Modelling inter- and transgranular fracture in piezoelectric polycrystals

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\section*{A B S T R A C T}
A cohesive zone finite element model for quasi-static fracture of piezoelectric polycrystals is proposed. Interface elements are used to model both inter- and transgranular fracture. Electromechanical constitutive relations are derived by enhancing commonly used mechanical traction-opening laws with relations for a parallel plate capacitor. Numerical simulations demonstrate that the proposed model correctly mimics several experimentally observed phenomena. Most importantly the switch from mainly intergranular to mainly transgranular fracture with increasing grain size is modelled correctly. The model also correctly mimics the influence of an electric field on the ultimate load.

\section*{1. Introduction}
Piezoelectric ceramics are materials that exhibit relatively large deformations through application of an electric field and vice versa. This electromechanical coupling makes piezoelectrics suitable for many applications, such as MEMS (micro electromechanical systems). The brittleness of piezoelectric ceramics makes the materials, however, sensitive to damage. An accurate and robust numerical model to assess this damage is an indispensable tool for increasing the reliability of piezoelectric systems.

Many theoretical studies of fracture in piezoelectric ceramics have been conducted [1–3]. In these theoretical studies the use of either impermeable (no surface charges on crack sides) or permeable (continuity requirements for the electric field and electric flux density over a crack) boundary conditions is discussed extensively. Various studies [4,5] demonstrated that the impermeable condition is less appropriate for modelling cracks in piezoelectric ceramics than the permeable crack assumption. The determination of a failure criterion for piezoelectric ceramics that correctly mimics the influence of an electric field has also been addressed frequently. The fracture criterion proposed in Ref. [6] is demonstrated to be in good agreement with experimental observations. Further enhancements of this fracture criterion by taking into account material nonlinearities have been considered [7,8]. A cohesive zone model for simulating fatigue has been proposed in [9].

In this contribution quasi-static fracture of a piezoelectric polycrystal is studied using a finite element model. The fracture process is described by a cohesive zone model. The novelty of this work is the use of a cohesive zone approach to model crack nucleation and propagation in a finite element model for the piezoelectric polycrystalline microstructure. In the considered cohesive zone model the crack evolves from being permeable at the moment of opening to impermeable when fully opened. The influence of an electric field as well as some material nonlinearities are incorporated in the model. On the one hand the proposed finite element model is developed to gain more insight in the micro-scale phenomena that lead to macro-scale fracture. On the other hand, modelling of fracture of a piezoelectric polycrystal might be applied directly in the case of MEMS, since these systems can be of the same order of magnitude as the piezoelectric grains.
In Section 2 the numerical treatment of the equilibrium equations for piezoelectric materials is discussed. The constitutive relations, required for determination of the solution of these equations, are then elaborated in Section 3. Some algorithmic aspects are discussed in Section 4 after which numerical experiments are performed in Section 5 to demonstrate the applicability of the proposed method. Finally, some conclusions are drawn in Section 6.

2. Discretisation of the electromechanical equilibrium equations

In this section the finite element method [10,11] is used to discretise the mechanical and electrical equilibrium equations. The grains are discretised using $C^0$-continuous finite elements. The grain boundaries and transgranular cracks are discretised by means of zero thickness interface elements. Based on the assumption of quasi-static behaviour, a coupled electromechanical system of equations is formulated.

2.1. Mechanical equilibrium equations

Consider a domain $\Omega$, with boundary $\Gamma$ and a predefined crack indicated by $\Gamma_c$ (Fig. 1). In the absence of body forces, the quasi-static mechanical equilibrium for any point in this domain is given by

$$\text{div}(\sigma) = 0,$$

(1)
where $\sigma$ is the Cauchy stress tensor. The body $\Omega$ is subjected to the boundary conditions as indicated in Fig. 1 such that

\begin{align*}
  \mathbf{t} &= \tilde{\mathbf{t}} \quad \text{on } \Gamma_t, \\
  \mathbf{u} &= \tilde{\mathbf{u}} \quad \text{on } \Gamma_u,
\end{align*}

where $\Gamma_t$ and $\Gamma_u$ are the parts of the boundary subjected to tractions and displacements, respectively. The weak form of the mechanical equilibrium equation can be derived by multiplication of (1) with a kinematically admissible displacement increment $\Delta \mathbf{u}$ (with $\Delta \mathbf{u} = 0$ on $\Gamma_u$). Integrating over the considered domain then yields

\begin{equation}
  \int_{\Omega} \text{div}(\sigma) \cdot \Delta \mathbf{u} \, d\Omega = 0.
\end{equation}

Using divergence and Gauss' theorem in combination with the symmetry of the Cauchy stress tensor, this equation can be rewritten as

\begin{equation}
  \int_{\Omega} \mathbf{t} \cdot \Delta \mathbf{u} \, d\Omega + \int_{\Gamma_c} \mathbf{t}_c \cdot \Delta \mathbf{u} \, dl = \int_{\Gamma_t} \tilde{\mathbf{t}} \cdot \Delta \mathbf{u} \, dl,
\end{equation}

where $\mathbf{t}_c$ is the traction over the crack. The symmetric gradient operator is denoted by $\text{grad}^s$ and $\mathbf{t}_c/C_1 = \mathbf{t}_c/C_0$ is used to indicate the discrete jump of a quantity over a crack. In this contribution linearised kinematics are assumed. This implies that the integrals in (4) are evaluated on the undeformed domain, where the volume of the crack is equal to zero. Furthermore note that the linearised kinematics $(\Gamma_c \equiv \Gamma_c^+ \equiv \Gamma_c^-)$ in combination with the traction equilibrium over the crack allows for description of the virtual work performed by the crack by an integral over $\Gamma_c$ only (e.g. [12]).

The weak form of the equilibrium Eq. (4) can be discretised using $C^0$-continuous finite elements for which it holds that

\begin{equation}
  \mathbf{u} = \mathbf{N}^n \mathbf{a}^n, \quad \mathbf{\varepsilon} = \mathbf{B}^n \mathbf{a}^m \quad \text{and} \quad [\mathbf{u}] = \mathbf{M}^n \mathbf{a}^m.
\end{equation}

In these equations $\mathbf{a}^n$ is the nodal displacement vector of size $n^m$, where the superscript “m” indicates that a mechanical quantity is considered. The matrices $\mathbf{N}^n$, $\mathbf{B}^n$ and $\mathbf{M}^n$ map the discrete displacement vector onto the displacements, engineering strains and crack openings, respectively. Note that since $C^0$-continuous finite elements are used, strains and stresses are in general discontinuous over element edges. Consequently, the traction distribution over a crack surface is generally also discontinuous. Substitution of these expressions in Eq. (4) then yields

\begin{equation}
  \delta \mathbf{a}^m \cdot \int_{\Omega} \mathbf{B}^m \sigma d\Omega + \delta \mathbf{a}^m \cdot \int_{\Gamma_c} \mathbf{M}^m \mathbf{t}_c d\Gamma = \delta \mathbf{a}^m \cdot \int_{\Gamma_t} \tilde{\mathbf{t}} d\Gamma, \quad \forall \delta \mathbf{a}^m \in \mathbb{R}^{m^2},
\end{equation}

where Voigt notation is used for the second-order stress and strain tensors $\sigma$ and $\mathbf{\varepsilon}$ and where the dot-product is defined as $\mathbf{a} \cdot \mathbf{b} = \mathbf{a}^T \mathbf{b}^T$. Since this equation should hold for any kinematically admissible displacement increment, the discrete mechanical equilibrium equations can be formulated as

\begin{equation}
  \mathbf{f}^m_{\text{int}} = \mathbf{f}^m_{\text{ext}},
\end{equation}

where the internal and external force vector are given by

\begin{equation}
  \mathbf{f}^m_{\text{int}} = \int_{\Omega} \mathbf{B}^m \sigma d\Omega + \int_{\Gamma_c} \mathbf{M}^m \mathbf{t}_c d\Gamma \quad \text{and} \quad \mathbf{f}^m_{\text{ext}} = \int_{\Gamma_t} \mathbf{M}^m \tilde{\mathbf{t}} d\Gamma,
\end{equation}

respectively. Note that the internal force vector requires a constitutive law for both the Cauchy stress $\sigma$ and the traction acting on the crack $\mathbf{t}_c$. The former is given by a stress–stress relation, whereas the latter is obtained using a traction-opening relation. Both constitutive relations are discussed in Section 3.
2.2. Electric equilibrium equations

In analogy with the mechanical displacement field, the electric field also needs to be in equilibrium. In the absence of body charges, this electrostatic equilibrium is represented by Gauss’ law as

$$\text{div}(\mathbf{D}) = 0,$$  \hspace{1cm} (9)

with $\mathbf{D}$ being the electric flux density. The corresponding electrical boundary conditions as indicated in Fig. 1 can be written as

$$q = \tilde{q} \quad \text{on } \Gamma_q,$$
$$\Phi = \tilde{\Phi} \quad \text{on } \Gamma_\Phi,$$  \hspace{1cm} (10)

where $\Gamma_q$ and $\Gamma_\Phi$ are the parts of the boundary subjected to surfaces charges and electric potentials, respectively. As in the case of mechanical equilibrium, the weak form of Eq. (9) is obtained by multiplying this equation with a kinematically admissible electric potential field increment $\delta \Phi$ and integrating over the domain $\Omega$ to yield

$$\int_{\Omega} \text{div}(\mathbf{D}) \delta \Phi d\Omega = 0.$$  \hspace{1cm} (11)

Application of Gauss’ theorem then gives

$$\int_{\Omega} \mathbf{D} \cdot \text{grad} \delta \Phi d\Omega - \int_{\Gamma_q} q \delta \Phi d\Gamma = - \int_{\Gamma_q} \tilde{q} \delta \Phi d\Gamma,$$  \hspace{1cm} (12)

where the surface charge density $q$ on a surface with outward normal $\mathbf{n}$ is defined as

$$q = -\mathbf{D} \cdot \mathbf{n}.$$  \hspace{1cm} (13)

The discretisation of Eq. (12) is performed using

$$\Phi = \mathbf{N}^e \mathbf{a}^e, \quad \mathbf{E} = \mathbf{B}^e \mathbf{a}^e \quad \text{and} \quad [\mathbf{f}^i] = \mathbf{M}^e \mathbf{a}^e.$$  \hspace{1cm} (14)

In these equations $\mathbf{a}^e$ is the nodal electric potential vector of size $n_e$, where the superscript “e” indicates that a electrical quantity is considered. Note that, as a consequence of the $C^0$-continuous finite element discretisation, the electric field $\mathbf{E}$ and electric flux density $\mathbf{D}$ are in general discontinuous over element edges. The surface charge density on a crack surface is therefore in general also discontinuous over element edges. Substitution of these expressions in Eq. (12) then yields

$$-\delta \mathbf{a}^e \cdot \int_{\Omega} \mathbf{B}^e \mathbf{D} d\Omega - \delta \mathbf{a}^e \cdot \int_{\Gamma_c} \mathbf{q}_e \mathbf{M}^e \mathbf{d} d\Gamma = -\delta \mathbf{a}^e \cdot \int_{\Gamma_q} \tilde{q} \mathbf{N}^e \mathbf{d} d\Gamma, \quad \forall \delta \mathbf{a}^e \in \mathbb{R}^{n_e},$$  \hspace{1cm} (15)

where use is made of the definition of the electric field given by

$$\mathbf{E} = -\text{grad}(\Phi).$$  \hspace{1cm} (16)

Since Eq. (15) should hold for any kinematically admissible potential increment, the discrete electrical equilibrium equations can be formulated as

$$\mathbf{r}^i = \mathbf{r}^e,$$  \hspace{1cm} (17)

where the electrical internal and external force vectors are given by

$$\mathbf{r}^i = \int_{\Omega} \mathbf{B}^e \mathbf{D} d\Omega + \int_{\Gamma_c} \mathbf{q}_e \mathbf{M}^e \mathbf{d} d\Gamma \quad \text{and} \quad \mathbf{r}^e = \int_{\Gamma_q} \tilde{q} \mathbf{N}^e \mathbf{d} d\Gamma.$$  \hspace{1cm} (18)

Evaluation of the internal force vector requires constitutive relations for the electric flux density $\mathbf{D}$ and surface charge density on the crack side $q_e$ in terms of the electric field $\mathbf{E}$ and potential jump over the crack $[\Phi]$. Both relations are considered in Section 3.

2.3. Electromechanical equilibrium equations

The discrete equilibrium Eqs. (7) and (17) as derived in the previous sections are coupled via the constitutive laws. From an implementation point of view it is therefore convenient to formulate both systems as a combined electromechanical system of equations. This combined system is derived by defining the electromechanical state vector as

$$\mathbf{a} = \left( \begin{array}{c} \mathbf{a}^m \\ \mathbf{a}^e \end{array} \right)$$  \hspace{1cm} (19)

and corresponding electromechanical $\mathbf{N}$, $\mathbf{B}$ and $\mathbf{M}$ matrices as
\[
\mathbf{N} = \begin{pmatrix} \mathbf{N}_m \\ \mathbf{N}_e \end{pmatrix}, \quad \mathbf{M} = \begin{pmatrix} \mathbf{M}_m \\ \mathbf{M}_e \end{pmatrix}, \quad \text{and} \quad \mathbf{B} = \begin{pmatrix} \mathbf{B}_m \\ \mathbf{B}_e \end{pmatrix}.
\] (20)

Using these relations, the discrete electromechanical system of \( n \) (\( = n_m + n_e \)) equations can be formulated as
\[
\mathbf{f}_\text{int} = \mathbf{f}_\text{ext},
\] (21)

where the internal and external force vector are given by
\[
\mathbf{f}_\text{int} = \int_{\Omega} \mathbf{B}^T \mathbf{d} \Omega + \int_{\Gamma_e} \mathbf{M}^T \mathbf{r} \, d\Gamma \quad \text{and} \quad \mathbf{f}_\text{ext} = \int_{\Gamma_i} \mathbf{N}^T \mathbf{r} \, d\Gamma.
\] (22)

The electromechanical “stress” and “traction” vectors are then given by
\[
\mathbf{S} = \begin{pmatrix} \mathbf{\sigma} \\ \mathbf{D} \end{pmatrix} \quad \text{and} \quad \mathbf{\tau} = \begin{pmatrix} \mathbf{t} \\ \mathbf{q} \end{pmatrix},
\] (23)

respectively.

### 3. Constitutive behaviour

In order to solve the electromechanical equilibrium Eq. (21), the constitutive behaviour of the material needs to be prescribed. Three constitutive laws are required for complete description of the polycrystal. First the stresses and electric flux densities need to be related to the strains and electric fields in the bulk material. Second, the traction and surface charge density on a grain boundary need to be described in terms of the opening of a grain boundary and the jump in electric potential over it. Finally, a similar law needs to be derived for the cracks in the bulk material.

#### 3.1. Piezoelectric bulk material

In this contribution one of the most commonly used piezoelectric ceramics, lead zirconate titanate (PZT) with chemical formula \( \text{Pb}(\text{Zr}_{x}\text{Ti}_{1-x})\text{O}_3 \) is studied. Experiments show that the magnitude of the piezoelectric effect in PZT significantly depends on the stoichiometric ratio of zirconate and titanate [13]. A significant piezoelectric effect is observed in the case that \( x = 0.5 \) when the PZT is in the morphologic phase boundary (MPB). On the scale of the crystal lattice, the piezoelectric effect observed in PZT is caused by the off-centred zirconium or titanium atom. In order to obtain a piezoelectric bulk specimen, a strong electric field is applied to a specimen in order to align the directions of the off-centred atoms. This process is referred to as the poling process and the direction in which the electric field is applied is called the poling direction.

Here the constitutive behaviour of the bulk material is assumed to be linear and isotropic [14]. Furthermore, the assumption of a plane strain condition is adopted. Both assumptions are made in order to keep the focus of this contribution on the development of the constitutive laws for the cracks. Although beyond the scope of this contribution, it should be mentioned that an improved material description is of special interest on the small scales considered.

Under the assumptions mentioned above, the mechanical stress and electrical flux density can be related to the strain and electric field by
\[
\mathbf{\sigma} = \mathbf{H} \mathbf{E} - \mathbf{e}_1 \mathbf{E},
\]
\[
\mathbf{D} = \mathbf{e} \mathbf{E} + \mathbf{\varepsilon} \mathbf{E}.
\] (24)

In this expression \( \mathbf{H} \) is the Hookean matrix (Voigt form of the fourth-order Hookean tensor) for an isotropic material under plane strain and \( \mathbf{\varepsilon} \) is the permittivity tensor with only values on its diagonal. The PZT considered has class 6 mm symmetry [14], for which the piezoelectric matrix \( \mathbf{e} \) (Voigt form of the third-order piezoelectric tensor) for a three-dimensional piece (with axis 1, 2 and 3) is given by
\[
\mathbf{e} = \begin{bmatrix}
0 & 0 & 0 & e_{15} & 0 \\
0 & 0 & 0 & e_{15} & 0 \\
e_{31} & e_{31} & e_{33} & 0 & 0
\end{bmatrix},
\] (25)

where the “3”-axis is aligned with the poling direction.

#### 3.2. Initially elastic grain boundary interface

Microscopic studies of the material in the grain boundaries show that the molecular composition of the material inside the grain boundaries differs significantly from the material in the grains [15]. Since a significant piezoelectric effect is only expected in the case that PZT is near the morphologic phase boundary, it is assumed that the material in the grain boundary itself is not piezoelectric. It is therefore assumed that a purely mechanical cohesive law can be employed to relate the traction to the opening of a crack. This law is then enhanced to include electrical effects.
3.2.1. Mechanical traction-opening relation

The traction is related to the opening by means of the Xu-Needleman cohesive law [16]. Assuming that the fracture toughness in normal (mode I) and shear (mode II) are both equal to \(\gamma_c\) and assuming that the opening in normal direction after complete shear failure is zero in the case of zero normal traction, gives the mechanical potential function

\[
\phi^m(\mathbf{u}) = \gamma_c \left[1 - \left(1 + \frac{[u_n]}{\delta_n}\right) \exp \left(-\frac{[u_n]}{\delta_n}\right) \exp \left(-\frac{[u_s]^2}{\delta_s^2}\right)\right],
\]

where \([u_n] = \mathbf{u} \cdot \mathbf{n}\) and \([u_s] = \mathbf{u} \cdot \mathbf{s}\) are the normal and shear components of the opening, respectively. Note that, due to the linearised kinematics, the normal and shear directions are the same on both sides of a crack. The parameters \(\delta_n\) and \(\delta_s\) are the characteristic length parameters that are related to the ultimate traction \(t^m\) (the same in normal and in shear direction) and fracture toughness by \(\delta_n = \gamma_c/(t^m e)\) and \(\delta_s = \gamma_c/(t^m \sqrt{2} e)\) with \(e = \exp(1)\). The mechanical traction components are obtained by differentiation of Eq. (26) with respect to the corresponding opening components to yield

\[
t_n^m = \frac{\partial \phi^m}{\partial [u_n]} = \gamma_c \frac{[u_n]}{\delta_n} \exp \left(-\frac{[u_n]}{\delta_n}\right) \exp \left(-\frac{[u_s]^2}{\delta_s^2}\right),
\]

\[
t_s^m = \frac{\partial \phi^m}{\partial [u_s]} = \frac{2 \gamma_c}{\delta_s} \frac{[u_s]}{\delta_n} \left(1 + \frac{[u_n]}{\delta_n}\right) \exp \left(-\frac{[u_n]}{\delta_n}\right) \exp \left(-\frac{[u_s]^2}{\delta_s^2}\right).
\]

The relations for pure mode I and pure mode II opening are illustrated in Fig. 2. Secant unloading is assumed and the loading condition is checked on the basis of the Kuhn-Tucker conditions with history parameter \(\kappa = \sqrt{[u_n]^2 + [u_s]^2}\).

3.2.2. Electrical relation

In order to enhance the mechanical traction-opening law (27) with electrical properties, the interface is considered as a parallel plate capacitor (Fig. 3). For such a capacitor it is assumed that:

- The plates are infinitely long, such that the effect of fringing fields can be neglected.
- The plates are not tilted with respect to each other, such that the electric field is aligned with the normal direction (n).

Since, strictly speaking, both assumptions are violated by the grain boundaries considered, the parallel plate capacitor assumption needs to be considered carefully. The appropriateness of this assumption for the configurations considered in this contribution is demonstrate in Section 5.

Assuming the grain boundaries to behave like a parallel plate capacitor, the electric field is given by

\[
E_{\text{cap}} = -\frac{\Phi}{d_{gb} + [u_n]},
\]

where \(d_{gb}\) is the thickness of the capacitor at zero normal opening. This thickness is required in order to avoid the singularity arising when the normal jump is equal to zero while a potential jump is present. Physically the thickness \(d_{gb}\) is interpreted as the grain boundary thickness, which for the material considered is typically of the order of 10 nm. It should be emphasised that an interface element has no thickness and that the thickness \(d_{gb}\) is a material parameter.

The permittivity of the medium in between the capacitor plates should represent the permittivity of the grain boundary. Initially, the permittivity of the grain boundary material \(\varepsilon_{gb}\) has a value depending on its constituents. Upon opening, cracks will emerge in the grain boundaries, decreasing the effective permittivity. Once the crack has fully opened, the permittivity
of the grain boundary has attained the value of vacuum \( \lambda_0 \) (or of the medium that filled the crack). This deterioration of the material is incorporated in the constitutive behaviour by introduction of a scalar damage parameter

\[
\omega = \left( 1 - \frac{k_n}{k_n^0} \right) \left( 1 - \frac{\lambda_0}{\lambda_{gb}} \right),
\]

where \( k_n \) is the secant stiffness in normal direction and \( k_n^0 = G_c/\delta_n^2 \) is the secant normal stiffness in the undamaged state. The effective grain boundary permittivity \( \lambda_{gb} \) is then given by

\[
\lambda_{gb} = \left( 1 - \omega \right) \lambda_{gb} + \omega \lambda_0.
\]

Consequently, the electric flux density in normal direction is found as

\[
D_{cap} = \lambda_{gb} E_{cap},
\]

which upon substitution of (28) and (30) gives the surface charge density as

\[
q = -D_{cap} = \frac{\lambda_{gb} \Phi}{\lambda_{gb} + \|u_n\}}.
\]

The positive and negative charges on the plates of the capacitor are attracting each other, causing an additional electrostatic traction contribution

\[
t_n^e = \frac{q^2}{2 \lambda_{gb}} = \frac{1}{2} \lambda_{gb} E_{cap}^2.
\]

Since no electrical traction contribution in shear direction is assumed, the electrical traction vector can be written as

\[
t^e = t_n^e \mathbf{n} = \frac{1}{2} \lambda_{gb} \left( \frac{\Phi}{\lambda_{gb} + \|u_n\}} \right)^2 \mathbf{n},
\]

where (28) is used to substitute the electric field.

### 3.2.3. Electromechanical interface

The mechanical and electrical relations derived in the previous sections can be combined to yield a general electromechanical cohesive law

\[
\mathbf{t} = \mathbf{t}([\mathbf{u}],[\Phi]) = \mathbf{t}^m([\mathbf{u}]) + \mathbf{t}^e([\mathbf{u}],[\Phi]),
\]

\[
q = q([\mathbf{u}],[\Phi]),
\]

where \( \mathbf{t}^m, \mathbf{t}^e \) and \( q \) are given by Eq. (27), (34) and (32), respectively. The absence of a mechanical contribution to the surface charge density is caused by the fact that the material in the grain boundary is assumed not to be piezoelectric. Nevertheless, the above relations are fully coupled by means of the surface charges and electrostatic forces.

### 3.3. Initially rigid bulk interface

The cohesive law for the interfaces in the bulk material differs from that for the grain boundary on two major points. First the cohesive law is initially rigid, which means that it has an infinite stiffness before opening and hence a non-zero traction at zero opening. After failure initiation, a constitutive law with finite stiffness is employed. The algorithmic treatment of the initially rigid interfaces is elaborated in Section 4.2. Secondly, the interface resides in the piezoelectric bulk material and hence a piezoelectric effect is to be expected.
3.3.1. Mechanical traction-opening relation

As for the grain boundary, a commonly used mechanical traction-opening law is considered as a starting point. The relation proposed in Ref. [17] is used to relate the traction components to the displacement jump using

\[
t_m^m = t_m^m \exp \left( -\frac{t_m^m \kappa}{\gamma_c} \right) \quad \text{and} \quad t_s^m = t_s^m \exp(h_s \kappa)(1 + k_s [u_s]),
\]

(37)

where \( \kappa \) is a history parameter defined as the maximum achieved value of the normal opening, \([u_n]\), up to the current time instant. The loading condition is checked using the Kuhn-Tucker conditions. As can be seen from Fig. 4, the normal mechanical traction remains constant while unloading. The mechanical traction in shear direction unloads along its secant.

In (37) the parameters \( t_n \) and \( t_s \) are the normal and shear traction in the undamaged state \((\kappa = 0)\) with zero opening \((s_{un} = 0)\), respectively. Furthermore \( \gamma_c \) is the mechanical fracture toughness and \( t_m^m \) a prescribed ultimate traction. The parameter \( h_s \) governs the degradation of the shear stiffness \( k_s \) and is here directly related to the damage in normal direction by taking \( h_s = -t_m^m / \gamma_c \). The initial value for the shear stiffness \( k_s \) is assumed to be equal to the initial shear stiffness of the Xu-Needleman law used for the grain boundary interfaces as

\[
k_s = \frac{(t_m^m \sqrt{\bar{e}})^2}{\gamma_c},
\]

(38)

which is obtained by differentiation of the shear traction (27) with respect to the shear opening \([u_s]\) and evaluating at zero opening. The traction-opening law in pure normal and pure shear directions is shown in Fig. 4. Note that the cohesive law as used in Ref. [17] has been adapted slightly in order to ensure the traction continuity condition \( t_m^m(0) = t_m^m \) in the undamaged state.

3.4. Electrical and piezoelectrical contributions

In order to derive the traction and charge contributions following from the piezoelectric effect, the interface is again considered as a parallel plate capacitor with normal \( n \). The traction and electric flux density on that capacitor are then given by

\[
t_{cap} = N^T \sigma \quad \text{and} \quad q_{cap} = n^T D,
\]

(39)

with \( N \) being a matrix that projects the Voigt form of the stress tensor on the virtual plane parallel with the capacitor. Requiring that the rate of work of the projected components equals the rate of work in the original domain gives the projected strain and electric field as

\[
\varepsilon = Ne_{cap} \quad \text{and} \quad E = E_{cap} \cdot n.
\]

(40)

Substitution of (39) and (40) in (24) yields

\[
\sigma_{cap} = H_{cap} e_{cap} - E_{cap} e_{cap}^T, \quad D_{cap} = e_{cap} e_{cap} + \lambda_{cap} E_{cap},
\]

(41)

with

\[
H_{cap} = N^T H N, \quad e_{cap} = n^T e N, \quad \lambda_{cap} = n^T \lambda n.
\]

(42)

Here it is assumed that these matrices damage in a similar way as the normal component of the traction (37), yielding

\[
\bar{H}_{cap} = (1 - \omega) H_{cap}, \quad \bar{e}_{cap} = (1 - \omega) e_{cap}, \quad \bar{\lambda}_{cap} = \lambda_0 + (1 - \omega)(\lambda_{cap} - \lambda_0).
\]

(43)
with \( \omega = 1 - \exp(-t^m/K) \) and assuming that once fully opened the permittivity in the crack is dropped to the permittivity of vacuum. As in the case of the grain boundary interface, electric charges (32) and electrostatic forces (34) add contributions to the constitutive law, yielding

\[
\begin{pmatrix}
  t \\
  q
\end{pmatrix}
= \begin{pmatrix}
  t^m \\
  0
\end{pmatrix} - E_{\text{cap}} \begin{pmatrix}
  0 \\
  \zeta_{\text{cap}}
\end{pmatrix} + \left( \frac{1}{2} \zeta_{\text{cap}} E_{\text{cap}}^2 n \right).
\]

In the case that the material under consideration is piezoelectric, an additional traction contribution is obtained as

\[
t^e = -E_{\text{cap}} \bar{e}_{\text{cap}}.
\]

The presence of deformations in the piezoelectric material will also cause additional surface charges according to

\[
q^m = -e_{\text{cap}} \bar{m}_{\text{cap}}.
\]

where the mechanical contribution of the strain, \( \epsilon_{\text{cap}}^m \), is related to the mechanical contribution of the traction using

\[
t^m = H_{\text{cap}} \epsilon_{\text{cap}}^m.
\]

Using this expression, the piezoelectric contribution to the surface charge density (46) can be written as

\[
q^m = -e_{\text{cap}} H_{\text{cap}}^{-1} t^m.
\]

Using (45) and (48), the constitutive law (44) can be reformulated as

\[
\begin{pmatrix}
  t \\
  q
\end{pmatrix}
= \begin{pmatrix}
  1 \\
  -e_{\text{cap}} H_{\text{cap}}^{-1}
\end{pmatrix} \begin{pmatrix}
  t^m \\
  0
\end{pmatrix} - E_{\text{cap}} \begin{pmatrix}
  \bar{e}_{\text{cap}} \\
  \zeta_{\text{cap}}
\end{pmatrix} + \left( \frac{1}{2} \zeta_{\text{cap}} E_{\text{cap}}^2 n \right).
\]

Note that the presence of piezoelectric material in the capacitor introduces an additional dependence of the total traction on the electric field. In the proposed model, this additional contribution represents the experimentally observed dependence of the fracture strength on the electric field [6].

4. Algorithmic aspects

Some aspects regarding the implemented finite element model require further explanation. The generation of a periodic polycrystal is first discussed. After that the fracture criterion for transgranular fracture is elaborated. Also the method for tracing the equilibrium path is explained.

4.1. Representation of a polycrystal

Polycrystals can effectively be generated using Voronoi tessellations [18]. The procedure is to generate a number of nucleation points in a domain. A subdomain can then be defined for each nucleation point as all points in the domain closest to that nucleation point. A schematic representation of this procedure is shown in Fig. 5. This geometrical procedure mimics the phenomenon of isotropic grain growth. Here, the nucleation points are generated according to a uniform distribution.

In this contribution periodic boundary conditions are employed. Geometrical periodicity of the polycrystal is therefore required. This geometrical periodicity is obtained by copying the nucleation points to neighbouring domains and then perform the Voronoi tessellation on the big domain. A periodic polycrystal is then obtained in the centre (Fig. 5).

4.2. Fracture criterion and traction continuity for transgranular fracture

In order to model transgranular fracture, interface elements are present in between all elements inside the grains. The interfaces used for modelling fracture in the bulk material are initially rigid [19,20]. For that reason interfaces are initially constrained, which has two advantages. First the bulk interfaces do not add additional flexibility to the polycrystal, avoiding severe mesh dependency. Second, the number of degrees of freedom remains limited, since the constrained degrees of freedom for the bulk interfaces are eliminated from the total system of equation.

Transgranular fracture is modelled by activation (removal of the constraints) of specific bulk interface elements. This is done by checking a mode-I fracture criterion on the basis of the mechanical traction as

\[
t^m_n \geq \Gamma_n.
\]

where \( \Gamma_n \) is the prescribed ultimate mechanical traction. The mechanical traction at all interface integration points are evaluated after each converged load step. This is done by computation of the nodal reaction forces and charges following from the applied constraints to keep the interfaces closed. Since a Newton–Côtes integration scheme (integration points are located at the same position as the nodes in the undeformed state) is used for the interface elements, the traction in an interface integration point is determined by
where \( w_i \) is the Newton–Côtes weight at integration point \( i \), \( f_i \) is the reaction force at the node corresponding to that integration point and \( t_i \) and \( q_i \) are the traction and surface charge density.

Since a piezoelectric material is considered, the traction is composed of two parts: a mechanical part and an electrical part. Since the mechanical traction is required to evaluate the fracture criterion (50), this part of the traction needs to be computed from the given total traction and charge density. This is done by considering expression (49) in the undamaged state as

\[
\begin{pmatrix}
    t_m \\
    q_m
\end{pmatrix} = E_{\text{cap}} \begin{pmatrix}
    e_{\text{cap}}^1 & 0 \\
    -e_{\text{cap}} & 1
\end{pmatrix} \begin{pmatrix}
    e_{\text{cap}}^1 \\
    q_{\text{cap}}^1
\end{pmatrix} + \left( \frac{1}{2} E_{\text{cap}}^2 \right) \begin{pmatrix}
    \frac{1}{2} E_{\text{cap}}^2 & 0 \\
    0 & 0
\end{pmatrix} \begin{pmatrix}
    t_m \\
    q_m
\end{pmatrix}.
\]

Since the total traction \( t \) and surface charge density \( q \) follow from (51), the only unknown quantities in this equation are the components of the mechanical traction \( t_m \) and the electric field in the capacitor \( E_{\text{cap}} \). The nonlinear system of Eqs. (52) can therefore be solved for these unknowns by using a Newton–Raphson procedure. As a starting point for this iterative method, the solution of the system in the absence of piezoelectricity (\( e_{\text{cap}} = 0 \)) is used as

\[
E_{\text{cap}} = -\frac{q}{\kappa_{\text{cap}}} \quad \text{and} \quad t^m = t - \frac{1}{2} \frac{q^2}{\kappa_{\text{cap}}} n.
\]

Once the mechanical traction is obtained, the fracture criterion (50) is checked. If the criterion is violated the constraints on the considered element are released (the element is activated). From a robustness point of view, it is important that traction

\[
f_i = w_i \begin{pmatrix}
    t_i \\
    q_i
\end{pmatrix},
\]

Fig. 5. Schematic representation of periodic polycrystal generation. From top-left to bottom-right: (1) Nucleation points are generated in the original domain. (2) A larger domain is considered and the original nucleation points are copied to the neighboring cells. (3) The Voronoi algorithm is applied to the large domain, yielding a non-periodic crystal. (4) The original domain is selected from the larger domain, yielding a periodic polycrystal.
continuity [21] is satisfied upon activation of an interface. This means that the traction and surface charge density at zero opening (and zero potential jump) should be chosen such that the forces resulting from the cohesive interface are equal to the reaction forces. This continuity condition is satisfied by adjustment of \(\mathbf{t}^n\) in the constitutive law (49).

4.3. Dissipation-based path-following constraint

For the computations considered, the external force is prescribed by

\[
\mathbf{f}_{\text{ext}} = \lambda \mathbf{f},
\]

where \(\lambda\) is a load scale and \(\mathbf{f}\) a unit external force vector. Substitution of this equation in the equilibrium Eq. (21) yields

\[
\mathbf{f}_{\text{int}}(\mathbf{a}) = \lambda \mathbf{f}.
\]

In order to solve this system of \(n\) equations with \(n + 1\) unknowns (the nodal degree of freedom vector \(\mathbf{a}\) and the load scale \(\lambda\)) an additional constraint equation is required. Since material softening is considered, a “force”-control constraint (additional equation only depending on \(\lambda\)) cannot be used to trace the equilibrium path beyond the maximum load. A “displacement”-control constraint (additional equation only depending on \(\mathbf{a}\) cannot be used since for the computations considered snapback occurs. In order to trace the complete equilibrium path, an “arc-length” constraint is used (additional equation depending on both \(\lambda\) and \(\mathbf{a}\)).

Since the rate of dissipation is monotonically increasing from a physical point of view, this quantity can be used to derive an arc-length constraint [22]. The rate of dissipation can be formulated as

\[
P = P - V,
\]

where \(P\) is the external power applied on the domain \(\Omega\) and \(V\) is the recoverable energy stored in the domain. The external power can be formulated as

\[
P = \mathbf{f}_{\text{ext}}^T \mathbf{a} = \lambda \mathbf{f}^T \dot{\mathbf{a}}
\]

and the recoverable energy is given by the Helmholtz free-energy as

\[
V = \frac{1}{2} \int_{\Omega} \sigma^e : \mathbf{E} d\Omega + \frac{1}{2} \int_{\Gamma_c} \mathbf{t}^T [\mathbf{u}] + q[\Phi] d\Gamma.
\]

Using (5) and (14) this can be rewritten as

\[
V = \frac{1}{2} \mathbf{a}^T \left[ \int_{\Omega} \mathbf{B}^T \mathbf{S} d\Omega + \int_{\Gamma_c} \mathbf{M}^T \tau d\Gamma \right] = \frac{1}{2} \mathbf{a}^T \mathbf{f}_{\text{int}}.
\]

Substitution of this equation and the expression for the external power (57) in Eq. (56) then yields the rate of dissipation as

\[
G = \frac{1}{2} \mathbf{f}^T (\dot{\mathbf{a}} - \lambda \mathbf{a}).
\]

The equilibrium path can then be traced by integrating the system

\[
\begin{bmatrix}
\mathbf{K} & -\mathbf{f} \\
\frac{1}{2} \mathbf{f}^T & -\frac{1}{2} \mathbf{f}^T \mathbf{a}
\end{bmatrix} \begin{bmatrix}
\dot{\mathbf{a}} \\
\dot{\lambda}
\end{bmatrix} = \begin{bmatrix}
0 \\
G
\end{bmatrix}.
\]

5. Numerical simulation

The proposed piezoelectric finite element model is tested using 40 \(\times\) 40 \(\mu\text{m}^2\) polycrystals with various average grain sizes. Each polycrystal is subjected to periodic boundary conditions for the displacements and electric potential and is loaded mechanically in the horizontal direction by a force \(\mathbf{F}\) in the horizontal direction (Fig. 6). Note that, as a consequence of the periodic boundary conditions, this concentrated force \(\mathbf{F}\) represents the resultant force of a distributed traction \(\mathbf{t}_{\text{right}}\) over the right edge. Therefore the combination of this discrete force and the periodic boundary conditions can be interpreted as a dis-
distributed load over the edges. The shape of these distributed loads is unknown in advance and generally changes during a simulation. The resultant force and charge on the top and bottom edge are equal to zero as a consequence of the applied loading conditions. Upon loading, a potential difference between the left and right edge will appear due to the piezoelectric effect of the considered material. The average potential jump \( V \) is measured using a voltage meter as indicated in Fig. 6.

The commonly used piezoelectric ceramic PZT-4 is used to define the material parameters. It is assumed that the grains are perfectly poled in the horizontal direction (the "3"-axis in (25) is aligned with the horizontal axis). The bulk material parameters for PZT-4 are used as in Ref. [6] and are assembled in Table 1. In Ref. [6] the fracture toughness of a bulk PZT-4 specimen is determined to be \( G_c = 2.34 \times 10^3 \) N/mm. Here this value for the fracture toughness is used for both the grain boundary constitutive law and bulk constitutive relation. The ultimate mechanical traction \( t_{\text{ult}} \) for PZT-4 is found as 80 MPa (e.g. [23]). For the ultimate traction, no clear distinct values for trans- and intergranular fracture were found. For that reason, the value of 80 MPa is used for both cohesive laws. The grain boundary thickness is approximated to be 10 nm [15]. Note that the grain boundaries are modelled using zero thickness interface elements and that this grain boundary thickness is merely a parameter used for the constitutive model. The grain boundary permittivity \( k_{gb} \) is assumed to be equal to \( k_{11} \). The shear stiffness \( k_s \) used for the transgranular cohesive law is computed using (38) as \( 7.4 \times 10^6 \) MPa/mm.

5.1. Accurateness of the parallel plate approximation

As outlined in Section 3.2.2, it is assumed that the electrical constitutive behaviour of the grain boundaries can be based on a parallel plate capacitor. Since the sides of a grain boundary are in general neither parallel, nor infinitely long, the appropriateness of this assumption is debatable. In the absence of experimental results, the accuracy of the parallel plate capacitor assumption is verified numerically. The effects of tilting of the plates, plate misalignment and fringing fields are examined on the basis of a micro mechanical model for the grain boundary.

In the presented numerical model, grain boundaries are considered as infinitely thin line elements. This assumption is made since the thickness of the grain boundaries is small compared to their lengths. Alternatively, the grain boundaries could have been modelled as very slender 2D (in a 2D simulation) material domains. This approach would require fewer assumptions and effects like grain boundary tilting and grain boundary shearing would be incorporated in the simulation. However, from a numerical point of view this approach is impractical. Discretisation of the grain boundaries would introduce an enormous amount of additional degrees of freedom, leading to a significant increase in computation time.
Although micro mechanical modelling of all grain boundaries throughout complete simulations is impractical, it can be employed to verify the assumption of the parallel plate capacitor. An intergranular crack (as simulated by the interface element model) is considered to derive boundary conditions for a micro mechanical finite element model that solves the electrostatic equation in the grain boundaries (see Fig. 7). The considered crack is chosen such that the total charge on a crack side is at its maximum, since in that case the largest absolute error in the internal force vector \( (22) \) is to be expected.

Considering a typical polycrystal, the crack for which the largest absolute error is expected is shown in Fig. 8. Although this is an intergranular crack, no fundamentally different results for transgranular cracks are expected when considering the appropriateness of the parallel plate capacitor. The magnitude of the opening, the mode-mixity of the opening, the tilt angle of the crack sides and the potential jump across the crack are shown in Fig. 9. The maximum opening is equal to 16.3 nm, the maximum mode-mixity is 6.41 and a maximum tilt angle of 2.93° is observed. The maximum jump in potential is equal to 3.9 V. From Fig. 9 is also observed that parts of the grain boundary are already significantly damaged. The micro mechanical model for the grain boundary is discretised using 336,454 linear triangular elements, leading to a system of 168,233 degrees of freedom.

The surface charge density resulting from the grain boundary finite element simulation is shown in Fig. 10. The surface charge density on the left side of the crack \( q^- \) is of opposite sign of the surface charge density at the right side \( q^+ \). As can be seen, the mismatch in surface charge density magnitude over the crack is small as a consequence of the slenderness of the crack. Furthermore it is observed that the parallel plate approximation accurately fits the results from the micro mechanical finite element simulation. The largest relative errors are observed in the regions where the mode-mixity is relatively large. In

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**Fig. 7.** The state of the interface element (left) is used to derive boundary conditions for a finite element model for the grain boundary (right). Note that the physical thickness of the grain boundary \( d_{gb} \) is incorporated in the construction of the deformed grain boundary domain. In this domain, the electrostatic equation is solved.

**Fig. 8.** An intergranular crack resulting from the proposed model is examined using a micro mechanical model for the fractured grain boundaries. The crack is discretised using 336,454 linear triangles, leading to 168,233 degrees of freedom. The details show the electric potential in the crack at: (A) the maximum mode-mixity; (B) the maximum opening; (C) the maximum tilt angle.
these regions, however, the absolute error remains small. It can therefore be concluded that, for the problem considered, the parallel plate capacitor assumption is appropriate for the derivation of the electrical contributions to the cohesive laws.

5.2. Fracture of polycrystals

The proposed numerical model is employed to model the fracture process of the six polycrystals shown in Fig. 11. The considered grain sizes correspond with typical sizes observed in literature (e.g. [24]) and are assembled in Table 2. The average grain size is determined using the mean intercept length [24].

The polycrystals are discretised using approximately 6000 three-noded triangular elements with a Gaussian integration scheme. The grain boundaries are discretised using four-noded interface elements with a Newton–Côtes integration scheme. The number of grain boundary interfaces depends on the total length of grain boundaries. In between the bulk elements approximately 8000 four-noded bulk interfaces with a Newton–Côtes integration scheme are present. In the undamaged state this discretisation leads to systems of approximately 50,000 degrees of freedom. The equilibrium path is traced using an ultimate dissipation increment $\Delta t$ of $5 \times 10^{-3}$ N mm, which satisfies the requirement (62) since the characteristic element edge size $l_e$ is equal to $7.5 \times 10^{-4}$ mm. The cohesive zone length can be approximated (for a mechanical problem) using

**Fig. 9.** Boundary conditions used for the micro mechanical simulation of the considered crack. The crack coordinate $s$ is defined in Fig. 8. Furthermore, $\|u\|$ is the magnitude of the crack opening, $|\|u_1\|/|u_n\|$ is the mode-mixity, $\theta$ is the tilt angle, $\|\Phi\|$ is the potential jump and $\omega$ is the damage parameter.
various methods [25]. Hillerborg’s model for example yields $E_G c_{22} t_m^2 = 30 \text{ m}$. Although this result might be affected by the electromechanical coupling, the characteristic element length is sufficiently small to assume that the cohesive zones are discretised appropriately. This setting typically requires 200–300 load steps and a computation time of approximately half hour on a 2 GHz Intel Core Duo with 2 GB SDRAM.

The response of a polycrystal is measured in terms of average stresses and strains. The average horizontal strain $\varepsilon_{xx}$ is defined as the average horizontal displacement over the right edge divided by the width of the specimen. The average horizontal stress $\sigma_{xx}$ is defined as the sum of the reaction forces over the right edge divided by the length of that edge. The responses of the considered polycrystals to the applied loading are shown in Fig. 12.

Initially a linear relation between the average stress and average strain is observed for all polycrystals. Also the potential difference as measured by the volt meter increases linearly. The ultimate loads and potential differences for the six polycrystals are collected in Table 2. As can be seen, the ultimate load (and potential difference) varies significantly for polycrystals with approximately the same average grain size (e.g. B.I and B.II). This variation is a consequence of the differing grain
shapes. The variation depends on the total number of grains in the domain considered. If a domain of e.g. 100 \(\times\) 100 \(\mu\)m would be considered, the ultimate values would have a significantly smaller spread. For the 40 \(\times\) 40 \(\mu\)m polycrystal considered here, significant variations are encountered. The results, however, show some important characteristics of the proposed model.

Table 2
Parameters used for the generation of the considered polycrystals and results of the finite element simulations performed on these polycrystalline structures

<table>
<thead>
<tr>
<th></th>
<th>(n_g)</th>
<th>(l_g) ((\mu)m)</th>
<th>IG (%)</th>
<th>TG (%)</th>
<th>(&lt;\sigma_{xx}\rangle_{\text{ult}}) (MPa)</th>
<th>(&lt;V_{\text{ult}}\rangle) (V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A.I</td>
<td>29</td>
<td>8.5</td>
<td>93</td>
<td>7</td>
<td>54.9</td>
<td>42.0</td>
</tr>
<tr>
<td>A.II</td>
<td>29</td>
<td>9.2</td>
<td>91</td>
<td>9</td>
<td>63.4</td>
<td>49.1</td>
</tr>
<tr>
<td>B.I</td>
<td>8</td>
<td>16.8</td>
<td>78</td>
<td>22</td>
<td>71.7</td>
<td>55.1</td>
</tr>
<tr>
<td>B.II</td>
<td>8</td>
<td>17.1</td>
<td>81</td>
<td>19</td>
<td>53.2</td>
<td>41.0</td>
</tr>
<tr>
<td>C.I</td>
<td>4</td>
<td>21.1</td>
<td>70</td>
<td>30</td>
<td>78.9</td>
<td>61.1</td>
</tr>
<tr>
<td>C.II</td>
<td>4</td>
<td>22.7</td>
<td>68</td>
<td>32</td>
<td>70.9</td>
<td>54.7</td>
</tr>
</tbody>
</table>

Here, IG and TG, respectively indicate inter- and transgranular fracture.

Fig. 12. Average horizontal stress \(<\sigma_{xx}\rangle\) (left) and average potential jump \(V\) (right) versus average strain \(<\varepsilon_{xx}\rangle\) for the performed numerical simulations.
After the ultimate load is reached, the polycrystals start to fracture. The wiggles observed in the fracturing part of the response curves are caused by the fact that not all grain boundaries are failing at the same moment. At the points where a crack propagates from one grain boundary into another, a strong curvature in the response curves is observed. It is in these regions where the use of an appropriate path-following constraint is required. The \( \sigma_{xx} \) stress field and electric potential field for the cracked polycrystals A.II and C.II is shown in Fig. 13. A potential jump over both inter- and transgranular cracks is noticed. The contribution of the electrostatic forces (33) is observed to be less than \( 1 \times 10^{-3} \text{ MPa} \) and is therefore negligible compared to the mechanical traction contribution. Comparison of the crack patterns for polycrystal A.II and C.II (Fig. 13) with the crack patterns for A.I and C.I (Fig. 14) shows the dependency of the crack pattern on the grain shapes.

An important aspect of the proposed numerical model is its capability to correctly mimic the transition from mainly intergranular fracture for relatively small grains to transgranular fracture in the case of relatively large grains. This transition is experimentally observed for PZT in [24], the results of which are collected in Table 3. In [24] the amounts of inter- and transgranular fracture are determined on the basis of the areal fractions observed on cracked specimens. Analogously, the amounts of inter- and transgranular fracture as observed from the numerical simulations is defined by the height ratios (as if one would look at a cracked specimen along the \( x \)-axis). The results of the numerical simulations are incorporated in Table 2. As can be seen the relative amount of transgranular fracture increases upon increasing the average grain size.

Although the model mimics this behaviour correctly from a qualitative point of view, quantitatively there is a significant difference between the results from the numerical simulations and those reported in [24] and Table 2. These differences can on the one hand be explained by the fact that the parameters used for the numerical simulations are obtained from different sources. On the other hand, assumptions like a plane strain condition and isotropy of the elasticity tensor are also likely to be sources of error.
In order to investigate the influence of the piezoelectric coupling, the simulations for polycrystal A.I and C.I have been done with the piezoelectric coefficients taken equal to zero. The responses are shown in Fig. 15. From the responses it is observed that, according to the numerical model, the maximum load is hardly affected by the piezoelectric coupling. The simulations show that the amount of transgranular fracture increases significantly for both polycrystals (from 7% to 20% for A.I and to 30% to 65% for C.I). This implies that the piezoelectric coupling prohibits transgranular fracture, which is a logical consequence of the electromechanical normal traction law (49). Although the ultimate mechanical normal traction, $t^m_n$, is taken as 80 MPa, the ultimate electromechanical traction is given by

$$\sigma_{xx} = \frac{t^m_n}{C_{22}}$$

### Table 3
Grain size dependence of the fracture mode of PZT as observed in [24]

<table>
<thead>
<tr>
<th>Average grain size ($\mu m$)</th>
<th>Intergranular fracture (%)</th>
<th>Transgranular fracture (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>9.4 ± 0.7</td>
<td>10.5</td>
<td>89.5</td>
</tr>
<tr>
<td>11.8 ± 0.2</td>
<td>20.0 ± 3.0</td>
<td>80.0 ± 3.0</td>
</tr>
<tr>
<td>14.5 ± 1.2</td>
<td>37.0 ± 7.0</td>
<td>63.0 ± 7.0</td>
</tr>
<tr>
<td>16.0 ± 0.8</td>
<td>70.0 ± 4.0</td>
<td>30.0 ± 4.0</td>
</tr>
<tr>
<td>17.8 ± 1.7</td>
<td>92.5 ± 2.5</td>
<td>7.5 ± 2.5</td>
</tr>
</tbody>
</table>

In order to investigate the influence of the piezoelectric coupling, the simulations for polycrystal A.I and C.I have been done with the piezoelectric coefficients taken equal to zero. The responses are shown in Fig. 15. From the responses it is observed that, according to the numerical model, the maximum load is hardly affected by the piezoelectric coupling. The simulations show that the amount of transgranular fracture increases significantly for both polycrystals (from 7% to 20% for A.I and to 30% to 65% for C.I). This implies that the piezoelectric coupling prohibits transgranular fracture, which is a logical consequence of the electromechanical normal traction law (49). Although the ultimate mechanical normal traction, $t^m_n$, is taken as 80 MPa, the ultimate electromechanical traction is given by

$$\sigma_{xx} = \frac{t^m_n}{C_{22}}$$

### Fig. 15
Average horizontal stress $\langle \sigma_{xx} \rangle$ versus average strain $\langle \epsilon_{xx} \rangle$ for the simulations of polycrystal A.I (left) and C.I (right), with (solid) and without (dashed) piezoelectric effect.
where the electrostatic forces are neglected. The negative electric field in the polycrystal therefore causes the ultimate traction to increase, which is in agreement with experimental observations [6].

6. Conclusion

A cohesive zone model for quasi-static fracture of piezoelectric polycrystals is proposed. Initially elastic interface elements are inserted in the grain boundaries of a polycrystal. Inside the grains, initially rigid interface elements are inserted in between all elements. These elements are initially constrained and activated once a fracture criterion on the basis of the internal reaction forces is satisfied.

The discrete fracture of the considered polycrystals is governed by electromechanical cohesive relations. The mechanical traction is related to the opening using commonly used models for purely mechanical problems. Electrical relations are derived based on the assumption that an interface can be considered as a parallel plate capacitor. On the basis of its molecular constituents, no piezoelectric effect is expected in the grain boundaries. Consequently, the only added traction contribution comes from electrostatic forces. The charges on the crack sides are purely generated by the electric field. For the cracks inside the grains, piezoelectric contributions are taken into account.

Numerical simulations are performed for six PZT-4 polycrystals with different average grain sizes and grain shapes. The numerical experiments demonstrated that the polycrystal with relatively small grains fails intergranularly, whereas in the crystal with larger grains transgranular cracks appear. This observation is in accordance with experimental observations. The numerical experiments correctly mimic the jump in the electric potential over cracks. Finally, the influence of a present electric field on the fracture criterion is observed to be incorporated in the model for transgranular fracture.

The proposed model is demonstrated to be applicable for modelling inter- and transgranular fracture in piezoelectric ceramics such as PZT-4. The enhancement of commonly used mechanical constitutive relations with parallel plate capacitor relations is demonstrated to correctly mimic several experimentally observed phenomena. The described model can be used to predict the maximum load of polycrystals. The suitability of the method for application of periodic boundary conditions offers the possibility for implementation in a multiscale framework.

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References