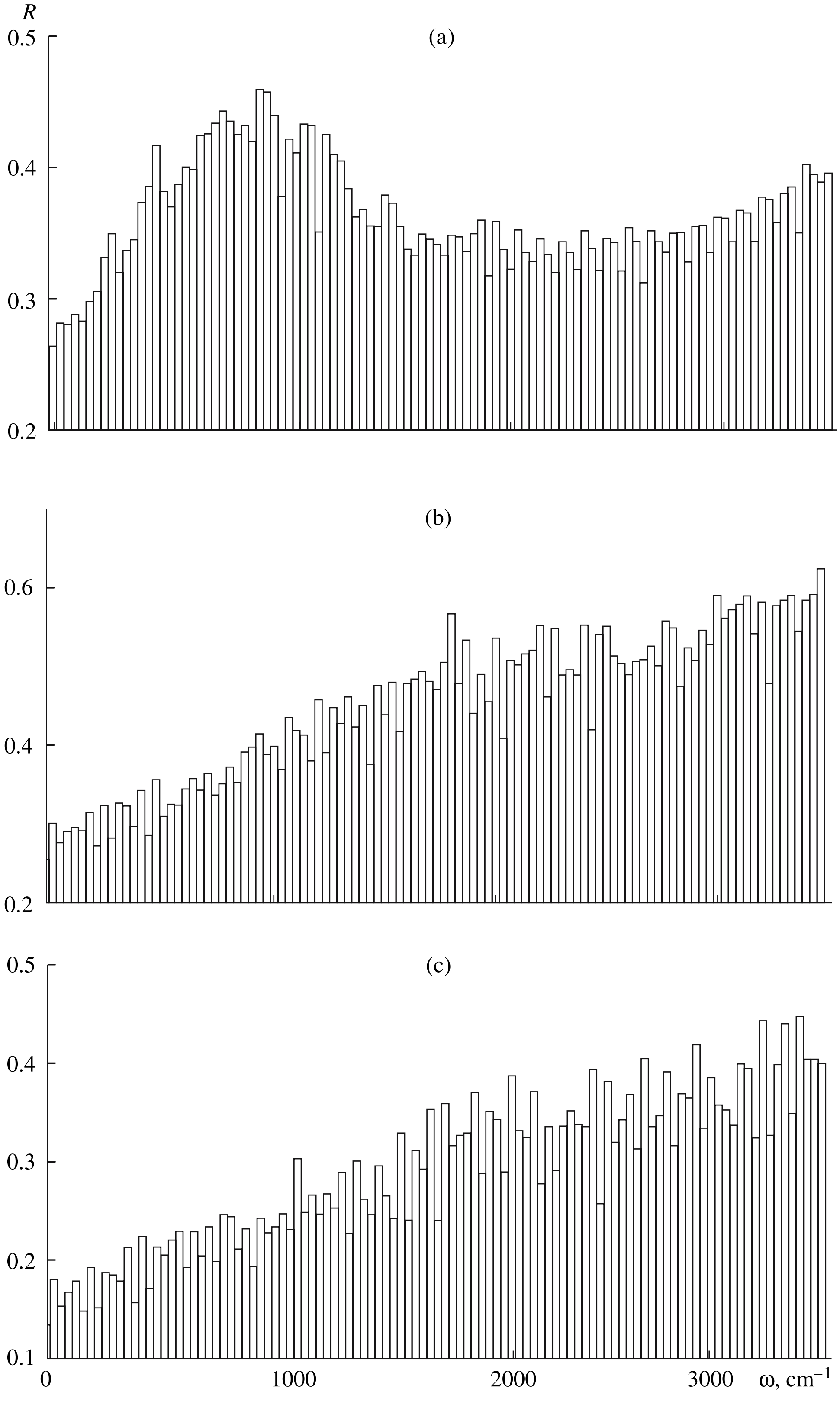

Fig. 5. Reflection coefficients of IR radiation for aqueous disperse systems (a) I, (b) II, and (c) III.

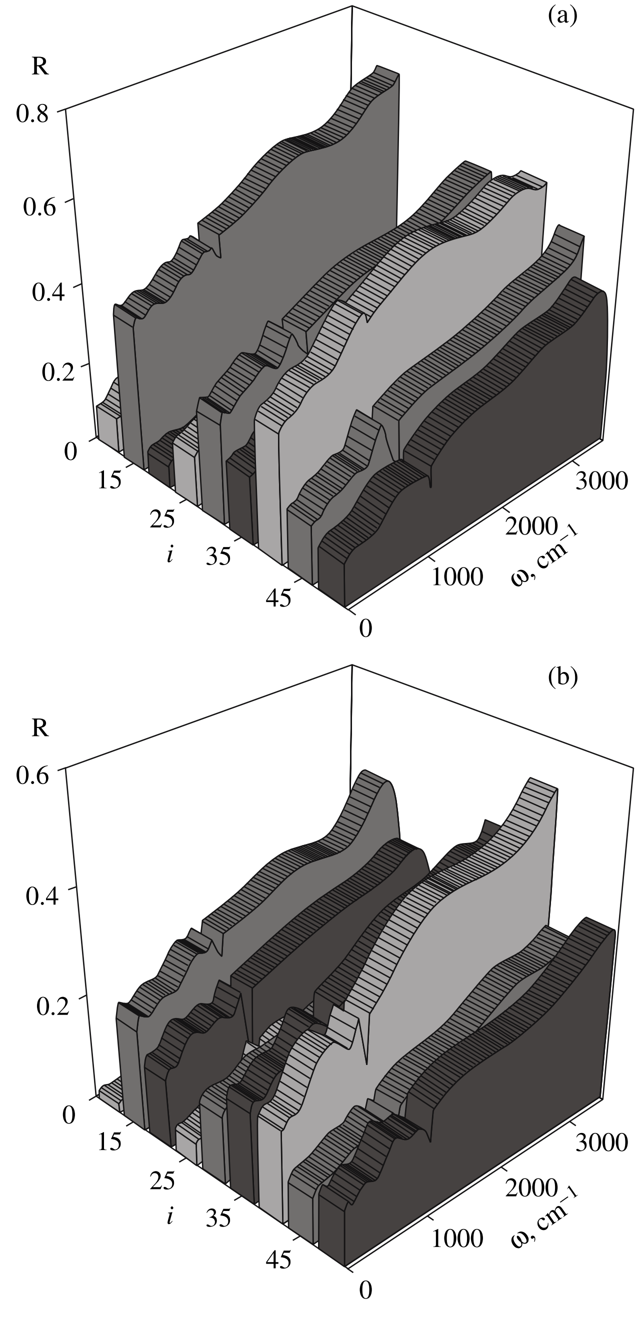

Fig. 6. Reflection coefficients of IR radiation of monodisperse systems of (a) $Ar(H_2O)_i$ and (b) $(Ar)_2(H_2O)_i$ clusters containing i water molecules.

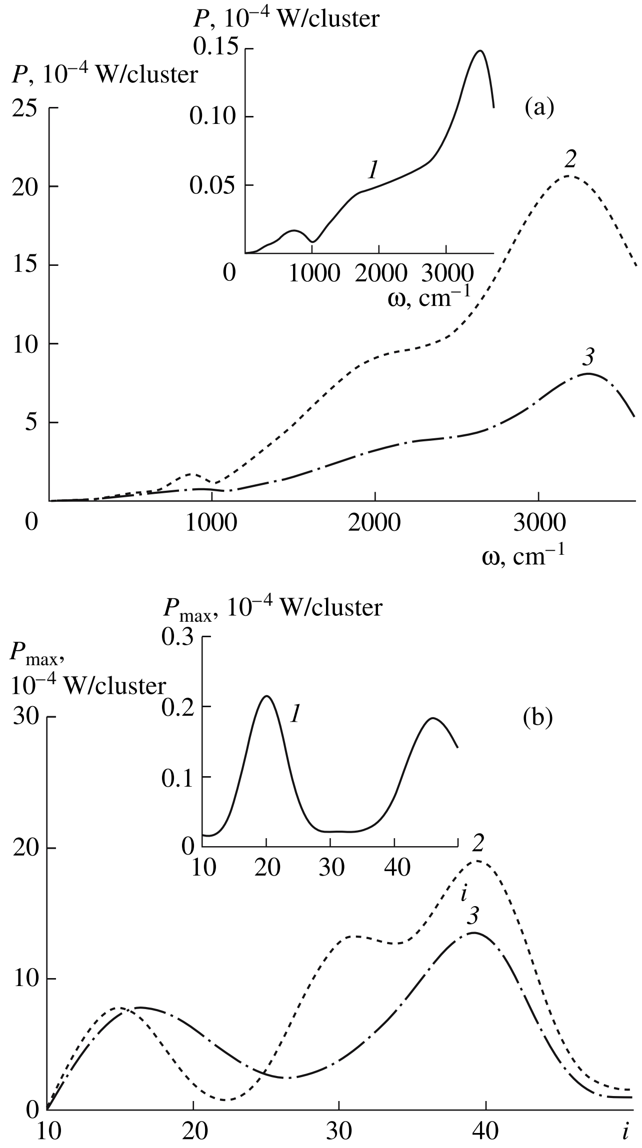

Fig. 7. (a) The power of radiation generated upon the dissipation of thermal energy; (b): maximum power of IR radiation in the ensembles of clusters containing i water molecules in different systems: (*1*) I, (*2*) II, and (*3*) III.

cluster, the reflection of the plane wave increases beginning with the frequency of about 1200 cm^{-1} and achieves the largest value ($R = 0.62$) at $\omega = 3500$ cm^{-1}. However, the addition of the second Ar atom to each studied cluster leads to a decrease in R values throughout the studied frequency range: the integral intensity of reflection for system III is lower than for system II.

The $R(\omega)$ spectra of systems formed by monodisperse water clusters with one or two Ar atoms considerably differ from each other by their patterns and intensities (Fig. 6). Among the systems of the first type (with one Ar atom in each cluster), the system of $Ar(H_2O)_{15}$ clusters is characterized by the $R(\omega)$ spectrum with the highest integral intensity, while among the systems of the second type (with two Ar atoms per cluster), the most representative is the system of $Ar(H_2O)_{10}$ clusters. The presence of the second Ar atom in clusters leads to the enhanced resolution of the peaks of $R(\omega)$ spectrum in the region of low frequencies ($\omega < 1000$ cm^{-1}).

Clusters are capable of emitting the absorbed energy. The rate of energy dissipation P or the radiation power are important characteristics. Frequency dependences of P value for systems I–III are shown in Fig. 7a. It is seen that, as a result of the addition of one Ar atom to each water cluster, the maximum emission power of IR radiation increases, at least, by the order of magnitude. However, the rate of energy dissipation drastically lowers due to the addition of one more Ar atom to each cluster. Upon consecutive increase in the argon concentration, the position of the major maximum of $P(\omega)$ function shifts first to the left and then to the right so that, finally, these positions correspond to 3480, 3190, and 3400 cm^{-1} for systems I, II, and III, respectively. The maximum of emission power as a function of the cluster size is shown in Fig. 7b. Among clusters of pure dispersed water, $(H_2O)_{20}$ and $(H_2O)_{45}$ aggregates emit the energy most intensely. In system II, whose each cluster absorbs one Ar atom, the $Ar(H_2O)_{40}$ aggregate demonstrates the highest rate of energy dissipation. The ability of this cluster to high-power emission is retained also after the absorption of one more Ar atom.

Qualitative characteristics of emission are represented by the model of interaction between electromagnetic waves and harmonic oscillator. Characteristic vibration times in the presence of the Lorentz force of radiant friction, together with the corresponding number of damping vibrations for each system, are listed in Table 2. It is seen that relaxation time τ_{rel} and the number of vibrations, n_{vib}, notably increase after the absorption of the N_2 molecules and especially of argon atoms by disperse systems. The period of vibrations of the damped oscillator represented aqueous systems with absorbed N_2 molecules or Ar atoms only slightly decreases, as compared with the τ_{vib} value of the oscillator characterizing pure dispersed water. The largest τ_{rel} and n_{vib} magnitudes are referred to system III; the lowest, to system I. If it had been assumed that the time of the propagation of vibrations in the medium (air) had coincided with the relaxation time, we could roughly estimate the distance at which thermal radiation produced by the considered systems is stopped. The IR radiation passes the longest distance (≈1270 km) for system III; the shortest distance (≈300 km), for system I.

For clusters of pure water, beginning with the size $i = 20$, the number n_{cl} of electrons participating in the electromagnetic interaction with the outer IR radiation changes slightly with increasing cluster size (Fig. 8). When one Ar atom is added to the cluster, the $n_{cl}(i)$ dependence becomes strongly oscillating. The $Ar(H_2O)_{15}$ and $Ar(H_2O)_{10}$ clusters are characterized by the largest and the lowest numbers of "active" electrons, respectively. The addition of one more Ar atom to the cluster lowers the vibration amplitude of $n_{cl}(i)$ function. In this case, the largest amount of electrons interacting with the radiation is present in the $(Ar)_2(H_2O)_{40}$ cluster; the smallest amount, in the $(Ar)_2(H_2O)_{10}$ cluster. In all the cases considered, the value of n_{cl} is by 5–7 orders of magnitude smaller than the density of free electrons in metals.

Table 2. Relaxation times τ_{rel}, vibration periods τ_{vib}, and numbers n_{vib} of damped vibrations of oscillator (cluster system)

System	τ_{rel}, s	τ_{vib}, ps	n_{vib}, 10^8
I – $(H_2O)_i$	0.00100	1.34	7.46
II – $Ar(H_2O)_i$	0.00301	1.27	23.69
III – $(Ar)_2(H_2O)_i$	0.00424	1.33	31.89
IV – $N_2(H_2O)_i$	0.00177	1.32	13.41
V – $(N)_2(H_2O)_i$	0.00261	1.29	9.44

DISCUSSION

In this work, we studied the absorption of argon atoms by water clusters. Resultant argon–water interaction potential is characterized by a weak attraction but having "soft" repulsive component similar to the repulsion between oxygen atoms upon the interaction of O_2 and H_2O molecules. Due to "soft" repulsion, argon atoms are brought into closer contact with water molecules than nitrogen molecules for which the repulsion branch characterizing N–O interaction has a steeper form. This allows Ar atoms to deeper penetrate into the water cluster, as compared with nitrogen molecules. The capture of one Ar atom by each cluster results in a considerable increase in the real and imaginary parts of

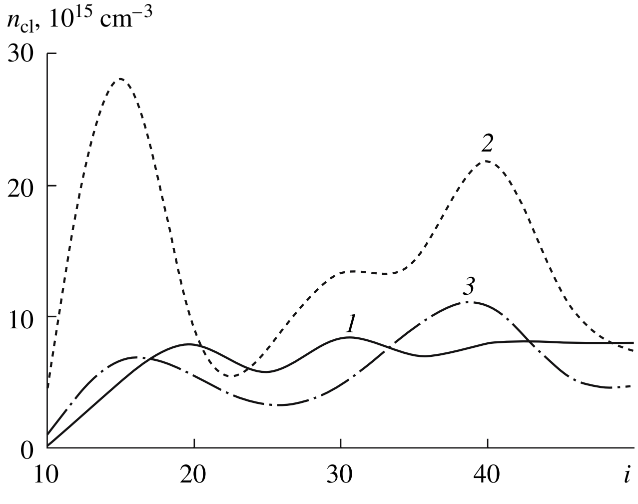

Fig. 8. Number of electrons interacting with outer electromagnetic radiation in clusters containing i water molecules in different systems: (*1*) I, (*2*) II, and (*3*) III.

dielectric permittivity of system II composed of these species at frequency $\omega > 1050$ cm^{-1}. Such a behavior of $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ functions is not observed upon the capture of two Ar atoms or the absorption of N_2 molecules by each cluster. The character of $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ dependences for system III (when two Ar atoms are present in each cluster) is mainly similar to the behavior of these functions for the system composed of $(N_2)_2(H_2O)_i$ clusters, except for the facts that $\varepsilon'(\omega)$ function of system III never intersects the plot of corresponding function for pure dispersed water and the $\varepsilon''(\omega)$ function of system III somewhat earlier (beginning with frequency 1660 cm^{-1}) than system V acquires the values larger than $\varepsilon''(\omega)$ function of system I. The intersection point of the plots of $\varepsilon''(\omega)$ function for $(N_2)_2(H_2O)_i$ system and pure dispersed water corresponds to the frequency 1680 cm^{-1}, and the $\varepsilon'(\omega)$ function for system V reaches the value of analogous dependence for system I at the frequency 3100 cm^{-1}.

The absorption of both argon and nitrogen by aqueous disperse system leads to an increase in the coefficient α of the absorption of IR radiation at the frequency $\omega > 1200$ cm^{-1}. As the concentration of absorbing gases rises twofold, coefficient α markedly decreases in this frequency range in the case of nitrogen absorption and slightly decreases upon the absorption of argon. Coefficient R of the reflection of plane monochromatic wave by aqueous disperse systems containing argon and nitrogen behave mainly in a similar manner depending on the frequency of the incident wave. However, on average, coefficient R of systems containing argon is by 26% larger than the reflection coefficient of corresponding systems that absorbed nitrogen. Besides, when the second molecule (atom) is added to each cluster, the integral intensity of the reflection of IR radiation increases in the case of argon and decreases in the case of nitrogen.

The rate of stored energy dissipation significantly rises after the capture of the impurity (argon or nitrogen) by the disperse system. When there is one argon atom per each cluster of the disperse system, the radiation becomes the most powerful. Twofold increase in the impurity concentration in a system lowers the average rate of energy dissipation by 2.7 and 1.5 times in the presence of argon and nitrogen in a system, respectively. The size of clusters that are most active to the emission of photons also depends on the type of impurity. In the case of argon, the most powerful energy emission is observed for $Ar(H_2O)_{40}$ and $(Ar)_2(H_2O)_{40}$ aggregates; among nitrogen-containing clusters, for $N_2(H_2O)_{50}$ and $(N_2)_2(H_2O)_{20}$ aggregates.

When aqueous disperse system absorbs argon so that one argon atom falls on each cluster, in general, the number of electrons interacting with outer electromagnetic radiation significantly (by 6 times) increases. This is not observed either upon the absorption of argon atoms (in twofold larger amount) or nitrogen by the disperse system.

CONCLUSIONS

The content of argon in the Earth's crust and atmosphere is increasing. Argon "is exhaled" by the lithosphere owing to volcanic processes, weathering, and recrystallization of rocks as well as by water sources. Argon has almost the same solubility in water as oxygen and is dissolved twice as faster as nitrogen. If the atmosphere had contained the same amount of argon as nitrogen, larger amount (compared to nitrogen) would be dissolved in rain droplets.

It was shown in this work that argon, being one of the most abundant atmospheric gases, could be present, along with nitrogen and oxygen, in atmospheric water clusters. Moreover, the behavior of impurity in aqueous aggregates is strictly individual and is manifested in spectral characteristics. Argon atoms adsorbed by water clusters are situated either on the surface or near the cluster surface. Such a position of Ar atoms considerably changes the spectra of atomic intramolecular vibrations in clusters. This leads to a significant increase in the intensity of IR absorption spectra in a frequency range determined by intramolecular vibrations. The argon atom more often collides with water molecules in the cluster than it is observed for the N_2 molecule. Atoms of hydrogen and oxygen in water molecules appeared to be more excited in the presence of argon atoms in clusters, as compared with the presence of N_2 molecules. Correspondingly, the power of radiation generated upon the dissipation of thermal energy by the clusters increases to a larger extent in the case, when the impurity is represented by argon, and to a lesser extent, when nitrogen molecules are present in clusters.

Thus, atmospheric absorption of argon by water clusters leads to a noticeable enhancement of the absorption of IR radiation by present moisture at frequencies $\omega >$ 1050 cm^{-1} and slightly changes the IR absorption spectrum at low frequencies ($\omega < 1050$ cm^{-1}). In general, the capture of argon from the atmosphere by the aqueous disperse medium should enhance the greenhouse effect.

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