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Interfacial Ion-Transfer Mechanism for the Intense Luminescence Observed When Opening Self-Seal Envelopes**

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Supporting information:
Video of the opening of seal, illustrating the mechanoluminescence. This material is available free of charge via the Internet at http://pubs.acs.org

Abstract

The unusually intense luminescence (commonly called triboluminescence) observed when opening self-seal envelopes has been studied using spectroscopy. Emissions from gas-phase species due to electrical discharge were observed, which in the case of air consists of vibronic transitions of $N_2(C^3\Pi_u$–$B^3\Pi_g)$ in the ultraviolet (UV) region (280–400 nm). However, the major cause of the intense blue luminescence (around 435 nm) is attributed to optical brightening agents added to the white paper. The results show that the emission from the brightening agents is caused by two mechanisms: (i) fluorescence due to excitation by the UV light from the gas discharge, and (ii) non-optically, by electron transfer. The electrical discharge results from contact electrification; the charge transfer is promoted by migration of hydroxide anions out of the paper into the wet latex-adhesive during drying.

1. Introduction

The emission of light in the absence of heat—luminescence—has fascinated man for many centuries. In times gone by we relied less upon artificial light, and it's probable that instances of luminescence around us were more noticeable.[1] Luminescence lurks where we least expect it, and it may have caught a number of us by surprise to find flashes of blue light emanating from our mail when opening an envelope. Not any type of envelope, we mean specifically the common self-sealing type that uses latex adhesive on two flaps that are pressed together to form the seal. If opened with any degree of quickness, the luminescence can be quite bright, even visible to an eye that is not adapted to the dark.

The self-seal envelope was patented in 1936 by William Hohwart.[2] The sealing part of a self-seal envelope is made up of two flaps of paper, each coated with a latex adhesive, see Fig. 1. The flaps are pressed together to give a paper–adhesive–paper seal. When the paper flaps are separated, the seal is broken, and the luminescence can be observed at the point where the two flaps separate. The phenomenon of luminescence when opening an envelope is closely related to that observed when peeling a roll of sticky tape (such as "Scotch tape[®]" or "Sellotape[®]").[3, 4] The mechanism behind luminescence of peeling tape has been picked apart by Dickinson *et al*.,[5-8] and indeed continues to surprise with the recent report that the peeling also produces X-rays.[9] In the case of self-seal envelopes, however, the luminescence is significantly brighter, and the keen observer may wonder whether stress or fracture of the thin strands of latex glue might have something to do with this (Fig. 1d).

The luminescence observed when peeling adhesive tape is caused by small electrical discharges in the gas between the separating surfaces.[6] This luminescence has often been classed as *triboluminescence* (TL), although we note that the *tribo* prefix implies friction or rubbing. The term *fracto-emission* has

also been applied, in recognition of the fact that both particles and radiation are emitted.[10] From an historical and etymological perspective, the term *electroluminescence* might seem more appropriate; this class of luminescence used to include gas discharge, but is now almost exclusively used to describe luminescence from charge recombination in solid-state materials.[1] A general term to describe luminescence on peeling adhesive is *mechanoluminescence* (ML); literally, luminescence from mechanical action.

We have been unable to locate any systematic study or details of the mechanism for ML from self-seal envelopes in the literature. There are, however, numerous notes of observations of the phenomenon and speculation about its origin; the effect is often simply explained as "triboluminescence". In this report we aim, therefore, to clarify the reasons why the luminescence is unusually strong, to identify the emitting species, and to identify unique chemical and physical characteristics of the mechanism. We show that the origin of the luminescence can be traced to hydroxide anion charge transfer during manufacture of the adhesive seal. As we shall discuss below, there has been quite some controversy surrounding mechanisms for contact electrification between insulating materials.[11-14] The present results contribute a readily available and visually stimulating example of ion transfer during interfacial contact.
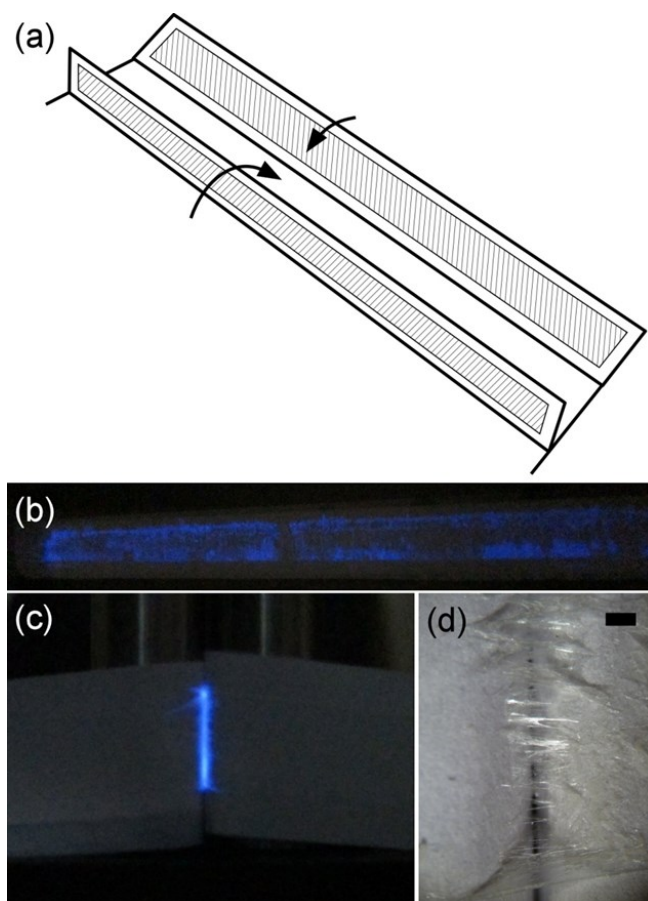
← *Fig. 1.* (a) Schematic diagram showing the layout of a self-sealing envelope: two flaps are folded with the latex adhesive coatings (shaded strips) pressed onto each other. (b) Image of the mechanoluminescence (ML) taken by peeling open the seal from end-to-end as a strip, lengthwise (Canon PowerShot S90, ISO 3200, F2.0, 2 s exposure). (c) Image of the ML at the point of separation of the two flaps (1/4 s exposure). (d) Image taken in white light, illustrating the thin threads of latex adhesive observed when opening the seal (scale bar = 1 mm).

2. Experimental

Measurements of ML spectra were carried out using a spectrometer (Acton 320PI, Princeton Instruments). Light was collected onto the end of a 1 m UV fiber-optic bundle, which was coupled to the entrance slit of the spectrometer. A grating with 300 grooves mm^{-1} was used, giving a wavelength range of 530 nm. The detector was an intensified charge-coupled device (ICCD) array. Wavelengths were calibrated against gas discharges in air and argon. The sensitivity of the system peaked in the deep UV (200–250 nm) and was broadly flat (within a factor of 2) in the UV–visible region (250–650 nm); therefore, the spectra presented here have not been corrected for sensitivity. The entire light-collection setup was covered by blackout material, and the experiments were conducted in a darkened room. Spectra of the ML were obtained by integrating the emission over sets of 5 s, at 95% gain of the detector. Spectra were also taken under inert-gas and hydrocarbon-gas atmospheres (1 bar pressure) using a plastic bag to enclose the light-collection optics. The bag was flushed 3 times before re-filling, and the envelope was sealed under the new atmosphere to reduce trapping of air in the seal.

A range of standard self-sealing envelopes from different sources were tested, and all were found to give the same results qualitatively. The results shown below were obtained with white (Q-Connect KF3454, DL size) and brown (Viking Q2D-2075814, C5 size) self-seal envelopes. It should be noted that self-seal envelopes have a shelf-life due to degradation of the adhesive; fresh envelopes give the best results.

To measure the transient flow of charge during the opening process, electrical probes were attached onto the paper surface of each flap, in line with the seal and opposite each other.[15] The probes consisted of the bare ends of the internal strands of BNC cable (RG58C) connected directly into separate channels of a digital oscilloscope, with the shielded strands grounded commonly (input impedance = 50 Ω). Voltage transients were recorded during rapid opening of the seal. The response of the probes *in situ* to streams of positively or negatively charged particles was verified using a piezoelectric static-discharge gun (Milty, Zerostat 3).

3. Results

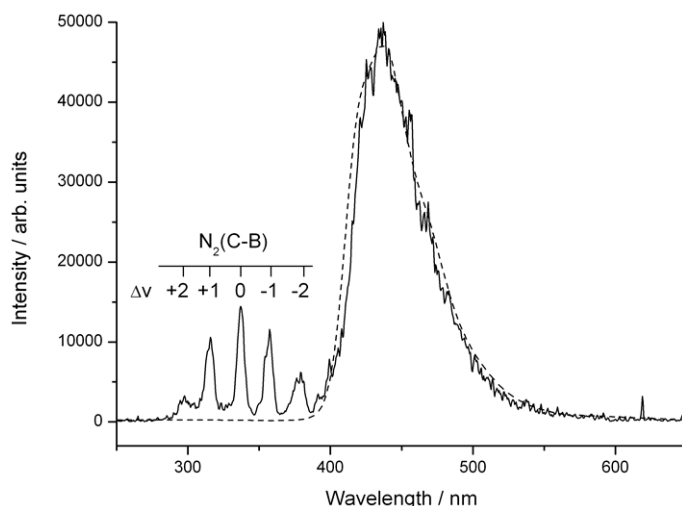### 3.1 Mechanoluminescence

The ML on opening a self-seal envelope is shown in Fig. 1, and also as a video (see Supporting Information) taken using only a compact digital camera (Canon Powershot S90). From Fig. 1(c) and the video, we see direct evidence of "light piping" where the luminescence is guided through latex fibers as the seal is broken; this was first noted by Ma *et al*. for adhesive tape.[6]

The ML spectrum recorded from opening a white self-sealing envelope is shown in Fig. 2, along with the fluorescence emission spectrum of the paper, excited in the UV at 300 nm. In the UV region (< 400 nm) the ML spectrum shows peaks characteristic of vibronic transitions of N_2 ($C^3\Pi_u$–$B^3\Pi_g$).[16, 17] It can be seen, however, that the ML is dominated by the visible emission from the paper, which peaks at *ca*. 435 nm (blue light). The sensitivity of the human eye to UV light varies from person to person: in general, the eye is insensitive to wavelengths < 390 nm. We conclude, therefore, that the visible (blue) emission from the paper is the major cause of the unusually strong luminescence perceived when opening a self-sealing envelope.

The blue emission can be traced to the optical brightening agents (OBA), such as triazinylaminostilbene dyes (Fig. 3), which are added to paper in order to enhance its white appearance. The effect of the brightening agent is demonstrated by comparison to the ML spectrum obtained when opening a brown-paper envelope, see Fig. 4. For brown envelopes we see only lines corresponding to the N_2 emission, and no strong feature in the blue. In addition, we see emissions from N_2^+ ($B^2\Sigma_u^+$–$X^2\Sigma_g^+$),[16, 17] confirming that ionization of air takes place. We note that it is still possible to see, by eye, the ML associated with opening a brown or manila envelope, although it is significantly weaker because the N_2 and N_2^+ emissions > 400 nm are less intense. Some brown envelopes made with recycled paper contain strands of white paper that can be seen to fluoresce under a UV lamp; these envelopes were found to give a stronger ML.

Fig. 2. Mechanoluminescence spectrum recorded from peeling open a white self-sealing envelope (solid line). A fluorescence emission spectrum of the paper excited at 300 nm is also shown (dashed line). In the region < 400 nm, the ML spectrum shows peaks corresponding to C(v')–B(v'') vibronic transitions of the N_2 molecule,[16, 17] these are labelled in groups according to $\Delta v = v' - v''$. In the region > 400 nm, the ML is dominated by emission from optical brighteners in the paper.
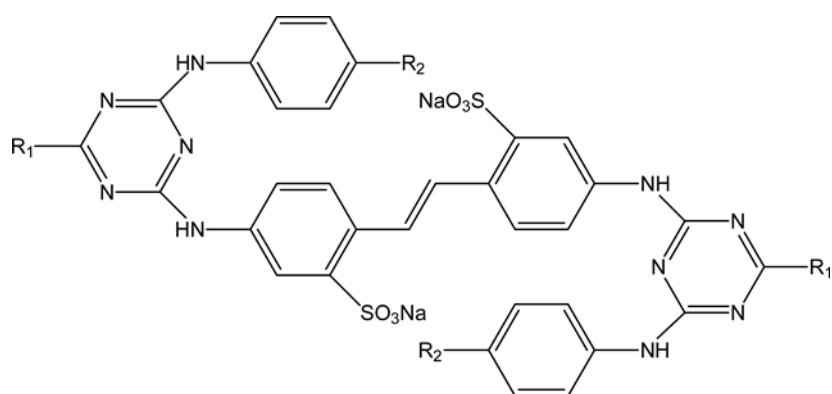
Fig. 3. General structure of optical brightening agents based on bis(4,4′-triazinylamino)stilbene-2,2′-disulfonic acid, commonly used in white paper manufacture.[18] Common functional substituents include R_1 = N(CH$_2$CH$_2$OH)$_2$; both disulfo (R_2 = H) and tetrasulfo (R_2 = SO$_3$Na) derivatives are used.
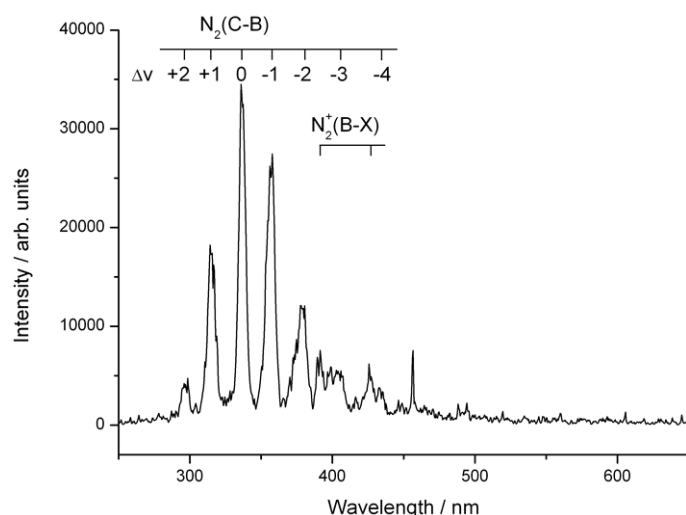
Fig. 4. Mechanoluminescence spectrum recorded from peeling open a brown-paper self-sealing envelope. By comparison to the white envelope (Fig. 2), no blue emission from the paper is observed. Peaks due to C–B vibronic transitions of N_2 and B–X vibronic transitions of N_2^+ are labelled.[16, 17] The sharp spike at 465 nm is an artefact.

The N_2 lines shown in Figs 2 and 4 are often seen during fracture of materials in air, e.g., sucrose,[19, 20] and are caused by electrical discharges resulting from the separation between positive and negative charges on the newly formed surfaces.[20-23] Luminescence due to fracture of crystalline solids is known as _fractoluminescence_, although still commonly referred to as triboluminescence. The charges formed are piezoelectric in origin: it has been observed that many TL-active compounds happen to have non-centrosymmetric solid structures, and are thus capable of being piezoelectric. For sucrose, TL is not

observed when crystals are fractured along a plane parallel to the unique axis because piezoelectric charge across this plane is forbidden by symmetry. For substances that are TL-active but centrosymmetric, the observed TL can be traced to local symmetry breaking, e.g., by impurity compounds.[21-23]

3.2 Luminescence from optical brightening agents

What is the mechanism for excitation of the strong blue emission from the paper? We consider two mechanisms: (i) UV emission from the N_2 discharge excites the optical brightener molecules in the paper, causing them to fluoresce; (ii) the brightener molecules in the paper are excited non-optically.

In Fig. 5(a) we show the ML spectra of white self-seal envelopes opened under argon and neon gas atmospheres. We observe a number of atomic and atomic-ion transitions, most of which are at the red end of the visible spectrum (> 575 nm) and are not capable of exciting blue fluorescence of the brighteners. The relatively high intensities of the atomic lines compared to the N_2 discharge lines in Fig. 2 can be explained by the lower dielectric strength of Ne and Ar, i.e., the electric field strength required to cause a discharge is lower (approximately 25% of N_2).[24] Under argon, the observation of a strong line in the UV (309 nm) means we cannot rule out the UV discharge as a source of excitation of the OBA fluorescence. By contrast, however, we see no strong UV discharge lines under Ne but the blue emission is still very strong. For both Ar and Ne atmospheres we do see some weak lines in the UV that can be assigned to residual N_2; however, the relatively strong intensity of the blue luminescence suggests that N_2 discharge is not the sole cause of the emission of the OBA.

In Fig. 5(b) we show the ML spectrum of a white envelope when opened under ethyne (C_2H_2) gas atmosphere. We note that the dielectric strength of ethyne is only 10% larger than that of N_2.[24] The only feature that can be reliably identified within the noise is the blue luminescence of the OBA molecules. There are hints of luminous activity in the region 300–350 nm, but no progressions could be identified for N_2. We made extensive comparisons for known electronic transitions of OH, CH and C_2, but none of the noisy spikes seen in the region 400–500 nm match to these species unambiguously. The observation of the strong blue luminescence in the absence of UV gas-discharge lines is strong evidence that the OBA can be excited non-optically. This excitation might occur by charge transfer to or from the dyes. However, given the high voltages and discharges involved, it is probable that the dyes are excited by electron bombardment.
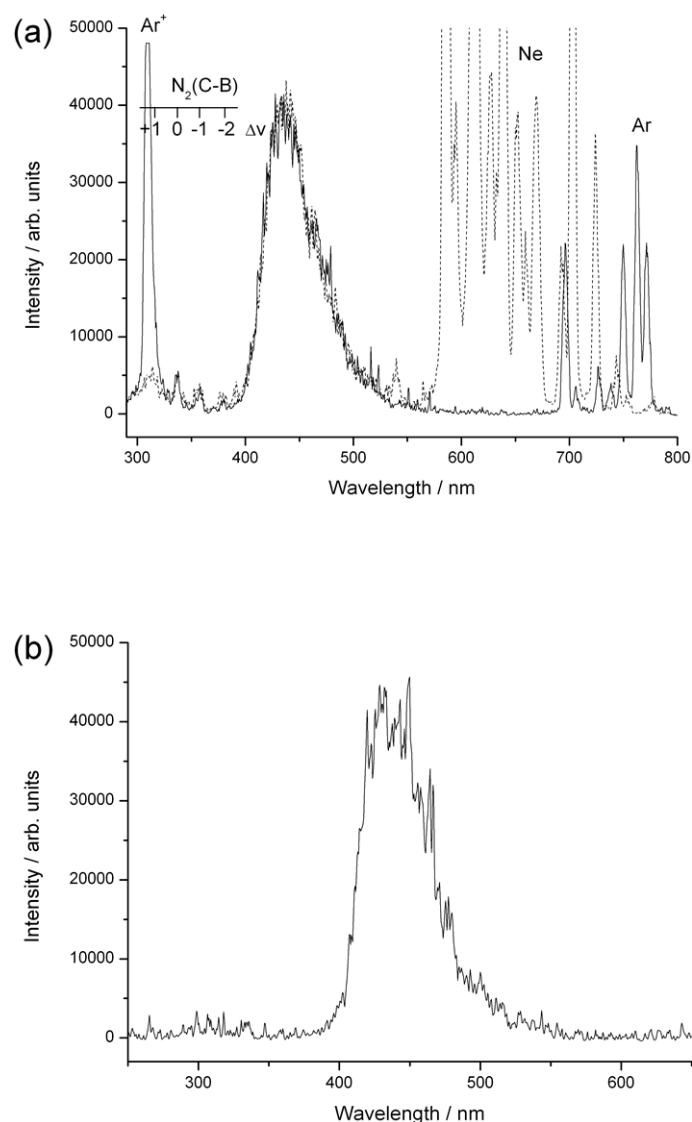
Fig. 5. Mechanoluminescence spectra recorded from peeling open white self-sealing envelopes under various gases: (a) argon (solid line), or neon (dashed line), and (b) ethyne (C_2H_2). Blue emission from the paper (peak 435 nm) is observed in all cases. In (a), the plots have been scaled to give the same intensity for the peak at 435 nm. The largest peak in the neon spectrum is at 594.5 nm (intensity at 2.41×10^5 on this scale) and is attributed to the $3p(^2[3/2])–3s(^2[3/2]^0)$ transition of Ne. Under argon, the peak at 309 nm can be attributed to the $4d(^2D_{5/2})–4p(^2P^0_{3/2})$ transition of Ar^+. Some weak peaks due to residual $N_2(C–B)$ can be seen under Ar and Ne. Peaks > 575 nm are attributed to Ar or Ne (various np–ns transitions). In (b), under ethyne, only the blue OBA emission is identified clearly. Peaks due to residual N_2 in the region 300–400 nm and transitions resulting from CH or C_2 could not be identified definitely. The region 200–300 nm was also investigated (not shown) but no emissions were observed.

3.3 Electrification and discharge mechanism

What causes the ML gas discharge when opening self-seal envelopes? By analogy to TL observed on fracturing materials, we might be tempted to consider fracture of the latex adhesive itself. TL of rubber upon stretching and breaking has been reported.[25, 26] While opening the envelope under ambient light, thin threads of latex are observed (Fig. 1d). On close inspection we observe that a number of thin strands break close to an adhesive anchor-point next to the paper. It was also observed that a large number of threads detach from the surface of the paper. The detachment can be verified by painting a thin layer of latex adhesive onto a white paper surface and peeling it off when dry: ML is observed at the point of detachment. We were unable to detect photoemission from stretching and breaking strands of latex in front of a photomultiplier tube.

To measure changes in electrical potential during opening, we attached probes to the paper flaps (as outlined in Section 2). The probes are sensitive to electric fields created by the proximity of nearby potential, and we used this method to give an indication of the sign and relative magnitude of charges formed. The results are shown in Fig 6. Instances of slower build-up of charge followed by sharp discharges are observed, see, e.g., 0.81–0.82 s. In this region of the seal, one probe measured build-up and loss of *positive* potential predominantly, while the other measured *negative* potential. It was noted generally that the paper flap associated with positive potential had lost its adhesive in the region of the probe; the flap that retained the adhesive yielded the negative voltage transients. We conclude that a negative potential builds up on the side of the adhesive, and positive potential builds up on the paper flap opposite. The resulting electrical discharge produces electronically excited molecules such as N_2, which subsequently luminesce.
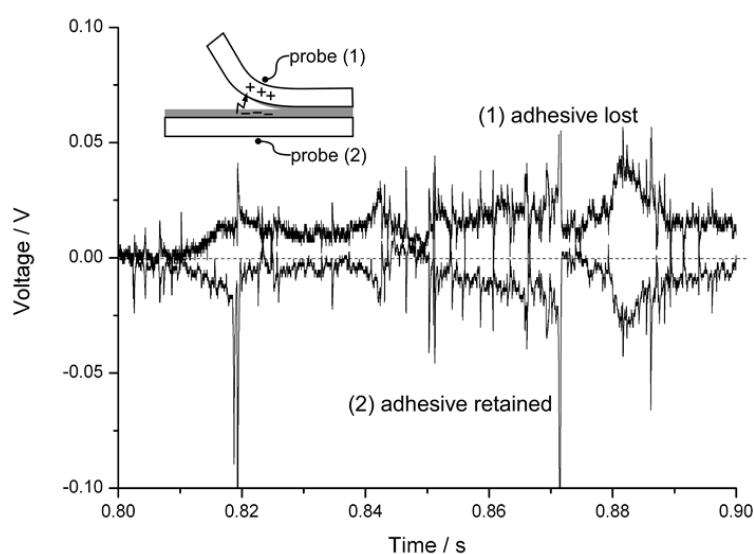
← *Fig. 6.* Voltage transients measured by two electrical probes: one attached to each flap of the sealed envelope, directly opposite each other (for details, see section 2). The flaps were separated rapidly. The traces can be seen to mirror each other; it was noted that the positive voltage is measured on the flap that loses adhesive, and negative voltage is measured on the flap that retains the adhesive.

It has been recognised since ancient times that electric charge can be built up by contacting or rubbing together two materials. This is known as contact electrification or triboelectric charging (triboelectric translates in Greek as "rubbing amber"). Despite many years of study, the mechanism for contact electrification is still hotly debated.[12, 27] Controversy surrounds the nature of the species that transfers the charge.[13, 14, 28, 29] Where both materials are conductors, it is clear that electrons can be transferred. For the case of two insulators, however, two mechanisms dominate the debate: (i) electron transfer via mid-bandgap, defect or surface states, (ii) transfer of adsorbed surface ions or other surface material.[30-33] Recent work appears to favor mechanism (ii); in particular, transfer of OH⁻ via adsorbed water.[12]

The direction and extent of charge transfer between insulators has been found to follow an empirical triboelectric series.[34] At the positive end of the series are materials such as glass, wool, paper; at the negative end are materials such as natural rubber, poly(ethylene), poly(tetrafluoroethylene) (PTFE). When two materials are contacted, the material closer to the positive end of the series will acquire a positive charge, and that closer to the negative end of the series will acquire a negative charge. Paper is nearer to the positive end of the series and natural rubber is closer to the negative end. Therefore, through contact, we might expect paper to receive a positive charge relative to latex adhesive, in accord with our observations (Fig. 6).

To understand fully the mechanism for charge transfer, we consider the structure of the paper and adhesive. Apart from the cellulose base, commercial fine white paper can be a complex mix of fillers, retention agents, strength additives, optical brightening agents, and more.[35] In general, however, most commercially produced paper is left with a net basicity, which among other things acts as a preservative against attack from acids. Fine white paper typically contains a large fraction of calcium carbonate as filler, resulting in a net residual pH of ~8. The latex adhesive used as a pressure adhesive in self-seal envelopes is an aqueous colloid of natural rubber which is preserved with ammonia at an overall pH of ~10. After the adhesive is applied to the paper, we note that drying will cause NH_3 and H_2O to evaporate, driving the following equilibria to the left,

$$NH_3 + H_2O \rightleftharpoons NH_4^+ + OH^-, \qquad pK_b = 4.75, \qquad (1)$$

$$H_2O + H_2O \rightleftharpoons H_3O^+ + OH^-, \qquad pK_a = 14. \qquad (2)$$

The evaporation sets up a pH gradient which promotes transfer of OH⁻ anions from the surface of the paper to the drying adhesive: see Fig. 7. Over time the gradient declines and eventually the H_2O solvent will evaporate whereby no further transfer can take place. The net result is a transfer of negative charge from the paper to the adhesive. When these two surfaces are separated, a potential difference is set up across the gap to the point that breakdown of the intervening gas occurs, and optical emission is observed, as described above.
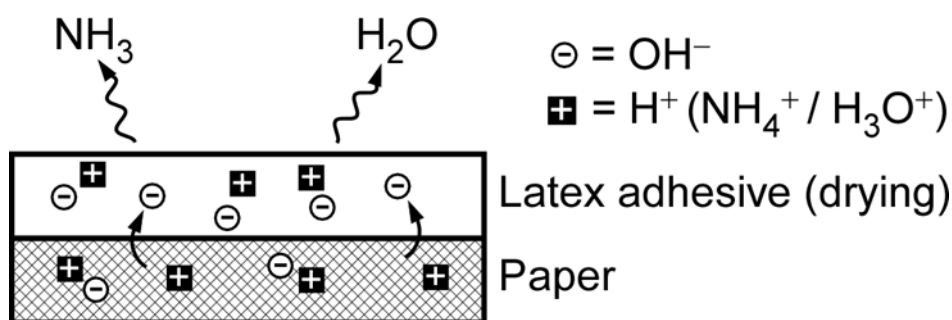
Fig. 7. Illustration showing how hydroxide anions (OH⁻) from the paper are transferred into the wet latex adhesive. The adhesive loses NH_3 and H_2O during drying according to equilibria (1) and (2). Transfer of ions is assisted by wetting of the surface of the paper by the aqueous adhesive. The overall result is that the dry adhesive is negatively charged relative to the paper, consistent with our measurements of charge (see Fig. 5 and text for details).

5. Conclusions

In summary, we have studied the mechanoluminescence observed when opening self-seal envelopes. Separation of strands of latex adhesive from the paper cause electrical discharges and luminescence in the surrounding gas atmosphere. Opening an envelope in air causes predominantly UV luminescence from N_2, which is barely visible to the eye. The reason for the unusually strong luminescence can be traced to excitation of optical brightening agents used in the paper, such as triazinylaminostilbene dyes. Our experiments suggest that these dyes can be excited not only by UV luminescence from the discharge but also directly by electron transfer.

The electrical discharge results from contact electrification between the two insulators of different materials (latex adhesive and paper). The mechanism for contact electrification between two dielectrics has been the subject of much debate recently. In the present case, the charge imbalance is promoted by transfer of hydroxyl anions from the paper to the adhesive during drying. The humble self-seal envelope represents a clear and simple example of contact electrification between dielectric materials that is caused by ion transfer rather than transfer of electrons between mid-bandgap or defect states.

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