Computational investigation of dielectric absorption at microwave frequencies in binary oxides

Eric J. Wu and Gerbrand Ceder a) 
Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

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In this work, a computational study of the dielectric absorption and Q-factor at microwave frequencies in rocksalt oxides is presented. This work is performed within a theory for the two-phonon anharmonic absorption process in terms of third-order force constant matrices, phonon eigendata, and Born effective charges. The complex dielectric permittivity is expressed by means of Green’s functions. This theory is used on model systems that are described using empirical Buckingham potentials with shells. The Q-factor and its temperature dependence in AO (A=Mg, Ca, Ba, Sr) oxides with the rocksalt structure is calculated. For MgO, the calculated Q-factor at room temperature agrees relatively well with experiments, and the temperature dependence is in qualitative agreement. The Q-factor in a model ‘‘AO’’ system is also calculated to determine the effects of cation mass and size on the Q-factor. Increased cation size is found to lower the Q-factor, whereas increasing cation mass is found to increase the Q-factor when the cation mass is higher than that of Ca. © 2001 American Institute of Physics. [DOI: 10.1063/1.1360223]

INTRODUCTION

Ceramic dielectric materials are widely studied because of their use as resonators, oscillators, and filters in microwave communications devices such as cellular phones and satellite communications equipment.1–5 For these communications devices to possess high frequency selectivity and low noise, the dielectric resonators must possess a high Q-factor, or quality factor, in the frequency regime of interest. Traditionally this has been at microwave frequencies, but as bandwidth becomes more scarce, the investigation of ceramics with a high Q-factor in the millimeter regime has become important as well.

The Q-factor is a measure of the dielectric losses in a ceramic and is defined by

\[ Q(\omega) = \frac{\varepsilon'(\omega)}{\varepsilon''(\omega)}, \]  

where \( \varepsilon'(\omega) \) and \( \varepsilon''(\omega) \) are, respectively, the frequency-dependent real and imaginary parts of the dielectric constant. Dielectric losses can be classified according to two types of mechanisms. The first loss mechanism is attributable to anharmonic forces within the crystal and is present even in perfect crystals. The second type can be attributed to defects such as vacancies, sample porosity, impurity phases, and grain boundaries. Some of these defects, such as vacancies and isotopic impurities, are present in equilibrium at nonzero temperatures.

To date, the search for high Q-factor ceramics has mostly been experimental in nature. One of the difficulties in interpreting experimental studies lies in the fact that observed losses can be a combination of losses attributable to many effects. For example, there is some disagreement as to whether the increased Q-factor of Ba(Mg1/3Ta2/3)O3 upon annealing is because the Mg and Ta cations order upon annealing, or because defects have been annealed out.\(^6,7\) Computational investigations of the Q-factor in oxides can be useful to help resolve these conflicts because of control over conditions. With computational modeling, one can isolate the effects of substitutional disorder, atomic mass, vacancies, atomic size, temperature, and so forth. For instance, one can investigate the effect of changing chemical composition without the effects of defects that may be caused by the change in chemical composition. The insight gained in this manner can be applied to suggest material improvements.

Although most of the work investigating losses in ceramics has been experimental there are some noteworthy theoretical studies. Silverman derived equations to show how losses attributable to defects as well as third- and fourth-order anharmonicity could explain the temperature dependence of experimental data in SrTiO\(_3\).\(^8\) Balagradov et al. and Mickiewicz performed similar analyses and applied it to ferroelectrics and alkali halides, respectively.\(^9,10\) Schlömann studied dielectric losses as a function of charge disorder.\(^11\) Meng et al. have studied temperature and frequency dependence of dielectric losses in NaCl owing to defects using approximate models.\(^12\) Gurevich and Tagantsev have examined the temperature and frequency dependence of dielectric losses for different crystal symmetries.\(^13–18\) Finally, it is worth noting that it is also possible to obtain the frequency-dependent imaginary part of the dielectric function using molecular dynamics simulation techniques.\(^19\) This method uses a form of the fluctuation-dissipation theorem to calculate the imaginary part of the dielectric constant from current-current density correlation functions. The real part of the dielectric function is then obtained through Kramers–Kronig relations.\(^20\) However, extremely long simulation
times and noise in the data make this method impractical for calculating the $Q$-factor at microwave frequencies.

The remainder of the article is organized as follows. First, the methodology which we use to calculate the $Q$-factor is described. This methodology is used to calculate the $Q$-factors for both real and model systems. The $Q$-factor for AO (A=Mg, Ca, Sr, Ba) oxides with the rocksalt structure is calculated, as is the temperature dependence of the $Q$-factor for MgO. Then, a systematic study of how the $Q$-factor in rocksalt oxides changes as cation mass and size are changed is performed.

THEORETICAL

In the present study, we focus on losses attributable to anharmonicity in perfect crystals, and do not consider losses attributable to defects. At microwave or millimeter frequencies, the ceramic dielectric absorbs energy from the electrical field by multi-phonon processes rather than by resonant single-phonon processes. The reason for this is that at microwave frequencies, the energy and momentum conservation requirements for single-phonon processes cannot be satisfied owing to the field frequency being much lower than the frequencies of the optical phonon modes. However, two-phonon processes can satisfy the energy and momentum conservation requirements

\begin{equation}
\Omega_{\text{field}} = \omega_{\text{phonon}},
\end{equation}

\begin{equation}
k_{\text{field}} = k_{\text{phonon}} \approx 0,
\end{equation}

Higher order absorption processes are also possible, though they will not be considered in the present work.

For two-phonon processes, an expression for the complex frequency-dependent dielectric constant has been derived by Cowley and Vinogradov, which we briefly outline here.\textsuperscript{22,23} The Hamiltonian of a crystal in an electric field can be written in terms of phonon creation and destruction operators and has the form\textsuperscript{22,23}

\begin{equation}
H = H_0 + H_A + H_E,
\end{equation}

where $H_0$ is the harmonic part and is written as

\begin{equation}
H_0 = \hbar \omega_a a^*_a a_a.
\end{equation}

$H_A$ is the anharmonic term in the crystal Hamiltonian truncated to third order and is written

\begin{equation}
H_A = \sum_{q,j_1,j_2} V^3(0; q; j_1; q; j_2)(a^*_a a_j)(a^*_a a_j) \\
\times(a^*_a a_j),
\end{equation}

where the $a$'s are phonon creation and destruction operators, the $q$'s are points in the first Brillouin zone, $j$'s are phonon branches, and $V^3$ is the Fourier transformed third order force constant matrix as defined in Born and Huang.\textsuperscript{23}

$H_E$ is attributable to the electrical field and is expressed as

\begin{equation}
H_E = -\mathbf{E} \cdot \mathbf{M},
\end{equation}

where $\mathbf{E}$ is the electrical field and $\mathbf{M}$ is the dipole moment. The dipole moment can be expanded in the same way as the crystal Hamiltonian

\begin{equation}
M_a = \sum_{k,j} M^1_a(k,j)(a^*_0 + a_0) \\
+ \sum_{k_1,k_2,j_1} M^2_{a,b}(k_1,j_1;k_2,j_2)(a^*_1 a_{k_1,j_1} + a_{k_1,j_1}) \\
\times(a^*_2 a_{k_2,j_2} + a_{k_2,j_2}) + \ldots,
\end{equation}

where the zeroth order term vanishes for crystals without spontaneous polarization. To obtain an expression for the frequency-dependent dielectric constant, the Heisenberg equation for the density matrix

\begin{equation}
\frac{d\rho}{dt} = [H,\rho]
\end{equation}

is solved using a perturbative Green’s functions approach,\textsuperscript{21,22,24} with

\begin{equation}
\rho = \exp(-H/kT) / \text{Tr} \exp(-H/kT),
\end{equation}

The final expression for the dielectric constant for a centrosymmetric crystal, taking only the linear term in the dipole moment expansion, keeping within the third order expansion of the crystal Hamiltonian, ignoring small anharmonic shifts in phonon frequency, and ignoring electronic dielectric losses is

\begin{equation}
e_{ab}(\omega) = e_{ab}(\infty) \\
+ \sum_{j=1}^{\text{TO modes}} \frac{M^\omega_{ab}(0,j)M^\omega_{ab}(0,j)}{\omega(0)^2 - \omega^2 - 2\omega(0)[i\Gamma(0,j,\omega)]},
\end{equation}

where the sum is taken over transverse optical phonon modes. $\Gamma(0,j,\omega)$ is the inverse phonon lifetime for the $j$th branch and is given by

\begin{equation}
\Gamma(0,j,\omega) = 18\pi/\hbar^2 \sum_{q,j_2} |V(q;j,j_1; -q;j_2)|^2 \\
\times\{n_1(q;j_1) - n_1(-q + \omega;j_2) \\
\times\{\delta(\omega - \omega_1(q;j_1)) - \omega_2(-q;j_2) \\
- \delta(\omega - \omega_1(q;j_1) + \omega_2(-q;j_2))\},
\end{equation}

where we have accounted for the effect of phonon lifetimes by the method of Sparks et al.\textsuperscript{25} In these expressions, $n(q,j)$ is the Planck distribution for the $q$th mode and $j$th branch. $M^\omega_{ab}$ is given by

\begin{equation}
M^\omega_{ab} = \sum_{\kappa,\rho} Z^\omega_{ab}\kappa,\rho / \sqrt{m_\kappa},
\end{equation}

where $\kappa$ sums over the atoms in the cell, $e_{\kappa}$ is the eigenvector for the $\kappa$th atom $j$th branch at $q=0$, $m_\kappa$ is the mass of atom $\kappa$, and $Z^\omega_{ab}$ is the Born effective charge tensor. These
expressions have been used to calculate phonon lifetimes for the alkali halides, as well as physical properties such as dielectric constants, optical coefficients, refractive indexes, and extinction coefficients.\textsuperscript{21,26–34}

The final expression for $Q(\omega)$ is

$$Q_{\alpha\beta}(\omega) = \sum_{\text{modes}} \frac{(\omega(0j)^2 - \omega^2)}{\left(\omega(0j)^2 - \omega^2\right)^2 + 4\omega^2(0j)^2 \Gamma^2(0j)}.$$ \hspace{1cm} (19)

In our study, we investigate compounds with the rocksalt structure with cubic symmetry, thus the diagonal components of the $Q_{\alpha\beta}(\omega)$ tensor are equal. Since the equations are somewhat cumbersome, it worth noting exactly what is needed for a calculation of the $Q$-factor within this model. To calculate the $Q$-factor, one needs the full phonon dispersion curve of a material with eigenvectors of each mode, the third order force constant matrices, Born effective charges, and the high-frequency dielectric constant.

**IMPLEMENTATION**

The $Q$-factor calculation can be implemented within any energy method, and we have chosen to implement our calculations within the empirical Buckingham pair-potential method. This method has been extensively used to study ionic interactions in oxides with relatively good success.\textsuperscript{35–38} The potential includes Coulomb and short-range terms, and the shell model is used to include electronic polarization effects.\textsuperscript{39} The short-range repulsive term is of the Buckingham type and takes the form

$$V(r) = A \exp \left( \frac{-r}{\rho} \right) - C \frac{1}{r^6},$$ \hspace{1cm} (17)

where $r$ is the distance between ions, and $A$, $\rho$, and $C$ are the potential parameters. Potential parameters for the alkaline-earth oxides were taken from Lewis and Catlow, who derived the potentials by performing an empirical fit to reproduce the experimental crystal structure, transverse optical phonon frequencies, high-frequency dielectric constants, and elastic constants.\textsuperscript{40} Potential parameters used in the calculations are listed in Table I.

<table>
<thead>
<tr>
<th>Ion pair</th>
<th>$\rho$(Å)</th>
<th>$C$(eV/Å$^2$)</th>
<th>$Y_+/e$</th>
<th>$k_+$ (eV/Å$^2$)</th>
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</thead>
<tbody>
<tr>
<td>Mg–O</td>
<td>1428.5</td>
<td>0.2945</td>
<td>0</td>
<td>1.585</td>
</tr>
<tr>
<td>Ca–O</td>
<td>1090.4</td>
<td>0.3437</td>
<td>0</td>
<td>3.135</td>
</tr>
<tr>
<td>Sr–O</td>
<td>959.1</td>
<td>0.3721</td>
<td>0</td>
<td>3.251</td>
</tr>
<tr>
<td>Ba–O</td>
<td>905.7</td>
<td>0.3976</td>
<td>0</td>
<td>9.203</td>
</tr>
<tr>
<td>O–O</td>
<td>22764</td>
<td>0.149</td>
<td>27.879</td>
<td>...</td>
</tr>
</tbody>
</table>

Table I. Potential parameters used in the calculations. $A$, $\rho$, and $C$ are potential parameters, $Y_+$ is the charge of the cation shell, and $k_+$ is the harmonic spring constant coupling the shell and core of the cation. Oxygen shell charges were taken to be $-3.0$, oxygen spring constants were determined individually for each compound using the method described by Lewis and Catlow (see Ref. 40).

data for all compounds were calculated for 166,375 (55 x55 x55 mesh) $q$ points in the first Brillouin zone with the GULP computer program.\textsuperscript{41} Third order force constant matrices for the repulsive terms were calculated numerically using the supercell method as described by Wei and Chou.\textsuperscript{42,43} The force constants were restricted to the first-nearest neighbor. Phonon inverse lifetimes were then calculated by performing the summation over $q$ points in Eq. (14).

To evaluate Eq. (14), we first note that, because of their finite lifetimes, the energy of phonons are defined only to within approximately the inverse of their thermal lifetime.\textsuperscript{34} Thus, the energy conservation requirement

$$\Omega_{\text{field}} = \Omega_{\text{phonon1}} - \Omega_{\text{phonon2}} \quad \text{[Eq. (4)]},$$

which is represented mathematically in Eq. (14) by a delta function, is not strictly correct. To account for this fact, the delta function in Eq. (14) is replaced with a Lorentzian of the form\textsuperscript{13,25,34}

$$\Gamma_{\text{avg}} + (\Omega - \omega_1 + \omega_2),$$ \hspace{1cm} (18)

where $\Gamma_{\text{avg}}$ is the inverse of the average phonon lifetime of the phonons participating in the absorption process, $\Omega$ is the frequency of the incoming radiation, and $\omega_1$ and $\omega_2$ are the energies of the two phonons participating in the absorption process. An estimate for the parameter $\Gamma_{\text{avg}}$ was chosen by the following procedure: The phonon inverse lifetime for the transverse optical mode at $q = 0$ was calculated using Eq. (14), and ten $q$ points that made the greatest contributions to Eq. (14) were noted. These inverse lifetimes at these $q$ points were calculated and the $\Gamma_{\text{avg}}$ parameter was adjusted until the difference between $\Gamma_{\text{avg}}$ and the calculated phonon inverse lifetimes at these points was as small as possible. In this way, we have used importance sampling to obtain a self-consistent estimate for the average phonon inverse lifetime of the phonons. The range of $\Gamma_{\text{avg}}$ estimated was between 0.03 and 0.15 THz for all calculations.

Once inverse phonon lifetimes were calculated for transverse optical phonons, the frequency-dependent $Q$-factor was then calculated using Eq. (16). Born effective charges used in this calculation were taken from the first-principles calculations of Posternak et al.\textsuperscript{44} The incoming radiation was assumed to come from the [100] direction in all calculations.

The $Q$-factor was calculated for a variety of simple oxide systems. First, the $Q$-factor for AO oxides ($A$=Mg, Ca, Sr, Ba) was calculated at 8 GHz and 300 K, using the poten...
tial parameters of Lewis and Catlow.\textsuperscript{40} Next, we calculated the $Q$-factor for MgO at 8 GHz as a function of temperature. In this calculation, since the average phonon lifetime at 0 K is infinite (the inverse lifetime is 0), we assume a linear form for $\Gamma_{\text{avg}}$ and scale $\Gamma_{\text{avg}}$ accordingly in Eq. (16).\textsuperscript{34} Next we calculate the $Q$-factor for a model “AO” system. In these calculations, we use the potential parameters of MgO from Lewis and Catlow as a base. Then, we vary the mass of the “A” cation to see the effect of changing mass on the $Q$-factor. Last, we vary the “size” of the A cation by varying the $\alpha$ and $\rho$ parameters of the cation-anion interaction. These model system calculations allow us to detect trends in how the $Q$-factor changes in rocksalt oxides when the mass or size of one of the cations is changed.

RESULTS AND DISCUSSION

The calculation of the $Q$-factor relies on four quantities: the phonon dispersion curves, the third order force constant matrices, Born effective charges, and the high-frequency dielectric constants. To obtain an idea of the sources and magnitudes of errors in our calculations, we have compared the calculated values of phonon dispersion curves, Born effective charges, and high-frequency dielectric constants with experiments for the alkaline-earth oxides AO ($A=$Mg, Ca, Sr, Ba). Dielectric data and Born effective charges are listed in Table II, and calculated and experimental phonon dispersion curves for the alkaline-earth oxides are shown in Figs. 1–4. High frequency dielectric constants and Born effective charges agree reasonably well with experiments. Calculated phonon dispersion curves agree well with experimental curves for MgO and CaO, slightly less well for SrO and BaO. We are unable to obtain an estimate of the errors in the third-order force constant matrices, thus it is possible that this quantity is where the greatest source of errors lie.

We investigated the effect of chemistry on the $Q$-factor for rocksalt oxides. Table III shows the calculated $Q$-factor for four AO ($A=$Mg, Ca, Sr, Ba) alkaline-earth oxides. Only the $Q$-factor for MgO has been measured experimentally. The other AO ($A=$Ca, Sr, Ba) compounds are hygroscopic, which makes sample preparation to measure the $Q$-factor difficult. However, our calculations indicate that, within our calculation errors, the other AO oxides may not be much better than MgO for use as high $Q$-factor materials.

At 8 GHz, the calculated $Q$-factor for MgO is 246 000 compared to the experimental value of 250 000 obtained by Mage \textit{et al.}\textsuperscript{45} The agreement with experiments is very good, however, it may be somewhat fortuitous given that we have considered neither losses owing to fourth-order anharmonic nor losses owing to defects. The neglect of other absorption mechanisms should make our calculated $Q$-factor be greater than the experimental value. As previously discussed, the use of empirical potentials as the energy model rather than more accurate first-principles methods leads to uncertainties in the third-order force constant matrices, as well as some uncertainty in phonon eigendata. These facts should lead to caution in interpreting the results.

The temperature dependence on the $Q$-factor has been studied for MgO. The normalized temperature dependence for MgO is shown in Fig. 5. In these calculations, we assume the effect of thermal expansion on the crystal structure is negligible. We have normalized both experimental and calculated $Q$-factors to 1 at room temperature to examine the

<table>
<thead>
<tr>
<th>Material</th>
<th>$\varepsilon(\infty)$ calc.</th>
<th>$\varepsilon(\infty)$ exp.</th>
<th>$Z_0^2$ calc.</th>
<th>$Z_0^2$ exp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>MgO</td>
<td>3.02</td>
<td>2.96</td>
<td>−1.98</td>
<td>−1.96/−2.02</td>
</tr>
<tr>
<td>CaO</td>
<td>3.36</td>
<td>3.35</td>
<td>−2.35</td>
<td>−2.26/−2.30</td>
</tr>
<tr>
<td>SrO</td>
<td>3.48</td>
<td>3.46</td>
<td>−2.44</td>
<td>−2.34/−2.47</td>
</tr>
<tr>
<td>BaO</td>
<td>3.91</td>
<td>3.61</td>
<td>−2.72</td>
<td>−2.69/−2.97</td>
</tr>
</tbody>
</table>

FIG. 1. Phonon dispersion curve for MgO. Lines are calculated using potentials of Lewis and Catlow (Ref. 40), circles are experimental points from Sangster \textit{et al.} (Ref. 47).

FIG. 2. Phonon dispersion curve for CaO. Lines are calculated using potentials of Lewis and Catlow (Ref. 40), circles are experimental points from Saunderson and Peckham (Ref. 49).
qualitative dependence on the $Q$-factor as a function of temperature. The calculated and experimental data agree roughly to within experimental error, except at extremely low temperatures\(^4\) where losses owing to defects dominate the experimental data.

Because the energy model used here has potential parameters with a simple physical interpretation, the effects of electronic polarizability, "size," and mass on the $Q$-factors, can be investigated with controlled changes in these parameters. Table III shows the calculated $Q$-factors for AO oxides \((\text{A}=\text{Mg, Ca, Sr, Ba})\) after the shell terms representing electronic polarizability have been removed from the energy description. This was done to examine the effect of electronic terms in the energy model. This changes both the high-frequency dielectric constants and also the phonon dispersion curves. Removing the shell terms seems to lower the calculated $Q$-factor by about 10%–20%. This is likely attributable to the fact that the electronic part of the real dielectric constant is lowered by removing shell terms in the energy model.

Figure 6 shows the $Q$-factor as a function of mass of the M cation. In these calculations, we have constrained the interatomic potentials to be those of MgO, while changing the mass of the cation. In this way, we can explore the effect of cation mass on the $Q$-factor. Increasing the cation mass lowers the frequencies of the acoustical phonon branches as the mass difference between the cation and oxygen increases. In addition, the transverse optical phonon frequencies are lowered somewhat. Our results indicate that as the mass difference between the cations increases, the $Q$-factor rises after a "cutoff" when the mass is greater than that of Ca. This can be understood in terms of phonon lifetimes and transverse optical mode frequencies, which appear in the expression for the $Q$-factor [Eq. (16)]. The frequency of the transverse optical mode at $q=0$ is lowered as the cation mass is increased from Be to Ba, from 16 to 10 THz. This has the effect of lowering the $Q$-factor as can be seen in Eq. (16). However, as the cation mass is increased from Ca to Sr, the phonon dispersion curve develops a gap between the acoustic and optical phonon branches. Thus, two-phonon processes involving optical and acoustical phonon branches are rare, since they do not satisfy the energy conservation requirement.

![SrO phonon dispersion](image)

**FIG. 3.** Phonon dispersion curve for SrO. Lines are calculated using potentials of Lewis and Catlow (Ref. 40), circles are experimental points from Rieder et al. (Ref. 50).

![BaO phonon dispersion](image)

**FIG. 4.** Phonon dispersion curve for BaO. Lines are calculated using potentials of Lewis and Catlow (Ref. 40), circles are experimental points from Chang et al. (Ref. 51).

![Normalized dielectric loss](image)

**FIG. 5.** Calculated and experimental temperature dependence on the $Q$-factor for MgO. Calculations performed at 8 GHz. Solid line is calculated, dotted line is experimental data from Mage et al. (Ref. 45).

<table>
<thead>
<tr>
<th>Shell model</th>
<th>No shells</th>
<th>Experimental</th>
</tr>
</thead>
<tbody>
<tr>
<td>MgO</td>
<td>246 000</td>
<td>201 000</td>
</tr>
<tr>
<td>CaO</td>
<td>33 000</td>
<td>29 000</td>
</tr>
<tr>
<td>SrO</td>
<td>315 000</td>
<td>256 000</td>
</tr>
<tr>
<td>BaO</td>
<td>108 000</td>
<td>97 000</td>
</tr>
</tbody>
</table>

**TABLE III.** Calculated and experimental $Q$-factors at 8 GHz and 300 K. Experimental data from Mage et al. (Ref. 45).
of Eq. (14). This results in longer phonon lifetimes, and lower dielectric losses. These competing effects explain the trends in the $Q$-factor in Fig. 6.

Next, we explore the effect of changing the effective size of the cation by varying the $A$ and $\rho$ parameters of the potential. In these calculations, we constrain the mass of the cation to be that of Mg, but change the interatomic potential parameters to those of CaO, SrO, and BaO. By increasing size in this way we can explore the effect of atomic size differences on the $Q$-factor. Figure 7 shows the $Q$-factor as a function of changing the size of the A cation. Our results show that as the size difference between the two ions increases, the $Q$-factor decreases. Increasing the size of the cation has two effects: one is to increase the strength of the third-order force constant matrices, and the other is to reduce the frequencies of all points in the phonon dispersion curves. As discussed before, lowering the phonon frequencies of the transverse optical modes will lower the $Q$-factor. In addition, since losses occur through the anharmonic mechanism, increasing anharmonic force constants will increase dielectric losses. Both of these effects work to lower the $Q$-factor as cation size is increased.

CONCLUSIONS

The problem of dielectric losses in microwave ceramics is a technologically important one, yet one in which the physics are not well understood. We have calculated the $Q$-factor based on a Green’s functions formalism based on third-order anharmonic effects. We have implemented this method to calculate the $Q$-factor for both real and model rocksalt oxides. We have calculated the $Q$-factor for AO (A = Mg, Ca, Sr, Ba) oxides. The $Q$-factor for MgO agrees well with experiments. The temperature dependence of the calculated $Q$-factor for MgO is qualitatively similar to experiments. We have found in our model AO system that as the mass difference between ions increases, the $Q$-factor first decreases as the cation mass is increased, then increases when the cation mass is greater than that of Ca. In our model system, we also find that as the size difference between ions increases, the $Q$-factor decreases. These changes have been explained in terms of the effects that mass and size have on phonon dispersion curves and anharmonic force constants.

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