

Charge density on fracture surfaces and contact electrification of identical materials

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Contact electrification of identical materials happens during collisions, rubbing, and mixing of granular materials. Fractures often accompany these processes, and the resulting fracture surfaces are typically charged. Based on visible-light and X-ray observations of fracture, we demonstrate that the charge density on fracture surfaces of organic and inorganic materials is consistent with those observed in collisions, rubbing, and mixing. This indicates that fractures involve contact electrification of identical materials.

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I. INTRODUCTION

It is known that two objects are electrically charged after contact. This occurs for most materials and is referred to as contact electrification [1]. When the objects in contact are of different materials, whether an object is charged positively or negatively can be determined from the ordering of materials in terms of the direction of charge transfer between materials, which is called a triboelectric series and has been extensively studied [1–5]. Many studies have linked such triboelectric series to some intrinsic material properties such as work function. Even between identical materials, however, contact electrification also occurs; there should be a mechanism other than the differences in material properties. Contact electrification of identical materials happens through various processes such as contact and separation of flat surfaces [6,7], collision [8–10], rubbing [2,11–13], and mixing of granular materials [1,14–17]. Several mechanisms have been proposed, therefore, in different situations to explain how electrification occurs during the contact between identical materials such as asymmetric contact between two materials [12], statistical distributions of material properties on contact surfaces [6], polarization due to the electric field from charged particles [18,19], the effect of curvature [20], and flexoelectricity [21,22].

Fracture often accompanies the processes causing contact electrification of identical materials: for instance, collisions cause fractures near the impact point, and friction and mixing in granular systems lead to many localized microfractures. Fractures are frequently associated with various emission [23–40] (e.g., visible light [26–31], radio waves [32–36], charged particles (i.e., ions and electrons) [37,38], and X – rays [31,39,40]), suggesting that the newly formed

surface after a fracture is charged. This means that, when contact between two materials is accompanied by fracture, the materials are electrified by contact: contact electrification occurs.

In this study, we conduct fracture experiments to investigate the electrification of fracture surfaces as an elementary process in contact electrification. We focus on the surface charge density, which is one of the key parameters for contact electrification. Since it is difficult to directly measure the charge on the surface of fragments immediately after fracture due to its motion, we utilize the radiation emitted during fracture, which reveals the electrical conditions of the surface [41,42]. Samples are sucrose and tartaric acid as organic materials, and muscovite (mica) as an inorganic material, known to emit visible light [23,26,27] and X – rays [39,40] when fractured. First, we analyze the time variation in visible-light emission to estimate the maximum distance between the opposing surfaces during fracture. Next, based on the X-ray energy spectra recorded in evacuated environments, we evaluate the energy of electrons traveling between the newly created surfaces, assuming that the bremsstrahlung from electron bombardment generates these X-rays. Using the electron energy and the maximum distance, we obtain the charge density on fracture surfaces. Then, we compare this charge density with the surface charge densities previously obtained in contact electrification of identical materials. Finally, as possible processes of charge generation on fracture surfaces, we evaluate the piezoelectric effect and then consider the effect of flexoelectricity. The regulation mechanism of the electric charge on the fracture surface is then discussed.

II. EXPERIMENTS

We place a sucrose or tartaric acid sample approximately 1.0–1.5 cm in size and 2–6 g in mass between two plastic (acrylic resin) cylindrical bars with a diameter of 12 mm inside a vacuum chamber. One bar contacts a metal (copper) cylinder inserted into the chamber through a gauge port. A hydraulic press applies force to disrupt the samples. The force loading rate is not precisely set but does not exceed

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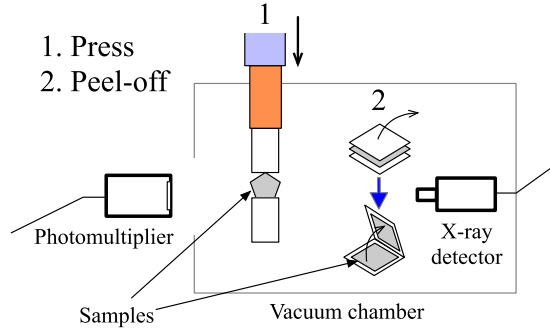


FIG. 1. Experimental setups: (1) press and (2) peel-off methods for the sample disruption. Visible-light and X-ray measurements were carried out in the atmosphere and vacuum, respectively.

approximately 1 mm/s. For a muscovite sample approximately 1 cm in size and 1–3 g in mass, we use a peel-off method. The sample is inserted and bonded between two plates. One plate is fixed and another one is opened by turning a rotator by hand from outside the vacuum chamber: the sample is cleaved. The schematic of both methods is shown in Fig. 1.

We observe visible light using a photomultiplier (H1051, Hamamatsu Photonics) in the atmosphere. The photomulti-

plier (PM) has a diameter of 15 mm. The distance between the samples and the PM in the press experiment is 90 mm, including a 20 μm thick polyvinylidene chloride film placed in front of the PM to prevent fragments from colliding with it, and is 150 mm in the peel-off experiment without film.

We detect X-rays generated during fracture using a Si detector with a 5 mm diameter and a 25.4 μm thick Be window (XR-100CR, AmpTek). The detector is placed in the vacuum chamber at a distance of 110 mm from the sample in the press experiment and 125 mm in the peel-off method. In the press experiment, a 12.5 μm thick polyimide film is positioned between the detector and sample to protect the detector from fine fragments. For sucrose, we vary the ambient pressure in the chamber from 5×10^{-1} to 1×10^3 Pa. For tartaric acid and muscovite, the ambient pressure is less than 20 Pa.

III. RESULTS

For organic samples, the visible-light radiation is occasionally observed over a long period of time [Fig. 2(a), left] because cleavage occurs on multiple planes during a single operation. The visible-light radiation consists of either one large pulse or several successive large pulses and numerous small pulses, as shown in the right panel of Fig. 2(a) (sucrose)

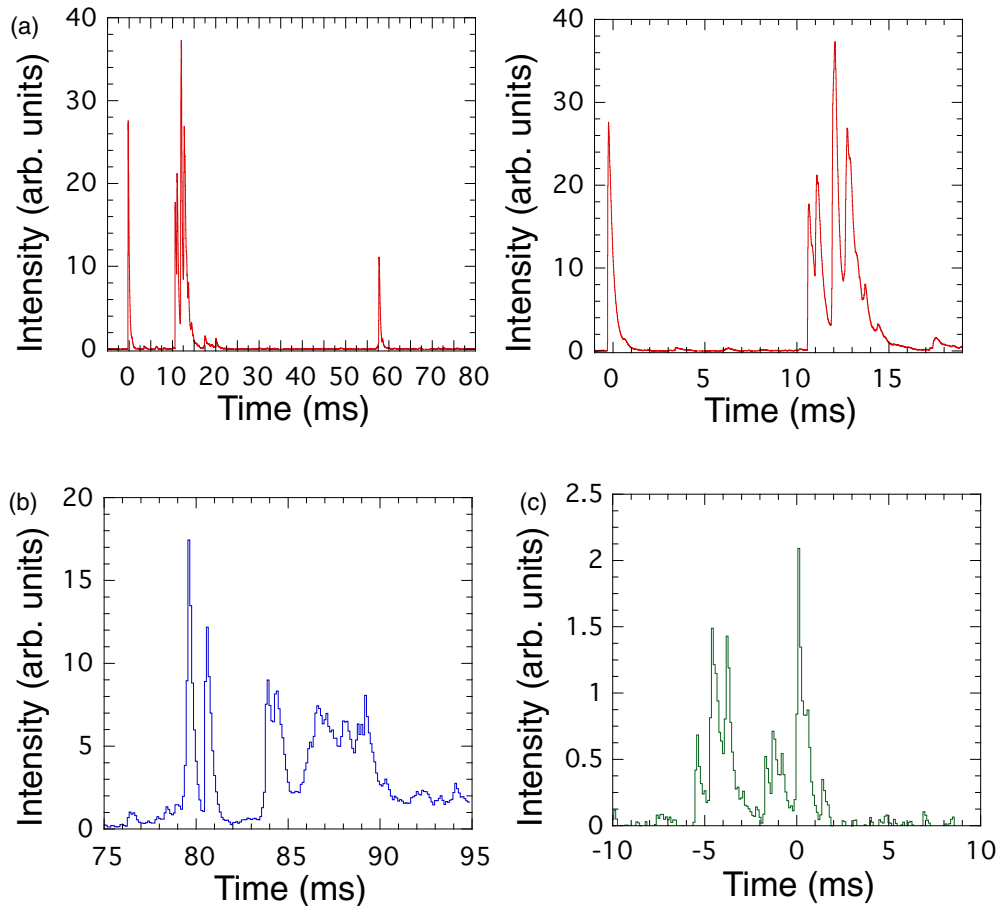


FIG. 2. Example of visible-light measurements during fracture as a function of time: (a) sucrose (left: long time range, right: the enlarged part of the left panel), (b) tartaric acid, and (c) muscovite (mica).

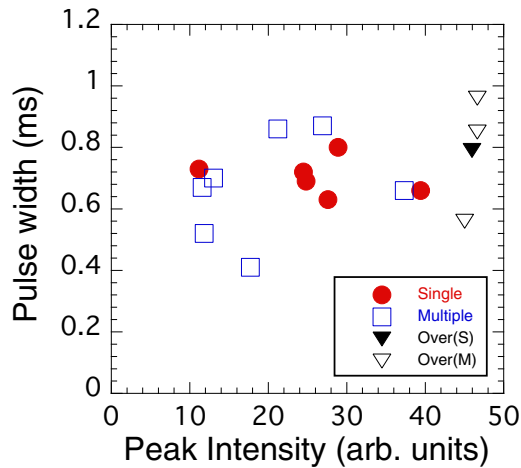


FIG. 3. Foot-to-foot (10% of peak intensity) pulse width for sucrose as a function of peak intensity. Data are from the fracture of six samples. “Single” and “Multiple” denote the pulses for which the time at 10% of the peak intensity can be determined before and after the peak and cannot be determined before and/or after the peak, respectively. “Over(S)” and “Over(M)” denote the pulses for which the peak intensity cannot be measured. The letters “S” and “M” in the parentheses represent the pulses where the time at 10% of the maximum measured intensity can and cannot be determined, respectively. In the case that the peak intensity could not be measured, the pulse width is plotted against the maximum measured intensity instead of peak intensity.

and Fig. 2(b) (tartaric acid). For muscovite, cleavage occurs on a single plane and usually there are several successive large pulses and numerous small pulses [Fig 2(c)]. These figures show that the occurrence of the pulses in any sample is sporadic. The sporadic variation in radiation is consistent

with the previous observations [23]. Furthermore, the width of each pulse obtained for all samples appears to be less than 1 ms. For sucrose, we examine the foot-to-foot (10% of peak intensity) widths of the largest 17 pulses, including four pulses with such high intensity that their peak intensities could not be measured because it exceeded the measurement limit of the photomultiplier. The results are shown in Fig. 3. “Single” (filled circles) denotes a pulse where the time at 10% intensity can be identified before and after the peak. “Multiple” (open squares) denotes a pulse where the time at 10% intensity cannot be determined due to proximity to another pulse and the time at the minimum intensity between two pulses is used to evaluate the pulse width. In the case that the peak intensity could not be measured, the pulse width is determined using either the time at 10% of the maximum measured intensity [denoted by “Over(S)”, filled inverted triangle] or, as in Multiple, the time at the minimum intensity between adjacent pulses [denoted by “Over(M)”, open inverted triangles]. We confirm that the foot-to-foot pulse width is actually less than 1 ms.

Figure 4(a) shows the spectra of X-ray counts per keV for sucrose at pressures of 1×10^3 , 5×10^2 , 1×10^2 , 5×10^1 , 1×10^1 , 5×10^0 , 1×10^0 , and 5×10^{-1} Pa as a function of photon energy in keV. The bin size is 0.5 keV. We have corrected the absorptions of the Be window and the polyimide film. The continuum component of the spectra decreases with photon energy. Below 5×10^1 Pa, the results were almost identical, but above this pressure, the X-ray counts decrease as the ambient pressure increases.

Figure 4(b) shows the spectra of X-ray counts per keV for tartaric acid (blue) and muscovite (green). The bin size is 0.5 keV. The continuum components of these spectra also decrease with photon energy. In the case of muscovite, the characteristic X-rays at Al (1.49 keV), Si (1.74 keV), and K

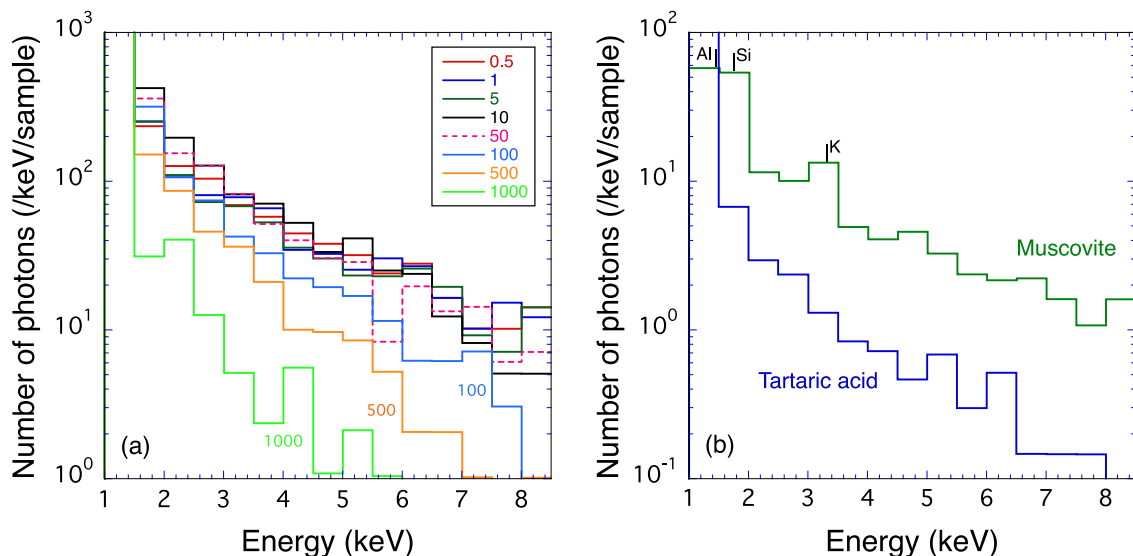


FIG. 4. X-ray spectra during fracture. The horizontal axis represents photon energy in keV, and the vertical axis denotes the photon counts divided by bin size (0.5 keV). (a) Sucrose: each spectrum represents the average of seven samples at each pressure. (b) Tartaric acid (blue) and muscovite (green): the results for tartaric acid and muscovite represent the averages of 14 and 15 samples, respectively. In the press method (sucrose and tartaric acid), postfracture vibrations of the vacuum chamber are propagated to the detector, resulting in noise at lower energies than 1.5 keV in the spectra.

(3.31 keV) result in higher photon counts at the corresponding energies than the level of the continuum component.

Note that the sample is intact until the emission is measured. In other words, no emission is observed before the sample is fractured. This implies that the emissions we observe are accompanied with the separation of fragments.

IV. DISCUSSION

Figure 3 shows that the pulse duration is less than 1 ms. When the charged fragments move and the distance between opposing fragment surfaces approaches the sample size, the electric field between the charged fragment surfaces is no longer uniform, resulting in a change in radiation. Therefore, the distance that the fragment moves during the maximum pulse width of 1 ms is expected to be less than the size of the sample, ~ 1 cm.

Crack propagation does not completely release the stress. Residual stress (i.e., elastic energy) causes the fragments to move after fracture, and the velocity of the fragments v is $\sim (E\delta^2/\rho)^{1/2}$, where E , δ , and ρ are Young's modulus, strain, and density, respectively [43]. For sucrose, with E set to 27.4 MPa and δ to 0.024 (Appendix A), and ρ to 1.59 g/cm^3 [44], the fragment velocity v is 3.2 m/s. Since the pulse duration of the visible-light radiation is less than 1 ms (Fig. 3), the maximum distance a fragment could move during one pulse is approximately $3.2 \text{ m/s} \times 1 \text{ ms} \sim 3 \text{ mm}$. This is consistently less than the sample size, ~ 1 cm.

Electrons traveling between charged fragment surfaces excite the ambient gas, leading to UV-visible radiation [23,25,27,28,45]. The mean free path l of an electron in the ambient gas is inversely proportional to the gas pressure. Assuming the velocity of the ambient gas molecules is much slower than that of the electrons and the radius of the gas molecules is $2.0 \times 10^{-10} \text{ m}$, l is 3.3 mm at 10 Pa, with the temperature assumed to be 300 K. When the pressure is so low that l exceeds the distance between opposing fragment surfaces, electrons can directly bombard the opposite surface, resulting in X-ray emission [31,39,40,46]. As the electron flight distance increases, their energy and the X-ray count should also increase. However, Fig. 4 shows that X-ray counts are independent of pressure below 50 Pa. This suggests that there is an upper limit to the electron flight distance, a few millimeters, which is consistent with the upper limit of the distance that a fragment can move during one visible-light pulse, approximately 3 mm.

We numerically estimated the counts of bremsstrahlung X-ray photons generated by electron bombardments on sucrose, tartaric acid, muscovite, and quartz using a model for X-ray tubes [47]. The composition of these samples is shown in Table I in Appendix B. Electrons with monochromatic energies of 3, 5, 7, 10, 15, and 20 keV collide with a flat sample surface at a 0° angle (vertical to the surface), and we investigate X-ray photons emitted at a 0° angle [48]. The numerically obtained X-ray spectra are shown in Fig. 5, compared with experimental results, including quartz obtained in previous experiments [31]. For sucrose [Fig. 5(a)], the result for incident electron energies of approximately 10 keV corresponds to the experimental X-ray spectra at ambient pressures below 50 Pa.

For tartaric acid [Fig. 5(b)], the result for incident electron energies of approximately 7–10 keV corresponds. For muscovite and quartz [Figs. 5(c) and 5(d)], electron energies of approximately 10–15 and 5–7 keV provide the corresponding continuum component in the spectra, respectively.

The charge density on fracture surfaces q is $\epsilon_0(V_c/L)$, where ϵ_0 is the dielectric constant of vacuum and V_c and L are the potential difference and distance between opposite fragment surfaces, respectively. Electrons gain energy corresponding to the potential difference during the acceleration between surfaces. Comparing experimental results and numerical results for X-ray counts for sucrose indicates that the maximum electron energy is 10 keV, corresponding to a V_c of 10 kV. Setting L to the maximum distance of approximately 3 mm, q becomes $3 \times 10^{-5} \text{ C/m}^2$ for sucrose. For tartaric acid, assuming L to be similar to that of sucrose, a few millimeters, q is also on the order of 10^{-5} C/m^2 . For muscovite, since the operating speed of the rotator is approximately 1 m/s, L attains a pulse width of 1 ms [Fig. 2(c)] times $1 \text{ m/s} \sim 1 \text{ mm}$. Hence q is obtained as $1 \times 10^{-4} \text{ C/m}^2$. For quartz, luminous areas with a size of 5–6 mm are observed [31], which corresponds to L . Hence q is $1 \times 10^{-5} \text{ C/m}^2$. Thus, for both organic and inorganic materials, q is in the range of $10^{-5} - 10^{-4} \text{ C/m}^2$.

We compared the charge density on fracture surfaces with the surface charge densities previously obtained in contact electrification through collision, rubbing, and mixing of granular materials. A summary of these results is presented in Table II in Appendix C. In a single collision of glass particles, the surface charge density ranges from approximately 10^{-6} to 10^{-4} C/m^2 in vacuum [8] and N_2 gas [9], and for nylon particles, the surface charge density is 10^{-5} C/m^2 in air [10]. These densities are consistent with our result within an order of magnitude. For rubbing of organic materials in a vacuum [12], the surface charge density is smaller, approximately $10^{-7} - 10^{-5} \text{ C/m}^2$, with the upper value close to our result. For the mixing of granular materials, electrification has been studied in various contexts, both natural and industrial [1]. We examined $300 \mu\text{m}$ $\text{ZrO}_2:\text{SiO}_2$ particles [16], sand particles simulating the Martian environment [49], volcanic ash [50], and dust devils [51]. Although the surface charge density varies widely, from 10^{-8} to 10^{-5} C/m^2 , the upper range is also close to our result. In rubbing and mixing, charge cancellation due to repeated contact between charged areas could have resulted in a decrease in the surface charge density.

New surfaces are electrically unstable immediately after crack propagation, and charges may be produced on fracture surfaces. Polarization is one of the possible processes that result in the charge production. First, we discuss the piezoelectric effects, which can play a role in producing charge on fracture surfaces when the crystal is asymmetrical [26,52]. Sucrose exhibits a piezoelectric effect with a piezoelectric strain coefficient d of $3.4 \times 10^{-12} \text{ C/N}$ [26]. With the strength T of sucrose set to 0.56 MPa (Appendix A), the charge produced on fracture surfaces is evaluated as $d \times T \sim 1.9 \times 10^{-6} \text{ C/m}^2$, suggesting that the contribution of piezoelectricity to the production of the charge on the fracture surface in sucrose is minor. In contrast, for quartz, $d \approx 10^{-12} \text{ C/N}$ [53] and $T \approx 10 \text{ MPa}$ (Appendix A) result in $d \times T$ on the order of 10^{-5} C/m^2 . Therefore, in quartz, the piezoelectric effect can be crucial to produce a charge on the fracture surface.

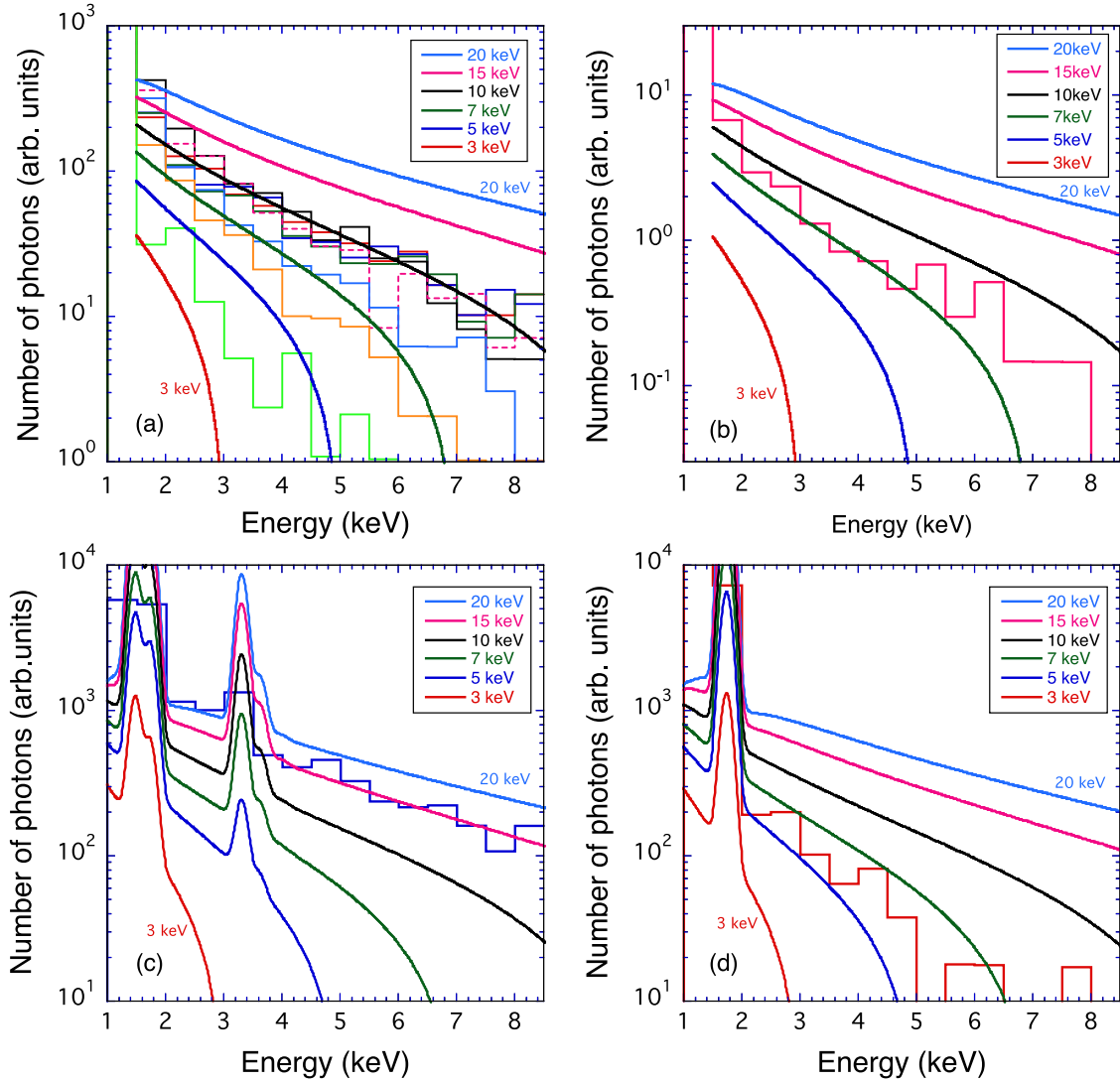


FIG. 5. Comparison of experimental results (shown in Fig. 4) with numerically obtained spectra of X-ray photons generated by electron collisions with (a) sucrose, (b) tartaric acid, (c) muscovite, and (d) quartz [31] at 3, 5, 7, 10, 15, and 20 keV. The numerically obtained spectra are scaled by a constant for comparison with the experimental results.

As another process resulting in polarization, we consider flexoelectricity, which can exhibit large local polarization at the nanoscale [54]. While the piezoelectric effect is limited to materials with asymmetric crystal structures, flexoelectricity can occur in any material. Such characteristic features allow us to apply it to the phenomena involving crack propagation because polarization in localized regions of very small size, such as crack tips, can be very large in any material. In fact, several studies on cracks have been conducted, including the effect on crack propagation [55], direct measurements of polarization at the crack tip [56–58], and bone fracture [59,60]. Polarization that exceeds the surface charge density observed in our experiments has been reported [57]. Thus, although the materials and stress conditions demonstrated have been limited at present, it is suggested that flexoelectricity contributes to the charge generation on fracture surfaces.

If the electrons generated after the crack propagation are bound to the fracture surface, their energy ζ is

approximately $-\hbar^2/(2s^2m)$, where $\hbar$ is the Planck constant over 2π , s represents a characteristic length over which the wave functions decrease from solid to vacuum, and m denotes the electron mass [53]. Assuming s is the surface layer thickness of about a few atomic layers (approximately 0.3 nm), ζ becomes -0.4 eV. On the other hand, the average distance λ between electrons is about $\sqrt{e/q}$. We consider a characteristic potential U defined as $U = e/(\epsilon_0\lambda) = \sqrt{eq}/\epsilon_0$ (eV). When q is large and U exceeds the binding energy ζ , electrons are no longer bound and escape. Therefore, setting $U = \zeta$, we obtain a critical charge density $q_c = (\epsilon_0\zeta)^2/e \sim 9 \times 10^{-5}$ C/m². This result is consistent with the experimental findings.

In this paper we focus on the electrification accompanied by fracture that results in crack propagation and separation of fragments. The electrification can also occur when two identical flat surfaces come into contact and then separate symmetrically [6,7], in which no crack propagation seems to be involved. In such case, the transfer of charge would

be caused by different mechanisms such as net transfer of charged species between two surfaces which have different numbers of the charged species trapped in high-energy states [6] and charge transfer caused by flexoelectric potential difference at nanoscale asperities as a thermodynamic driver [21,22].

It should be noted that air breakdown also limits the surface charge density during electrification in the atmosphere, with a threshold of approximately $3 \times 10^{-5} \text{ C/m}^2$. Since our results are based on the measurements conducted at pressures lower than 10 Pa, which provide sufficiently lower values than the Paschen minimum ($\sim 100 \text{ Pa cm}$), there should be no effect of the air breakdown. On the contrary, in most mixing experiments of granular materials, gas flow fluidizes the particles. This threshold is close to the upper range of the surface charge density observed in mixing experiments. Therefore, mixing experiments of granular materials in a vacuum are necessary for validation.

V. CONCLUSIONS

We investigated visible light and X-rays generated during the fracture of sucrose and tartaric acid as organic materials, and muscovite as inorganic materials, to evaluate the charge density on the fracture surfaces. For sucrose, the pulse width of the visible light indicates that the maximum distance between opposite surfaces during radiation is approximately 3 mm. The X-ray spectra revealed that the maximum energy of the electrons traveling between the fracture surfaces is 10 keV. These results indicate that the charge density on the fracture surfaces of sucrose is approximately $3 \times 10^{-5} \text{ C/m}^2$. All of the samples have the charge density on fracture surfaces of approximately $10^{-5} - 10^{-4} \text{ C/m}^2$. This value is consistent with the surface charge density observed during a single collision and the upper limit observed during rubbing and mixing. Thus fracture can be a cause of contact electrification, though further verification experiments are needed, particularly for electrification in the mixing of granular materials in a vacuum.

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APPENDIX A: MECHANICAL PROPERTIES OF SUCROSE AND QUARTZ

Sucrose. To measure the Young's modulus, strength, and maximum strain of sucrose used in our experiments, we conduct static disruption experiments using a uniaxial-compressive testing machine (MCT-1150, A&D Company, Ltd.) at the University of Occupational and Environmental Health, Japan. The press speed is set to 10 mm/min. We record the load as a function of displacement. The obtained data show that the load is proportional to the displacement. The slope of the force-displacement line corresponds to the Young's modulus E . Using the load F at the point of disruption, the tensile strength T is evaluated as $2F/(\pi ab)$ (Brazilian test), where a and b are the sample dimensions parallel and perpendicular to the load, respectively. Furthermore, the maximum strain δ is calculated as λ/a , where λ is the displacement at the point of disruption. We conduct the measurement for 13 samples. The averaged results, E , T , and δ , are $27.4 \pm 5.9 \text{ MPa}$, $0.56 \pm 0.07 \text{ MPa}$, and 0.024 ± 0.003 , respectively.

Quartz. To measure the tensile strength of quartz used in [31], we carry out the static disruption experiments using a uniaxial-compressive testing machine TENSILON UCT-25T (ORIENTEC) at Institute of Space and Astronautical Science of JAXA. The press speed is 5 mm/min. We record the load M (N) at the disruption and recover the fragments. The high-speed camera shows the position of the fracture surface, and we can identify the surface on the recovered fragments and obtain the area of the surfaces S (m^2). The tensile stress is evaluated as M/S . The average of five samples is $6.3 \pm 4.0 \text{ MPa}$.

APPENDIX B: COMPOSITION OF THE SAMPLES

Here we show a model composition of the samples in mass fraction used in the numerical simulation for X-ray emission in Table I.

TABLE I. A composition of the samples in the numerical simulation.

Element	Sucrose	Tartaric acid	Muscovite	Quartz
H	6.5	4.0		
C	42.1	32.0		
O	51.4	64.0	45.1	53.3
Al			25.4	
Si			19.8	46.7
S				
K			9.2	
Ca			0.5	

APPENDIX C: COMPILATION OF SURFACE CHARGE DENSITY

We have compiled surface charge density data previously obtained in contact electrification through collision, rubbing, and mixing of granular materials. These are summarized in Table II.

TABLE II. Previous results in contact electrification of identical materials.

Process	Charge density (C/m ²)	Material	Ambient pressure	Remarks	Reference
Collision	$10^{-6} - 10^{-4}$	Glass	1.4 Pa		[8]
	$10^{-6} - 10^{-4}$	Glass	1 atm (N ₂)		[9]
	10^{-5}	Nylon	1 atm		[10]
Rubbing	$10^{-7} - 10^{-5}$	PTFE, Nylon, PS	0.01 Pa	$\sim 1 - 100$ pC per ~ 10 mm ²	[12]
Mixing	1×10^{-6}	ZrO ₂ : SiO ₂	1 atm	3×10^{-13} C per 0.3 mm sphere	[16]
	5×10^{-8}	Sand	10^3 Pa (CO ₂)	500 V/10 cm	[49]
	$10^{-6} - 10^{-5}$	Volcanic ash	1 atm	50 μ C/kg, 1 – 250 μ m	[50]
	2×10^{-7}	Dust devil	1 atm	20 kV/m	[51]

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