Repetitively pulsed gas discharges: memory effect and discharge mode transition

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Abstract: Repetitively pulsed gas volume and dielectric surface discharges have gained growing attention because of peculiar and exciting physics phenomena, high efficiency, high reactivity, and potential to obtain conventionally unachievable plasma properties. Nevertheless, incomplete understanding of fundamental mechanisms renders the repetitively pulsed discharge far less predictable and controllable, where the inherent memory effect and the discharge mode transition are universal challenges. In this topical review, the authors will explore the macroscopic characteristics of the gas gap breakdown and the surface flashover, state-of-the-art mechanisms and dominant agents of discharge memory effects, operation regimes and transitions of the discharge mode, and how waveform parameters affect the pulsed discharge properties. Challenges and potential approaches for further understanding the memory effect and the discharge mode transition in the repetitively pulsed discharge are discussed.

1 Introduction

Recent years have witnessed the dramatic expansion of the applicability of the non-equilibrium discharge plasma into various industrial applications, such as the bio-medical treatment [1], surface modification [2], trace heavy metal detection [3], gaseous pollutant treatment [4], plasma-assisted combustion [5], plasma-assisted flow control [6, 7], nanomaterial synthesis [8]. General requirements for the discharge plasma include the high electron energy, high reactivity, room temperature, arc-free feature in heat-sensitive scenarios, scale-up capability etc. [9–12]. Most parameters of the discharge plasma are sensitive to and controlled by the voltage excitation [9]. The repetitively pulsed voltage has illustrated the exclusive and feasible features for achieving desired plasma properties. Emerging physics phenomena under repetitive nanosecond pulses have attracted growing attention in the plasma science community, such as the runaway electron phenomenon [13], fast gas heating [14], hydrodynamic effects [15], discharge regime classification [16], potential to achieve large-area glow discharge [17], and inherent memory effects [18, 19].

As fundamental and inherent processes in repetitively pulsed gas and dielectric surface discharges, the memory effect and the discharge mode transition are crucial for understanding the discharge evolution mechanism, achieving the predictive control, and modulating chemical reaction environments.

The memory effect is usually ambiguously utilised to explain the ‘following’ trend in the repetitively pulsed discharge and to describe profound effects of previous discharges on initiation and development of subsequent discharges if the pulse repetition frequency (PRF) is sufficiently high. Meanwhile, the general decreasing tendency of the insulation capability under repetitive pulses compared with that under single pulse is believed to be induced by the memory effect [19, 20]. Despite the facilitative memory effect is traditionally emphasised for empirical ‘degradation’ trends (easier to achieve the breakdown) under repetitive pulses, the abnormal and unexpected tendencies have been experimentally observed (e.g. the increment in the number of applied pulses before breakdown with increasing PRF [21]), which, however, have not been satisfactorily explained. Meanwhile, the present framework of proposed memory effect agents (such as free electrons, positive ions, metastable species etc.) is not self-consistent [18]. Several candidates simultaneously exist in the discharge space while their impacts on further discharge developments are either the same or contradictory. Besides, impacts of the pulse timescale and the long-term operation on dominant memory effect agents have not been further investigated and need to be clarified.

The manipulation of the discharge mode is crucial for the controllable chemical reaction and processing (e.g. the glow discharge is frequently desired). The discharge mode transition refers to (i) modulations by pulse parameters and (ii) progressive transitions in the same long pulse train (e.g. formation of the gas gap breakdown and the surface flashover). Progressive breakdown/flashover mechanisms under long-term repetitive pulses have not been fully revealed, specifically why the initial corona discharge eventually triggers catastrophic breakdown/flashover in the pre-breakdown stage and what the insulation thresholds are at a long-term operation. Despite the fact that the progressive ‘corona–glow–spark’ transition is frequently observed under long-term repetitive pulses [16, 22–24], a reversed transition from the spark to diffusive discharge has been reported [25]. The real-time tracking and actively confining desired discharge modes are technically challenging, especially the temporal integration of complex memory effects in the long-term operation and the bounds of discharge instabilities are unclear.

The work aims to provide a review of the memory effect and discharge mode transition in repetitively pulsed gas and surface discharges as well as impacts of waveform parameters, including the state-of-the-art progress, challenges, and potential solutions and approaches. The coverage of this review paper mainly includes the fundamental direct gas gap discharge and the dielectric surface discharge, not presently extended to the plasma-jet source although exciting phenomena have been discovered and universal theories are similar. The whole paper consists of four parts:

(i) Characteristics and developing process of the gas gap breakdown and surface flashover.
(ii) Mechanisms and agents of memory effects in the repetitively pulsed discharge.
(iii) Discharge mode control and transition.
(iv) Effects of waveform parameters on the discharge characteristics.
2 Characteristics and developing process of the gas gap breakdown and surface flashover

Insulation capabilities of the gas gap and the dielectric surface are generally lower under repetitive pulses than those under a single pulse. Meanwhile, a relatively long stressing period (a certain number of applied pulses) is required for achieving breakdown and flashover [19], which suggests that accumulations of memory effect agents are critical for formations of breakdown and flashover.

2.1 General discharge tendencies under repetitive pulses

Shao et al. [19, 21, 26, 27] systematically investigated the breakdown characteristics of gas gaps under repetitive nanosecond pulses (pulse width $t_{pw}$: 30 ns, PRF: 0–1 kHz), and proposed the characterisation system for breakdown features, including the breakdown electric field strength, breakdown time lag, repetitive stressing time, and number of applied pulses before breakdown. Non-linear dependencies of the breakdown electric field strength and the repetitive stressing time on PRF [19, 21] and a negligible breakdown polarity effect [26] were discovered, which were qualitatively attributed to accumulations of metastable and excited species. In Fig. 1a, the breakdown time lag generally decreases with increasing PRF; meanwhile, a remarkable deviation from the traditional streamer criteria existed in the breakdown curve as shown in Fig. 1b [19]. Pai et al. [28] demonstrated in Fig. 2 that the number of consecutive pulses ($t_{pw}$: 10 ns, PRF: 1–30 kHz) for achieving a spark breakdown in a pin–pin gas gap was sensitive to the voltage amplitude; however, hardly dependent on PRF from 1 to 30 kHz.

Zhao et al. [29] investigated the evolution of corona discharge dynamics in high-pressure nitrogen under long-term repetitive sub-microsecond pulses ($t_{pw}$: 800 ns, PRF: 0.02 Hz–2 kHz). A special data logging method was utilised based on the segmented memory technique to capture the evolution trend of the emission light intensity of corona discharges in a long-term pulse train (conventionally 4000 pulses). A three-stage evolution pattern was discovered as illustrated in Fig. 3a under positive repetitive pulses [29], consisting of the intermittent mode at low PRF, the transition mode at medium PRF, and the successive mode at high PRF. In the intermittent mode, the interval pulse number $\Delta N$ between consecutive corona discharges unexpectedly increased with increasing PRF in Fig. 3c. The spark breakdown rapidly occurred in the successive mode, and the envelope curve of the number of applied pulses before breakdown generally incorporated a fast decreasing edge and a relatively steady stage (except for 0.4 MPa) in Fig. 3b. The evolution pattern of the corona discharge challenged the traditional metastable-species-dominated memory effect since the monotonically facilitative influence on subsequent discharge development was enabled by metastable species. A space-charge-dominated memory effect was discussed based on electron depletion processes in voltage rising/trailing edges and the impact of the space–charge region on the spatial distribution of the electric field strength [30].

The dielectric surface flashover illustrated similar tendencies compared with the breakdown of a gas gap. Declining trends of the number of applied pulses before flashover with PRF were demonstrated by Ran et al. [20] for flashover characteristics of a cylinder epoxy resin insulator in SF$_6$. Stone et al. [31, 32] presented the ageing process and electroluminescence of an epoxy insulator stressed by unipolar repetitive pulses. The number of applied pulses to initiate a tree increased with increasing PRF, which was devoted to the necessary period for the dissipation of injected space charges [32]. Besides, profound degradations of surface properties have been observed under intensive repeated surface discharges [33].

2.2 Empirical formula for pulsed gas breakdown

The empirical formula is important for demonstrating the general impacts of variables and applicable for practical insulation design. However, most of the current researches focus on the plasma chemistry and hydrodynamic properties after the initiation of the repetitively pulsed discharge rather than the breakdown/flashover insulation criteria. Besides, the gas gap breakdown characteristics under repetitive pulses are more complicated than those under a single pulse. Insulation characteristics of typical structures need to be further investigated and associated empirical formula is not sufficiently available.

Martin [34], Mankowski et al. [35], and Shao et al. [21] summarised and proposed empirical formulae for the gas gap breakdown under single and repetitive pulses. Martin’s formula is applicable for a wide range of the pulse timescale (single pulse, from nanosecond to microsecond) and a wide spectrum of gas compositions. Mankowski et al. [35] extended Martin’s formula to the sub-nanosecond regime. Shao summarised the breakdown characteristics of repetitive discharges under different conditions.
characteristics of a plane–plane structure in nitrogen and air under repetitive nanosecond pulses (when PRF is higher than 100 Hz):

\[
\rho \tau = 97800 \left( \frac{E}{\rho} \right)^{-3.44}, \quad \text{Martin}
\]

\[
\rho \tau = 0.9 \left( \frac{E}{\rho} \right)^{-2.25}, \quad \text{Mankowski}
\]

\[
\rho \tau = 0.78 \left( \frac{E}{\rho} \right)^{-2.14}, \quad \text{Shao}
\]

where \( \rho \) is the gas density in g/cm\(^3\), \( \tau \) is the breakdown time in seconds, and \( E \) is the mean electric field in kV/cm.

When compared with the gas gap breakdown, the surface flashover voltage is affected by more external factors, such as the surface roughness, surface trap distribution, electrode contact condition. Martin et al. [36] proposed the empirical equation for the pulsed surface tracking voltage

\[
L = 1.6 \cdot \left( V - 5.5 \cdot \delta^{1/3} \right)^{1/4}
\]

where \( \delta \) is the distance tracked in cm, \( V \) is the peak voltage in kV, \( \delta \) is the thickness of mylar in 10\(^{-3}\) inch, and \( t \) is the time width of the pulse at 87% in a microsecond. Laghari [37] summarised an empirical formula for the surface flashover voltage

\[
V_f = 12.4 \cdot \left( \frac{K_s}{K_c} \right) \cdot \frac{\ln(V_{BD})}{\ln(V_f)} \cdot V_{BD}
\]

where \( K_s \), \( K_c \), and \( \epsilon_i \) are the parameters related to the surface roughness, the gap distance, and the relative permittivity, respectively. However, current investigations on flashover characteristics under repetitive pulses mainly focus on the dependencies of general discharge tendencies on external parameters [18, 20, 32], and available data is not sufficient for deducing the empirical formula.

Importantly, in contrast to the single pulse, both the breakdown and flashover tend to occur after a certain number of pulses under repetitive excitation. Therefore, the curve correlating the voltage level and the number of pulses before the breakdown is more feasible to represent the insulation capability than only the voltage level.

2.3 Similarities and differences between volume and surface streamers

A volume streamer and a surface streamer occur in the gas phase without and with the presence of an insulator, respectively. Major similarities and differences between volume and surface streamers are briefly summarised.

Discharge processes in a volume streamer also exist in a surface streamer, such as the ionisation, recombination, and excitation. Dominant memory effect agents discussed in a volume streamer are also crucial in a surface streamer, such as the metastable species, free electrons, and space charges. Nevertheless, a surface streamer exhibits distinct features compared with a volume streamer because of the presence of a solid insulator.
(i) Surface streamer characteristics and surface flashover criteria are affected by more factors than the volume streamer, such as the electric field distribution, insulator shape, surface condition, surface charges, surface defects, contamination [37, 38]. For example, a volume streamer conventionally initiates from tips or edges of electrodes; nevertheless, a surface streamer could start to develop from a middle position, which is governed by the distribution of the electric field strength [39].

(ii) Impacts of surface charges (long decay time constants up to several hours) on subsequent surface streamers are indispensable, including the significant distortion of the electric field distribution [40] and higher availability of seed electrons [41]. Experiments provide shreds of evidence that minimum deposited surface charges are sufficient to impact flashover characteristics [42]. Deposition characteristics of surface charges under various voltage types have been extensively studied as a fundamental high-voltage insulation issue [43, 44]. Deng et al. [40] reported that the length of the second surface discharge dramatically decreased if the second voltage pulse had the same polarity and explained this trend based on the effect of surface charges on the electric field distribution. Besides, the back discharge is frequently observed in the surface discharge at the falling edge of a negative pulsed voltage, which is related to the locally reversed electric field at the electrode edge [18, 45, 46]. Although the binding energy of intrinsic electrons in the solid dielectric surface is as high as 10 eV, the depth of the surface trap centre is conventionally around 1 eV, which enables frequent trapping and detrapping processes [41].

(iii) More physical processes are involved with the presence of an insulator in the repetitively pulsed discharged. Surface charges affect the propagation of subsequent surface streamers by introducing additional electric field components [18]. Meanwhile, diffusions of charged and metastable species are physically bounded by the dielectric surface. Besides, metastable species will be further lost at the dielectric surface. Akishev et al. [47] suggested different current sources for volume and surface streamers. The current flowed into the volume streamer body through the streamer head, however, through the lateral area in a surface streamer. Furthermore, Allen and Mikropoulos [48] proposed that a surface streamer consisted of two components, i.e. the ‘surface’ and ‘air’ components, and a competing mechanism existed between a higher ionisation efficiency related to the photon electron emission and a higher attachment loss to the insulator surface.

3 Mechanisms and agents of memory effects in the repetitively pulsed discharged

Dominant memory effect agents are significantly affected by many factors, such as the voltage pulse waveform, gas pressure, gas composition, discharge history/mode, electric field inhomogeneity, and “pulse off” period. Technically, all leftovers or variations of discharge conditions by preceding discharges have potentials to become candidates of memory effect agents and affect the inception and development of subsequent discharges, although their impacts are inconsistent. Metastable species, free electrons, residual conductivity, positive ions, gas heating, and surface charges (with the presence of a dielectric surface) are usually emphasised in previous theoretical and experimental investigations. Memory effect agents, their influential mechanisms, and decay pathways have been summarised by Zhao et al. [18] for understanding repetitively pulsed gas and surface discharges.

3.1 Metastable species

Acker and Penney [49], Hartmann and Gallimberti [50], and Shao et al. [19] proposed that high-density metastable species were the dominant memory effect agents because of the super-elastic collision, the extra energy gain, and relatively long lifetimes concerning the conventional “pulse off” period. Acker and Penney [49] measured the propagation time of a positive streamer under repetitive pulses based on a photomultiplier (PMT) array. The propagation velocity of subsequent streamers was significantly higher than previous ones inside the same ‘metastable trail’ as shown in Fig. 4, which was related to lower ionisation thresholds of metastable species [49]. Hartmann and Gallimberti [50] suggested that the lower ionisation threshold could not adequately explain the streamer development mechanism. It was proposed that metastable species acted as energy reservoirs and enabled electrons to undergo super-elastic collisions and a streamer energy balance formula was summarised. Simek [51] experimentally determined the temporal evolution of the density of metastable species [49]. Hartmann and Gallimberti [50] discussed a multi-stage profile of the classical ‘double-pulse’ method in low-pressure N₂-O₂ mixture. Nijdam et al. [53] discovered that the development of the second streamer experienced several distinct stages with increasing the pulse interval time as illustrated in Fig. 5a. If the pulse interval time was extremely short (e.g. less than several hundred nanoseconds), the second streamer would continue where the first one stopped at the end of the first voltage pulse (reignite). If the pulse interval time was slightly longer (e.g. several microseconds), the second streamer occupied new channels and preferably developed at edges of previous streamer channels instead of the continuation. The number of new streamer channels increased and the second streamer discharge was eventually independent of the previous one after a long enough pulse interval time (e.g. several dozens of milliseconds). The critical pulse interval time of each stage was sensitive to the gas composition and related to the electron loss mechanism. Nijdam et al. [53] emphasised that the relatively high electron density at edges of previous streamer channels guided subsequent streamers and the high residual conductivity prevented the second streamer from penetrating inside old trails. Li et al. [54] discussed a multi-stage profile of the second streamer length (Fig. 5b) and proposed that the extension of the second streamer length resulted from the earlier discharge inception. The facilitative impact of residual free electrons on initiations of subsequent streamers was also demonstrated in Tholin and Bourdon [22] simulation.

The initial distribution of free electrons before the next voltage pulse is affected by the gas composition and pulse waveform. Free electrons are directly left from previous discharge (e.g. in high-purity nitrogen) or detached from negative ions (e.g. in N₂-O₂ mixture and SF₆) if the electric field exceeds a threshold value [55]. Remnant free electrons quickly diffuse and recombine in the ‘pulse off’ period; however, negative ions reside around previous channels for a relatively long time. Furthermore, free electrons are swept away if the next pulse rising rate is too low, which is
Net space charges affect the spatial distribution of the electric field strength. In contrast to the high mobility of free electrons, the mobility of positive ions is much lower by a factor of several magnitudes. Kazemi et al. [57] emphasised the effect of residual ions on the electric field distribution to explain the second lower streamer current in a wire–pipe electrode system. The second streamer current gradually increased with increasing the pulse interval time, and recovered to the same level of the first streamer when the pulse interval time was approximately longer than 2 ms. Zhao and Li [30] superimposed an extremely low DC bias voltage on long-term repetitive sub-microsecond pulses to intentionally manipulate the position of a space–charge region and verify the effect of space charges on breakdown characteristics. Fig. 6 presents that breakdown curves of a rod-plane gas gap significantly and non-linearly varied with increasing the amplitude of the DC bias voltage [30]. The number of applied pulses before breakdown increased before saturation in the high-PRF region with increasing the amplitude of the DC bias voltage; however, initially decreased and then increased in the low-PRF region. Meanwhile, the breakdown regime extended towards the low-PRF direction. The formation of a positive space–charge region in high-pressure nitrogen was probably related to electron depletion in pulse rising and trailing edges, which was supported by the transient corona discharge under a pulsed voltage [30], the electric field step phenomenon observed by Zeng et al. [58], and the discharge detour phenomenon observed by Hayakawa et al. [59]. Variations of breakdown characteristics were related to the reduction of space-free electrons in the high-PRF region and the spatial drift (low DC bias voltage) or diminishment (high DC bias voltage) of the space–charge region in the low-PRF region [30].

Low-mobility negative ions can provide a copious source of initial electrons through collisional detachment under the following pulsed voltage, especially in electronegative gases (e.g. O₂ and SF₆). MacGregor et al. [60] emphasised the effect of negative ions on the recovery curve of a high-pressure gas closing switch. A low-amplitude DC bias voltage can effectively improve the recovery rate by removing the remaining negative ions.

### 3.4 Gas temperature

The channel temperature can be dramatically elevated up to 1000–2000 K within a time scale of 10–100 ns in the nanosecond repetitively pulsed spark discharge because of a fast gas heating mechanism at high over-voltage ratios [15]. Consequently, the reduced electric field significantly increases due to a lower gas density induced by the channel expansion in the ‘pulse-off’ period. A ‘self-focusing’ tendency of the spark channel was imaged by Starikovskiy et al. [61] and simulated by Nikipelov et al. [62] as presented in Fig. 7.

### 3.5 Dielectric surface charges

In the presence of a dielectric surface, interactions and bidirectional transformations between space and surface charges significantly contribute to characteristics of a surface streamer [18]. The binding energy of surface charges is usually around 1 eV, which indicates that surface trapped electrons are expected to be released and get involved in subsequent surface discharges [41]. Höff et al. [63], Nemschokmichal et al. [41], and Akishev et al. [66] also investigated the volume and surface memory effects in the dielectric barrier discharge (DBD). The important role of the electron desorption from the cathodic dielectric surface was validated by Nemschokmichal et al. [41]. Laser-triggered discharge results and fluid simulation trends matched well in Fig. 8 [41]. A higher pre-ionisation resulted in a lower breakdown voltage, a lower discharge current, and an earlier discharge inception moment. Li et al. [65], Wang et al. [66], and Yao et al. [67] also presented the significant impacts of surface trap features and the secondary electron yielding coefficient in DBD.

In a classic DBD configuration, both the spatial and temporal memory effects appear. In a conventional AC-voltage-driven DBD operating in the filamentary regime where numerous microdischarges (MDs) illustrate spatial–temporal chaotic behaviours, the discharge memory effects incorporate two aspects of reproducibility, i.e. spatial memory and temporal memory [64]. A traditional explanation for the spatial reproducibility of MDs is...
based on the electric field enhancement induced by deposited surface charges [68]. However, Akishev et al. [64] clarified that the remaining conductivity of a preceding channel is responsible for the spatial memory because of the redistribution of surface charges (deviation from the Gaussian assumption) and the consequent appearance of strong radial electric fields at the periphery of a plasma column base (surface breakdown). Residual surface charges help to retrieve the same plasma column; nevertheless, do not determine the local position of MDs, which is also supported by the small position jitter of subsequent MDs. Furthermore, the temporal scattering in the inception phase of MDs is affected by the stochastic nature of the distribution of the residual surface charge.

Direct characterisations of the distribution and evolution of surface charges are beneficial for interpreting the effect of surface charges on the initiation and propagation features of subsequent discharges. The surface charge distribution can be diagnosed and evaluated by the Lichtenberg diagram [69], electrostatic probe [70, 71], Pockels crystal [72, 73], and associated electric field [74–76]. The measurement of the associated electric field provides online monitoring and spatial–temporal information despite the indirect characterisation of surface charges. The electric field can be measured based on the electric-field-induced second harmonic generation [77], the four-wave mixing method [74], optical emission spectroscopy [75], capacitive sensor [76], and electro-optical sensor [58]. Winters et al. [76] monitored the surface potential decay in the continuous repetitively pulsed surface discharge based on the capacitive sensor with different pulse polarities.

4 Discharge mode transition in the repetitively pulsed discharge

Discharge mode (e.g. corona discharge, glow discharge, spark discharge) is crucial for tailoring the plasma chemistry and hydrodynamic effects. The transition characteristics and mechanisms provide insights into actively confining a favourable or preventing an adverse discharge plasma environment especially in a long-term operation.

The operation limit of a discharge mode in the repetitively pulsed discharge is usually controlled and defined by voltage amplitude, PRF, and ambient environment (e.g. gas pressure and gas temperature). Pai et al. [16, 28] investigated the discharge operation regimes and mode transition criteria under repetitive nanosecond pulses ($t_{\text{pw}}$: 10 ns, PRF: 1–30 kHz) in a pin–pin gas gap. The discharge mode sequentially experienced transitions (no discharge–corona–glow–spark) with increasing voltage amplitude and PRF [16]. The lower voltage limit of the spark discharge regime decreased with increasing PRF as shown in Fig. 9 [16]. Ding et al. [78] and Shcherbanev et al. [79] discussed the streamer-to-filament transition in the pulsed surface discharge. The electron density increased by 3–4 orders of magnitude and continuous

![Fig. 7](a) Self-focusing phenomenon in the repetitively pulsed spark discharge. (a) Partial temporal-resolved intensified charge-coupled device (ICCD) images of the spark channel of ten consecutive pulses ($t_{\text{pw}}$: 5 ns, PRF: 1 kHz) [61]. Δ$t$ is the ICCD camera delay with respect to the voltage starting moment, (b) Simulation results of the radial distribution of the electron density [62]. Both figures are directly from literature figures.

![Fig. 8](a) Comparison between the laser-triggered discharge and simulation results [41]. Directly from literature figures (a) Measured voltage, current, optical emission in the 1064 nm laser photo-desorption experiment, (b) Simulated voltage, current, spatiotemporally resolved He(33S) excitation rate with and without 0.1 nC electrons released from the surface.
accumulations of memory effect agents and their profound progressive discharge mode evolution usually exists before an inception and adjustment stage of regimes [16]. N: no discharge, C: corona, G: glow, S: spark. Directly from spectra in ultraviolet and visible ranges appeared during the transition. Under a long-term and waveform-fixed pulse train, a progressive discharge mode evolution usually exists before an equilibrium state in the repetitively pulsed discharge, indicating accumulations of memory effect agents and their profound influences on the discharge regime. A spark breakdown is conventionally achieved after a number of applied pulses, which is necessary for the plasma-assisted combustion; nevertheless, unacceptable for heat-sensitive applications. Naidis [24] simulated the progressive formation of the spark discharge in 0.1 MPa air under repetitive pulses ($t_{\text{pw}}$: 5 ns, PRF: 30 kHz) and proposed the three-stage evolution profile as illustrated in Fig. 10: initial heating in the low current stage (Stage I), inception and adjustment stage of the first several spark discharges (Stage II), and quasi-stationary spark discharge stage (Stage III). Similarly, the number of pulses prior to the spark breakdown showed an inverse dependence on the voltage amplitude. Nagaraja et al. [80] developed an integrated theoretical and numerical framework and provided temporal evolution and dynamics of chemical kinetics, energy coupling, gas heating, and generation of active particles excited by long-term repetitive pulses in low-pressure air ($t_{\text{pw}}$: 3 ns, PRF: 1–100 kHz, consecutive pulse number: up to 100 pulses). Tholin and Bourdon [22, 23, 81, 82] systematically simulated external factors on the development of repetitively pulsed nanosecond discharges ($t_{\text{pw}}$: 12 ns, PRF: 1 Hz–100 kHz, consecutive pulse number: up to 7 pulses), including the background gas temperature, hydrodynamics expansion, and external circuit impedance. As illustrated in Fig. 11, the negative discharge occupied almost 50% of the gas gap and the positive discharge just started to ignite at the first pulse [22]. In contrast, positive and negative discharges successfully connected and a glow discharge channel was maintained under several subsequent pulses. The connection time was required to be roughly equal to the voltage pulse width if a glow discharge was desired [23]. In terms of the ‘glow-to-spark’ transition before the spark state, Fridman and Kennedy [83] and Raizer [84] summarised the potential approaches such as the accumulation of metastable species and the positive feedback involving the electron density, gas density, and reduced electric field.

Regarding the progressive formation of a surface flashover from the initial corona discharge, Zhao et al. [18] measured the evolution of the propagation velocity of the surface streamer under repetitive sub-microsecond pulses and discussed mechanisms of the back discharge and the reversed polarity effect of the repetitive working coefficient. The surface discharge configuration resembled the surface needle defect structure in partial discharge studies [46]. Evolutions of surface streamers and flashover regimes were different from the gas volume breakdown, although the majority of discharge tendencies were similar. The development of a subsequent surface streamer was qualitatively influenced by the previous volume charge region and assisted or retarded by surface charges as illustrated in Fig. 12 [18].

In contrast to the traditional scenario where a spark is ultimately maintained in the high-frequency repetitive pulsed discharge, we have provisionally discovered a periodical discharge mode transition sustained by long-term repetitive 20 kV 15 ns voltage pulses (0.1 MPa nitrogen and no gas flow). The repetitive nanosecond pulses were produced based on a hybrid pulse combined topology [85, 86]. In 5 mm cone–cone and cone–plate configurations, the discharge mode experienced a periodical transition at 5–10 kHz excitation rate, i.e. no discharge $\rightarrow$ corona discharge inception $\rightarrow$ corona discharge enhancement $\rightarrow$ glow discharge $\rightarrow$ transient spark discharge $\rightarrow$ dramatic extinction of spark discharge $\rightarrow$ corona enhancement $\rightarrow$ glow discharge $\rightarrow$... The periodical transition process is expected to be governed by the ‘plasma–source’ interaction and will be presented in our future paper through varying the pulse source impedance. Furthermore, Zhang et al. [25] demonstrated the possibility of a reversed mode.
Gas heating mechanism. The discharge plasma chemistry and stability can be tailored based on waveform selections [83, 87, 88].

Impacts of waveform and operation parameters on discharge characteristics in the plasma-jet structure have been systematically reviewed by Lu et al. [9]. Universal physics are applicable. Here we mainly focus on discharge peculiarities at high overvoltage ratios and effects of selected waveform parameters on discharge features associated with memory effect and mode transition.

### 5.1 Discharge peculiarities at high overvoltage ratios

The overvoltage ratio (compared with the DC excitation) generally increases with shortening the pulse rise time because of statistical and formative lags. The overvoltage ratio exceeds 2–3 if the pulse width is reduced to within 10–30 ns [36, 89] as illustrated in Fig. 13 or even becomes bigger than 5 for subnanosecond pulse rise time [90]. Korolev and Mesyats [91] measured the impact of the pulse rising rate on the pulsed corona inception voltage and found that the pulsed corona inception voltage was 2.4 times higher at 3 kV/ns than that at \( \sim 0.1–0.2 \) kV/ns. The electron energy increases with increasing the overvoltage ratio, and the runaway electron phenomenon becomes possible at an extremely high overvoltage ratio [13, 92, 93].

Korolev and Mesyats [91] summarised the peculiarities of electron avalanches developing at high overvoltage ratios based on numerical simulations:

1. The temporal dependence of the number of charge carriers in an electron avalanche illustrated a deviation from the exponential curve as early as 0.4 ns after the avalanche onset (Fig. 14a).
2. Longitudinal and transverse electron distributions over an avalanche suggested the electron cloud was deformed at a large number of charge carriers, which was mainly induced by the internal electrostatic repulsion (Fig. 14b).
3. A significant difference in the electron drift velocity existed between the front side and back side of an electron cloud (Fig. 14c).
4. The critical number of electrons for the ‘avalanche-to-streamer’ transition (AST) decreased by 15–20% at a high overvoltage ratio compared with the classical condition.
5. In a moderate electric field and a short gas gap, the streamer travelling time was negligible, and the discharge forming time was close to the period needed for the charge-carrier number in an avalanche developing to the critical value. Nevertheless, in an extremely high electric field, the lifetime of excited species becomes much longer than \( t_{cr} \), which suggested that emitted photons had a certain delay relative to the ionisation process in an avalanche. Consequently, the discharge forming time will be considerably longer than \( t_{cr} \).
6. The electric field at the cathode surface was possibly enhanced by positive ions in an electron avalanche by a factor of around 20% (background electric field: 100 kV/cm), which was favourable for the field emission at cathode micro-points.

For a conventional AST occurring in a moderate electric field (e.g. around 100 Td), Raether–Meek criterion states that a reasonable estimation of the product of the impact ionisation coefficient \( a \) and the distance \( d \) is 18–20 [84]. However, Montijn and Ebert [94] and Pai et al. [28] suggested that the critical number of charge carriers required for AST decreased at a high reduced electric field, and \( a \cdot d \) remained as 16 when the reduced electric field was approximately higher than 250 Td. This reduction matches well with the prediction of the numerical simulation by Korolev and Mesyats [91].

Kawada and Hosokawa [95] experimentally examined the nanosecond-pulse breakdown of gas-insulated gaps and divided the whole breakdown delay into three steps, i.e. the time required for the AST, the time in which the streamer propagates towards the anode until the connection, and the time in which the channel instability grows. It was observed from the luminosity in the gas gap that a streamer occurred near the cathode surface only a few nanoseconds after the application of the pulsed voltage. Similar to

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Fig. 12: Impacts of volume and surface charges on the propagation of subsequent surface streamers in N\textsubscript{2} [18]. \( E_0 \) is the external electric field. In both figures, \( F_1, F_2, \) and \( F_3 \) are the electric forces on the streamer head generated by the perpendicular component of the external electric field, surface charges, and residual space charges, respectively. The triple point condition is crucial for the initiation of a subsequent streamer. Both figures are replotted from literature figures (a) Multiple electric field components for the second positive surface streamer, (b) Multiple electric field components for the second negative surface streamer.

Fig. 13: Impacts of the pulse width and the product of the gas pressure \( p \) and gas distance \( l \) on the 50% breakdown voltage in air [89]. Rectangular, triangular, and circle data represented the scenarios where the gas distance was equal to 3, 2 and 1 cm, respectively. \( t_p \) was the pulse rise time. Replotted from the literature raw data.

5 Effects of waveform parameters on discharge characteristics under pulsed voltage

Macroscopic and microscopic properties of the discharge plasma are governed and modulated by voltage waveform parameters [9, 10]. Both the rise time and the pulse width determine the overvoltage ratio, the electron avalanche development, streamer morphology, pre-ionisation distribution, discharge instability, and
the prediction by the numerical simulation [91], the streamer travelling time is significantly longer than the AST time $t_{ct}$ by an approximate factor of 6.6 in a 3 cm air gap [95].

The build-up of the positive ion layer near the cathode surface enhances the cathode field. Ivanov et al. [96] and Zubarev and Ivanov [97] investigated the initial phase of a sub-nanosecond pulsed breakdown of a high-pressure gas gap. It was found that the electric field near the cathode surface was significantly increased, which was related to the formation of a charge-depleted region [96]. The locally enhanced electric field near the cathode surface in the high-overvoltage discharge also provides the possibilities of generating a short pulse of runaway electrons and short-wave radiation as suggested by Shao et al. [92], Kostyura and Tarasenko [98], and Bratchikov et al. [99].

Naidis et al. [90] systematically reviewed experimental and computational results of pre-breakdown characteristics at ultra-high overvoltage ratios during the subnanosecond diffusive discharge formation. Series of unique features are expected to be induced by high overvoltages, including (i) the possibility of the dense plasma formation without the preliminary gas ionisation and occurring in subnanosecond breakdown delays, (ii) the possibility of increasing propagation velocities and cross-sections of streamers due to the high electric field intensity, (iii) the generation of runaway electrons which has significant impacts on breakdown processes [90]. Favourable diffusive discharge is suggested to be dependent on runaway electrons, X-rays, and streamers of large cross-sections. Intense gas pre-ionisation will be produced by runaway electrons and diffuse form is resulted from overlapped electron avalanches [100].

Levko et al. [101, 102] and Levko [103, 104] have recently performed a series of simulations of the subnanosecond pulsed gas discharges. Both kinetic and fluid models can predict the shift of Paschen curves minima towards higher pressure and voltage with decreasing the pulse rise time, which correspond to the conditions of electron runaway [92]. The electron velocity distribution function on the left branch of the Paschen curve is non-local both in time and space [102]. Two stages existed for the subnanosecond breakdown in pressured nitrogen, i.e. an initial slow stage of conventional fast ionisation wave and a following fast stage determined by the runaway electrons generation [103]. The fast stage started from the moment when the electric field between the electric field, which is crucial for the plasma hydrodynamic effect and chemical reactions [105–107]. Xu et al. [15] and Rusterholtz et al. [108] demonstrated the formation of a shock wave after a pulsed discharge and the ultra-high heating rate of $5 \times 10^{10}$ K/s ($t_{pw}$: 10 ns, PRF: 10 kHz). The fast gas heating in a N$_2$–O$_2$ mixture mainly follows the two-step mechanism [81]. Firstly, electronically excited nitrogen molecules are generated from the electron impact under the pulsed voltage. Secondly, excited nitrogen molecules are rapidly and efficiently quenched by O$_2$ molecules, followed by the dissociation of O$_2$ molecules. These oxygen atoms thermalise within a few collisions and increase the gas temperature. The energy transfer efficiency is dependent on the reduced electric field [14].

5.2 Effect of the pulse rising rate

The discharge morphology is significantly affected by the pulse rising rate. Hinterholzer and Boeck [109] found that the accumulated launch angle of multiple spark channels in SF$_6$ decreased with increasing the voltage steepness. Komuro et al. [110] simulated the effect of the pulse rising rate on the channel radius and the propagation velocity of a streamer and summarised that a lower pulse rising rate resulted in a lower discharge current, a slower primary streamer, and a thinner streamer channel. Mesyats and Korolev [111] demonstrated that the spatially uniform and multi-electron initiated volume discharge appeared at a higher electric field (100 kV/cm), while shrank into a filamentary channel at a lower electric field (73 kV/cm).

The pulse rising rate also dramatically affects the spatial distribution of space-free electrons (i.e. the actual pre-ionisation condition). A temporary electron depletion region appears near the cathode at the next pulse rising edge because electrons instantaneously drift towards the anode in the pulse rising edge if the electron multiplication before the static breakdown voltage and cathode field emission is neglected [28]. Consequently, a multi-electron initiated uniform discharge benefited from a high-density and uniform pre-ionisation is undermined, and transforms into the single-electron-initiated filamentary discharge at a lower pulse rising rate [91]. Geiman et al. investigated the dependence of the length of the uncomplicated filamentary channel on the pulse rising rate in a CO$_2$/N$_2$/He mixture. As the voltage rising rate decreased, the glow discharge became inhomogeneous throughout the gap, and an uncomplicated filamentary spark channel was observed near the cathode surface [91]. Fig. 15a illustrated that the length of the uncomplicated filamentary channel was shorter at a higher pulse rising rate and always bigger than the calculated length of the temporary electron depletion region [91]. Furthermore, Levatver and Lin [112] and Pai et al. [28] derived the formula of the temporary electron depletion thickness, which was dependent on the voltage rising rate and gas pressure. Besides, Ito et al. [113] proposed a rapid breakdown mechanism under repetitive nanosecond pulses, where the volume charge movement and the surface charge deposition were emphasised. Remnant positive ions from preceding discharges were left behind because of the swift electron drift in the pulse rising edge. As verified in Fig. 15b, significant net space charges (around 30%) had already been accumulated near the cathode before a detectable optical emission [113].

The voltage rising rate controls the shape of the electron distribution function, which largely determines the rates of associated chemical reactions [101]. Iza et al. [114] presented the
possibly prevented. Furthermore, Pai et al. [115] suggested that the evolution of a modification and bio-medical applications and is promisingly pulse width. The selective production of different radicals in the high-temperature environment (typically: 10–100 ns [83]), the ‘glow-to-spark’ transition will be considerably prevented. Furthermore, Pai et al. [16] verified that the high-temperature region (100–300 V [84]). Similarly, as illustrated in Fig. 16, Qi et al. [17] presented a possibility of achieving the atmospheric-pressure large-scale glow plasma sheet by repetitive nanosecond pulses (τpw: 15 ns, PRF: 40 kHz), and discovered that the discharge plasma was simultaneously generated in the whole gap within a 0.125 ns temporal resolution.

5.4 Effect of the voltage polarity

Stream propagation and properties are determined by the voltage polarity because of different ionisation mechanisms. For example, the generation of electron beams and X-rays are usually observed for negatively stressed subnanosecond discharges [90, 117]. Remaining electrons have distinct impacts on positive and negative streamers, critical for the initiation and development of a positive streamer; nevertheless, negligible on a negative streamer [52]. Shao et al. [26] discovered a weakened polarity effect of the breakdown voltage under repetitive nanosecond pulses in a point-plane configuration, which was related to the accumulation of metastable species before the final breakdown.

Takashima et al. [118] compared the surface streamer morphology under positive and negative repetitive nanosecond pulses. Under negative repetitive pulses, the surface discharge exhibited a uniform structure at relatively low PRF, while became strongly filamentary at high PRF. This trend was less dramatic under positive repetitive pulses. In the surface streamer-to-filament transition under single and repetitive nanosecond pulses [78, 79], the streamer-to-filament transition voltage was less dependent on the gas pressure under negative pulses than positive pulses. Meanwhile, the effect of the oxygen concentration on the transition voltage is more evident under positive polarity.

Zhao and Li [30] compared the effect of the voltage polarity on the evolution of the light intensity of corona discharge under repetitive sub-microsecond pulses, and found that the transition PRF from the intermittent mode to the successive mode was much lower under negative pulses than positive pulses. Meanwhile, the allowable repetitive working coefficient (defined as the ratio of the voltage amplitude of applied repetitive pulses to 50% breakdown voltage under single pulse) was higher under negative pulses in a rod-plane structure. For the surface flashover, a reversed polarity effect of the allowable repetitive working coefficient existed with increasing gas pressure, which was tentatively correlated with the corona stabilisation effect, surface electron de-trapping process, and the effect of the background electric field [18].

5.5 Effect of PRF

The repetition rate directly determines the pre-ionisation level of subsequent discharges. Conventionally, a spark breakdown is easier to be achieved at high PRF, where the number of applied pulses before breakdown decreases with increasing PRF as illustrated in Figs. 1 and 5, although abnormal trends occur at high pressures [19, 21, 29].

Huang et al. [119, 120] conducted experimental and theoretical investigations on repetitively pulsed discharges, especially on the impact of PRF on discharge characteristics. In the pin-to-pin MD, a high residual charge density at high PRF tends to reduce the breakdown voltage, the peak power, and the peak electron density [119]. Meanwhile, the rising rates of the electron density and
interdisciplinary fields require one to thoroughly understand the charges during the breakdown period [119]. Furthermore, for a features and fascinating physics phenomena compared with electron temperature are both higher near the cathode than the anode. It was proposed that the distribution of electric field and metastable species densities. In hardly reproducible repetitively pulsed discharges, such as the electric field, electron density, and metastable species densities.

6 Challenges and potential approaches

Repetitively pulsed discharge plasmas have illustrated superior features and fascinating physics phenomena compared with conventional excitations. The dramatic expansion and tremendous potentials of the repetitively pulsed discharge plasma into interdisciplinary fields require ones to thoroughly understand the fundamental mechanisms and tackle technical challenges.

6.1 High-performance repetitive pulse generator

Unfortunately, as the crucial equipment, the repetitive pulse generator is currently far less robust and flexible than traditional AC voltage sources. Critical issues of designing a repetitive pulse generator incorporate the high-voltage switch with a satisfactory repetitive operation capability, the novel circuit topology, and the accurate and fast charging method. Although peculiar and exciting physics phenomena have been experimentally observed and ground-breaking records have been achieved with assistance of high-performance repetitive nanosecond pulse generators, the cutting-edge switches are still rare and not commercialised yet. Besides, the output pulse parameters of most repetitive pulse generators could not be flexibly adjusted. The pulse combining topology is a promising approach to deliver a high-voltage pulse especially for semiconductor switches.

6.2 Characterisation methods and diagnostics

Characterisation methods of the pre-breakdown and pre-flashover stages are critical to reveal differences between the single-pulse and repetitive-pulse excitations. The classical ‘double-pulse’ method enables one to explore the memory effect with the equivalent ‘pulse off’ period down to several hundred nanoseconds. However, the long-term evolution of streamer dynamics and the discharge mode transition could not be effectively obtained by a limited number of applied pulses. Furthermore, present characterisation methods for the gas gap breakdown under repetitive pulses mainly focus on the final breakdown data rather than the evolution process. Besides, the dedicated parameters in the aspect of the insulation capability between the single and repetitive excitations have not been established yet. Several novel characterisation parameters have been recently introduced for the evolution process, including the repetitive working coefficient, the ‘sequence–phase–intensity–density’ diagram, and the corona discharge following coefficient [30]. However, these parameters are still mainly devoted to macroscopic evolution trends. Laser-based diagnostics are urgently required for obtaining fundamental plasma parameters in hardly reproducible repetitively pulsed discharges, such as the electric field, electron density, and metastable species densities.

6.3 Distinguish dominant memory effect agents

Many memory effect agents have the same facilitative or prohibiting impacts. Different memory effect agents were separately emphasised in previous studies. The dominant memory effect agent might change in a long-term operation, e.g. due to the charge accumulation, or different discharge modes. Besides, effects of waveform parameters on dominant memory effect agents and the evolution of discharge mode have not been explored yet.

6.4 Effect of the plasma–source interaction

The plasma–source interaction affects the discharge mode transition, plasma properties, and energy transfer efficiency [116, 121]. Plasma properties could be different under the same voltage waveform while coupled with distinct generator topologies [121]. The suitable waveform for achieving uniform discharge needs to be further investigated (e.g. how short the voltage pulse needs to be). Another important challenge is how to prevent the spark discharge in the long-term operation when the voltage pulse is already sufficiently short. Therefore, the ‘smart’ pulse generator integrated with the data-driven artificial intelligence and feedback control is an interesting approach to actively optimise and modulate plasma features (e.g. tailoring the waveform or modulate PRF before the glow-to-spark transition based on optimised strategies).

6.5 High fidelity and realistic simulation for the long-term repetitive pulsed discharge

Numerical simulations can provide quantitative explanations and validations to the experimental results. However, the numerical simulations of long-term repetitive gas discharge are technically difficult. Requirements for implementing simulations of long-term repetitively pulsed gas discharges are briefly summarised:

(i) Unlike the single streamer discharge, the long-term repetitively pulsed discharges exhibit discharge mode transitions (e.g. gas gap spark breakdown) probably after a certain number of applied pulses, suggesting the crucial role of the self-consistent continuous simulation of the accumulation of memory effect agents.
Simulations of long-term repetitively pulsed discharges encounter many technical difficulties, such as the huge timescale difference between the fast streamer discharge in the ‘pulse on’ period and the slow plasma decay in the ‘pulse off’ period [80], the repeated update of the pre-ionisation condition, numerical errors etc. Specifically, the prolonging tendency of a positive streamer at high PRF before breakdown implies that the simple uniform pre-ionisation assumption throughout the whole gas gap is not applicable in the dynamic pre-breakdown stage [29].

(ii) Many associated applications desire the chemical reactivity and hydrodynamic effect enabled by repetitively pulsed gas volume and dielectric surface discharges. The inherent multi-physics nature requires the involvement of interfaces among different physics processes (e.g. chemical reactions, heat dissipation, wall reaction).

(iii) In the presence of a dielectric surface, the surface charge features should be included. Conventionally, the dielectric surface is regarded as a set of perfect electron absorption centres [122]. Consideration of actual surface properties (e.g. surface charge trap functions) provides more possibilities to improve the simulation fidelity [41, 45].

6.6 Interpret emerging insulation phenomena under repetitive pulses based on discharge plasma theories

Novel transportation demands (civil and space applications) are driven by the deep-electrification trend, where the repetitive pulse stress is common for the internal insulation. For instance, insulations of the motor winding and silicone gel in a wide-band-gap power module [123, 124] are required to withstand long-term, high-frequency, high dv/dt pulses and be immune to severe partial-discharge-induced degradations. How to understand and control partial discharge behaviours under such extreme conditions are challenging. The ‘sequence and phase resolved partial discharge’ diagram proposed in our previous papers [29] is promising to characterise the partial discharge behaviours under repetitive pulses.

7 Conclusions

This review has focused on foundations in the repetitively pulsed gas and surface discharges: the memory effect and the discharge mode transition, which provide insights towards the controllability and predictability. Macroscopic characteristics of the gas gap breakdown and the surface flashover, mechanisms and dominating agents of memory effects, discharge mode transition, and effects of waveform parameters have been discussed. Research consensuses have been reached regarding general trends, including:

(i) Insulation capabilities of the gas gap and the dielectric surface generally decrease under repetitive pulses compared with single pulse. A steady spark state is usually progressively reached after a long-term excitation.

(ii) Several dominant memory effect agents have been identified and their influential mechanisms have been analysed, such as the metastable species, free electrons, positive ions, and surface charges.

(iii) The high-amplitude, high-PRF, short-width repetitive pulse is beneficial for achieving the uniform and highly reactively discharge plasma. Peculiar discharge phenomena exist in the high-overvoltage ratio scenario.

Although general discharge tendencies have been agreed by the plasma community, emerging observations have challenged previous results and assumptions, which require further understanding and investigation from the long-term evolution and the charge transportation perspective.

Repetitively pulsed gas and surface discharges incorporate rich physics, complicated dynamic phenomena, and diverse temporal and spatial scales. The predictive control of the repetitively pulsed discharge plasma is expected to be built on the thorough interpretation of underlying mechanisms. We would like to emphasise that only two aspects of the repetitively pulsed gas and surface discharges have been briefly reviewed considering a number of relevant effects are continuously discovered and an inherent interdisciplinary basis exists. We hope that our efforts could eventually help the improvements of the predictability and controllability of the transient plasma.

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