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Review Article

Droplet Drying Patterns on Solid Substrates: From Hydrophilic to Superhydrophobic Contact to Levitating Drops

Sujata Tarafdar,[1] **Yuri Yu. Tarasevich**,[2] **Moutushi Dutta Choudhury**,[3] **Tapati Dutta**,[1,4] **and Duyang Zang**[5]

[1]*Condensed Matter Physics Research Center (CMPRC), Physics Department, Jadavpur University, Kolkata 700 032, India*
[2]*Laboratory of Mathematical Modeling, Astrakhan State University, Astrakhan 414056, Russia*
[3]*Centre for Advanced Studies in Condensed Matter and Solid State Physics, Department of Physics, Savitribai Phule Pune University, Pune 411 007, India*
[4]*Physics Department, St. Xavier's College, Kolkata 700 016, India*
[5]*Functional Soft Matter and Materials Group (FS2M), Key Laboratory of Material Physics and Chemistry under Extraordinary Conditions, School of Science, Northwestern Polytechnical University, Xi'an, Shaanxi 710129, China*

Correspondence should be addressed to Sujata Tarafdar; sujata_tarafdar@hotmail.com

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This review is devoted to the simple process of drying a multicomponent droplet of a complex fluid which may contain salt or other inclusions. These processes provide a fascinating subject for study. The explanation of the rich variety of patterns formed is not only an academic challenge but also a problem of practical importance, as applications are growing in medical diagnosis and improvement of coating/printing technology. The fundamental scientific problem is the study of the mechanism of micro- and nanoparticle self-organization in open systems. The specific fundamental problems to be solved, related to this system, are the investigation of the mass transfer processes, the formation and evolution of phase fronts, and the identification of mechanisms of pattern formation. The drops of liquid containing dissolved substances and suspended particles are assumed to be drying on a horizontal solid insoluble smooth substrate. The chemical composition and macroscopic properties of the complex fluid, the concentration and nature of the salt, the surface energy of the substrate, and the interaction between the fluid and substrate which determines the wetting all affect the final morphology of the dried film. The range of our study encompasses the fully wetting case with zero contact angle between the fluid and substrate to the case where the drop is levitated in space, so there is no contact with a substrate and angle of contact can be considered as 180°.

1. Introduction

The study of drying droplets, especially those containing colloidal particles or salts, has become a topic of wide interest in recent years. This is evident from the fact that international conferences on droplets are regularly organized (Droplets Conferences and EMN Droplets) and books exclusively devoted to this subject have been published [1–8]. There are, in addition, several excellent review articles on droplets [9–21]. Different features of this problem, such as the rate of drying, evolution of the drop geometry, and the final pattern formed, depend on a number of parameters, notably the composition of the drying fluid, ambient conditions during drying, and the substrate which supports the drop.

The widespread interest in the drying droplets with inclusions stems from important and innovative applications, mainly in medical science and in technology. When the fluid in the droplet evaporates, the solid material left behind can be distributed in a wide variety of patterns on the substrate. Inclusions may be salts, nanoparticles in the form of nanorods, nanotubes, or any other shape, starches, proteins, and so on. Patterns formed can range from a simple ring at the periphery of the droplet, the so-called coffee ring [22, 23], to multiple rings forming bands, fractal and

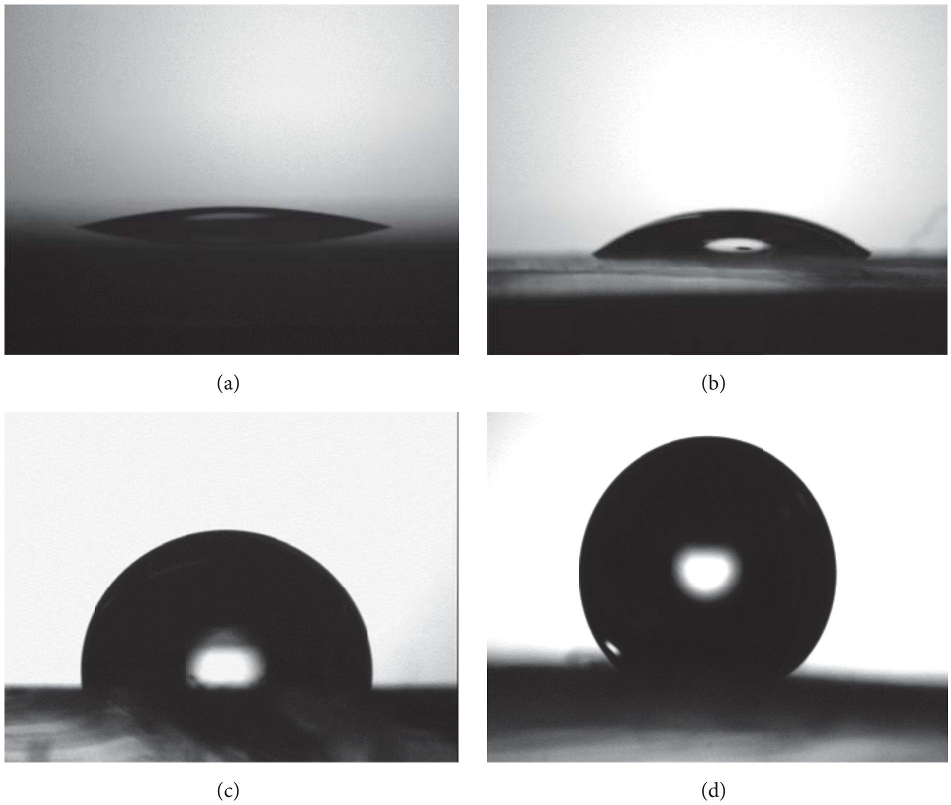


Figure 1: 5 μl drop of water (a) on glass substrate (treated with Piranha solution), (b) on nontreated glass substrate, (c) on polystyrene (PS) substrate, and (d) on hydrophobic TEFLON substrate.

multifractal aggregates of salt crystals, or nanoparticles [24–27]. In addition, the dried drop may develop crack patterns [8, 28–43], which can also be induced by external fields [35, 44]. It is important to realize that the shape of the droplet plays a crucial role in generating these patterns. Unless the drop is too large, its shape may be roughly approximated by a section of a sphere or a spherical cap. In some cases, when a crust or skin forms on the free surface [45], buckling instability may develop [46–49]. Nonuniform evaporation over different regions of the drop surface generates convection currents that determine mass transfer. Temperature and concentration gradients also develop [50–53], leading to surface tension gradients, resulting in thermal Marangoni flow [54–59] or/and solutal Marangoni flow [60]. Obviously, drying out of a large flat film of fluid would lead to a different situation. Figure 1 presents sessile droplets on hydrophilic and hydrophobic surfaces. A thin section of a sphere, where the height at the center is small compared to the lateral dimension, represents a sessile drop that wets the surface well (Figure 1(a)). Here, the angle of contact is very small. On the other hand, a section much larger than a hemisphere represents a strongly hydrophobic contact (Figure 1(d)).

Another possible geometry of a drop in contact with a solid surface is the *pendant drop*, which is suspended from a support *above* it, like a rain drop hanging from a leaf. The pendant drop has also been studied [69], but we do not discuss it further in the present article.

To eliminate the effect of substrate, one may turn to the pendant drop hung by a thin needle or nonwetting drop supported by a superhydrophobic surface. In this case, though its shape was quasi-spherical, the boundary condition for evaporation was, however, influenced by the contact points, thus in turn influencing the evaporation flux. Via levitation techniques, for instance, electrostatic, magnetic, and acoustic levitation, the contact of solid substrate can be completely avoided; thus it somewhat can provide a more uniform evaporation flux along the drop surface. But for most of levitation techniques, the levitation force to balance gravity is surface force, not body force; therefore, the natural convection cannot be suppressed. To further study evaporation without the influence of natural convection, experiments under microgravity, that is, levitation experiments in space, are expected.

Recent experiments on levitated droplets [70] represent the extreme case of a spherical drop with no contact to a substrate. This situation is relevant in space research. In the present review, we aim to discuss the whole range of contact angles and how it affects the morphology of the dried residue. Interplay of related factors such as the chemistry of the materials, interface tensions involved, and physical properties such as elastic or viscoelastic moduli and ambient drying conditions all need to be taken into consideration [71]. There can also be external perturbations such as electric/magnetic fields, heating/cooling of the substrate, or mechanical perturbations. We try to discuss these factors and their contribution as far as possible within the brief span of this review.

We excluded from our consideration some specific topics such as evaporation of sessile droplets on inclined (including vertical), patterned (textured), dissolvable, and porous substrates. To obtain some information about these topics, we can refer the reader to [72–76]. Besides, we omitted all effects connected with artificially inhomogeneous evaporation such as obstacles [77], masks [78], modulated gas phase convection [79], and infrared heating [80], which can force or impede evaporation at particular parts of the free surface of the drop.

The medical science application of the droplet problem utilizes characteristics of the droplet patterns found in dried body fluids, such as blood, serum, tear drops, saliva, and cervical fluid, for identifying diseases in patients. The technique offers a simple and inexpensive method in pathological diagnostics, which has been in use for several decades [3, 81].

A more recent technological innovation is the use of dried droplets with conducting inclusions such as silver nanoparticles or carbon nanotubes (CNT) to create a nearly invisible but connected conducting network on a transparent surface [27, 82, 83]. The demand for such transparent conductors is extremely high in photovoltaic devices of everyday use. The droplet method has a potential to fabricate such surfaces easily and inexpensively. There are other applications related to, for example, ink-jet printing [84] and high throughput drug screening [60].

2. Experimental Studies

In this section, we give an overview of experimental studies on evaporating droplets. We have tried to classify the large body of earlier work into subsections to facilitate the discussion. However, separating the experiments into clear-cut nonoverlapping sections is not always possible. We hope the reader will bear with us in this attempt to present an array of a colourful mass of tangled threads, sorted out as far as possible.

2.1. Evaporation of a Pure Fluid. A sessile drop of a pure liquid, such as water or methanol, when placed on a solid substrate evaporates completely without leaving any residue. The points of interest here are the spreading rate of the drop, the equilibrium shape reached, and how it evolves with time up to complete evaporation. These observations help in understanding the more complicated situations that follow. Obviously, the nature of fluid and ambient are important as well as the properties of the substrate and interaction between fluid and substrate [85]. So we have to consider the substrate here as well, but we leave situations where the substrate (or its absence) has a crucial role to Section 2.2.1.

A liquid drop on a solid surface evolves spontaneously until it reaches its equilibrium with minimum surface area consistent with Laplace's equation (see, e.g., [86]). The shape of the drop on a surface depends on the surface energy of the substrate and the liquid drop, as well as the interface energies between different phases. An additional energy is needed to create an interface between the liquid and solid; this is called

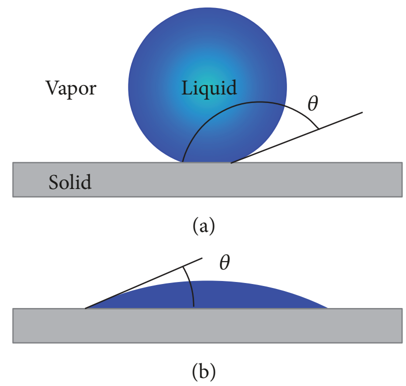


Figure 2: Contact angle between a drop and a substrate: (a) nonwettable substrate and (b) a partially wetted substrate.

the interface free energy. In equilibrium, a liquid drop satisfies Young's equation (see, e.g., [86]).

$$\gamma_s = \gamma_{sl} + \gamma_l \cos\theta, \quad (1)$$

where γ_s, γ_{sl}, and γ_l represent the energy of the surface, solid-liquid interface, and the liquid-vapor interface, respectively. θ is the contact angle between liquid and the surface (Figure 2).

The effects of vapor adsorption and spreading pressure are not considered here. Further, the drop is considered to be much smaller than the capillary length l_c, which is defined as

$$l_c = \sqrt{\frac{\gamma_s}{\rho g}}; \quad (2)$$

here, ρ is the density of the liquid and g is the acceleration due to gravity [1, 86]. Conventionally, the term "drop" is used when the volume of the liquid drop is more than 100 μl and for lower volumes the term "droplet" is used [87].

Assuming the drop to be a spherical cap, the drop boundary on the substrate, that is, the three-phase contact line (TPCL), where solid, liquid, and vapor meet, has the shape of a perfect circle, for an ideal smooth surface. In this article, we do not elaborate on rough or prepatterned surfaces, where there may be interesting deviations [88, 89]. We also leave out from our discussion sessile drops describing works on soft or liquid surfaces.

As the liquid evaporates with time, the drop can shrink in two ways [90] (Figure 3):

(1) The TPCL remains "pinned" to the substrate, leaving the circular solid-liquid contact surface constant; this is referred to as the *"Constant Contact Radius (CCR) mode"* of drying. Obviously, the angle of contact will decrease with time in this case.

(2) Alternatively, the *radius* of the TPLC can decrease, while the angle of contact remains constant. This is called the *"Constant Contact Angle (CCA) mode."*

These are two extreme cases but many real experiments show mixed behavior, where both the contact angle and the contact radius vary (so-called stick-and-slip mode; see, e.g., [91]).

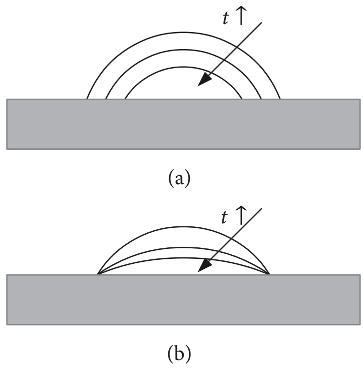


Figure 3: (a) Constant Contact Angle (CCA) mode and (b) Constant Contact Radius (CCR) mode.

CCR is the favored mode for drying on high energy surfaces. For example, in case of Piranha treated glass substrate, the contact angle of a water droplet is less than 15° and it is pinned during drying [92, 93]. On the other hand, in the case of low energy surfaces, for example, a TEFLON coated substrate, the contact angle is more than 150° and the mode of drying is CCA. However, things get complicated in the case of liquid drying on a PMMA or polystyrene substrate. The equilibrium contact angles are 70° and 90°, respectively, but the drying mode switches from CCR to CCA mode with time (Figure 4) [94]. Yu et al. [95] explain the switching from CCR to CCA mode from a thermodynamic point of view. Since we are not discussing rough or patterned surfaces, we refer the interested reader to relevant literature in this area. Modifications of Young's equation required for such problems are discussed in [96–99]. The question of contact angle hysteresis (CAH) also needs to be addressed here [100]. The effect of CAH on pattern formation is discussed in [101].

2.2. Evaporation of a Mixture. In Section 2.1, we discussed how the shape and size of an evaporating drop of pure liquid change during drying. However, once the liquid evaporates completely, no trace of the drop is left, if the substrate is hard and inert. In this section, we come to more interesting observations. What happens when a drop of a solid-liquid suspension or a salt solution evaporates? What pattern does the solid residue leave on the substrate? Study of this phenomenon as a physics problem started with Deegan et al.'s [22] work on the "coffee stain." Noticeably, at the same time, similar independent researches have been published by Parisse and Allain [90, 102]. Moreover, pattern formation in desiccated drops of biological fluids has been known in medical community even several decades earlier [103, 104].

2.2.1. Evaporation of a Suspension of Micro- or Nanoparticles. It is a common observation that when a spilt drop of coffee dries, it leaves a dark ring along the periphery of the droplet, the so-called *coffee-stain* pattern. Initially, the drop looked uniform, so why do the microscopic grains of coffee crowd along the edge during drying, leaving the central portion nearly clean?

Deegan et al. [22, 23] explained that this happens for a pinned boundary (CCR mode). This causes "capillary flow" internal currents from the center towards the TPCL, carrying the suspended solid particles to the boundary, where they get deposited in the form of a ring.

The necessary conditions for formation of the coffee stain are (i) evaporation and (ii) the fact that the drop should be pinned to the substrate during evaporation (Figure 5). Note that an alternative mechanism for coffee-ring deposition based on convection [105] and on active role of free surface [106] has been proposed.

Different patterns of drying are noted when the above conditions are not satisfied. If the TPCL does not remain pinned to the substrate but slips and sticks during drying, a series of concentric rings with varying radius are formed [91, 107, 108].

There is another important effect that competes with the capillary flow causing "coffee-ring effect" and leads to deposition of the suspended particles near the center of the drop. On the other hand, in some cases, the whole solid is deposited near the center of the drop [13, 23, 109]. This is more prominent when the angle of contact is large and is named the *Marangoni effect* [54, 110–112]. It arises due to a gradient in surface tension. There are two sources of such a gradient. First of all, a gradient in surface tension can be produced by a temperature gradient created by different rates of evaporation along the surface of the drop (thermal Marangoni effect). Since evaporation absorbs latent heat, regions near the TPCL cool more than the surface near the top of the drop. This leads to a convection current from the center of the drop surface to the periphery then to center of the bottom of the drop and again to the center of the surface. The temperature gradient in turn produces a gradient in surface tension. This drives the suspended particles along with the fluid in circulating paths from the top of the drop downward towards the center (Figure 6). Here they may get adsorbed on the surface or move towards the periphery and recirculate with the vortex formed. Notice that direction of the Marangoni flow depends on the relative thermal conductivities of the substrate and liquid, $k_R \equiv k_S/k_L$, reversing direction at a critical contact angle over the range $1.45 < k_R < 2$ [85]. It is important to note that a *single* circular flow or vortex is not the only possibility. It has been demonstrated that Marangoni convection induced by thermal conduction in the drop and the substrate can result in multiple vortices, depending on the ratio of substrate to fluid thermal conductivities, the substrate thickness, and the contact angle [113]. The Marangoni effect has been clearly demonstrated in organic liquids. Hu and Larson [111] show that, during evaporation, PMMA microparticles suspended in octane collect near the center of the drop. In water, however, the coffee-ring is observed. Another origin of a Marangoni flow is concentration gradient. Thermal and solutal Marangoni effects may compete with one another [60].

Marangoni effect should always be present in clean fluids but is suppressed in water if surfactants are present. According to Hu and Larson [111], it is difficult to avoid presence of trace surfactants even in "pure" water, so the coffee ring dominates.

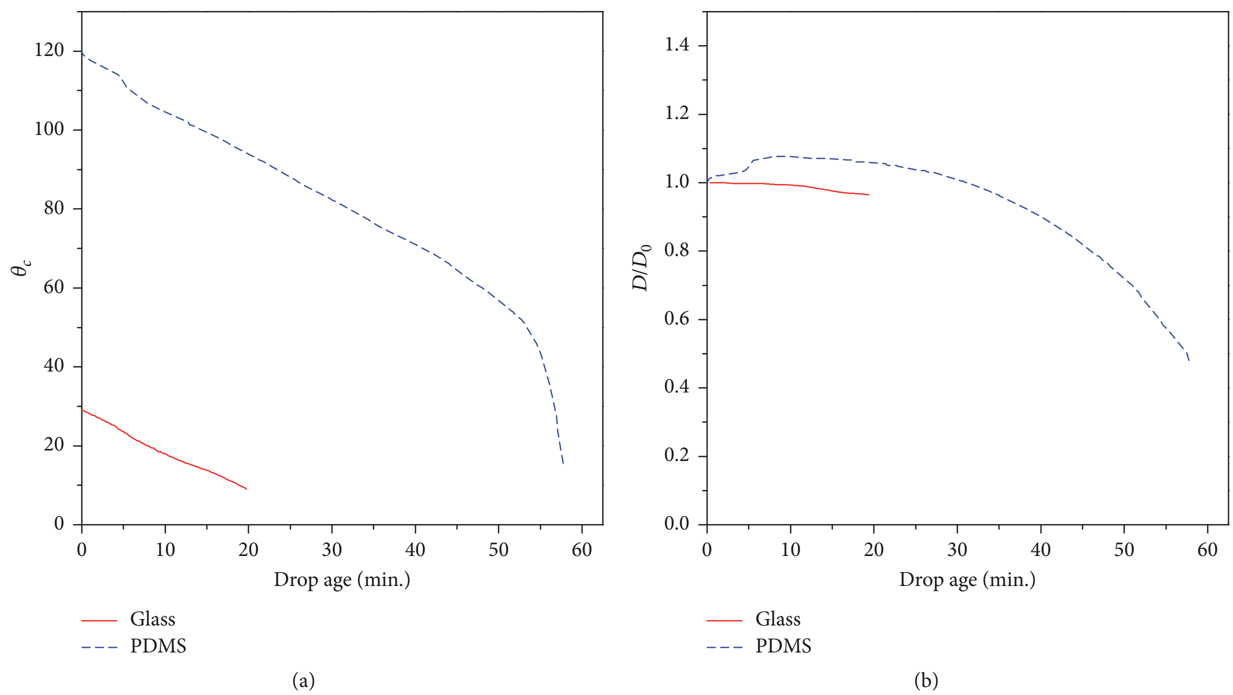


Figure 4: (a) Contact angle variations for a water droplet nontreated glass substrate (red solid curve) and PDMS substrate (blue-dashed curve); (b) normalized diameter of same drops on glass substrate (red solid curve) and PDMS substrate (blue-dashed curve).

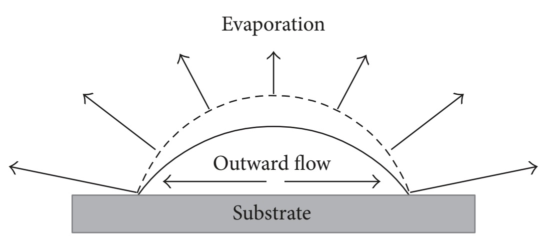


Figure 5: Evaporation and TPCL pinning as the origin of outward flow inside evaporating sessile droplet [22]. Initial position of the free surface is shown as the dashed line, whereas its new position due to evaporation is shown as the solid line.

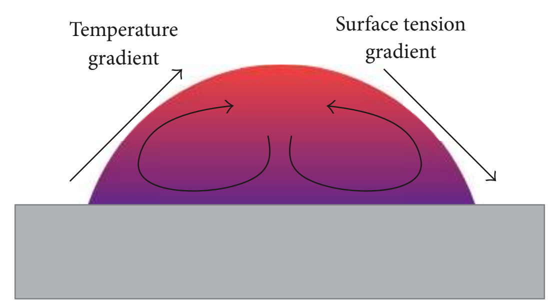


Figure 6: Thermal Marangoni effect is produced by inhomogeneous cooling of the free surface due to evaporation. The temperature gradient in turn causes a surface tension gradient.

The competition between the capillary effect leading to the coffee ring and Marangoni effect is decided by several parameters including the thermal conductivities of the substrate and fluid [85, 113–117] and a Marangoni number (thermal) (Ma) has been defined to characterize this [54, 118]:

$$\mathrm{Ma} = -\frac{\partial \gamma}{\partial T}\frac{H\Delta T}{\mu\kappa}. \qquad (3)$$

Here, ΔT is the temperature gradient, μ is the viscosity of the fluid, κ is the thermal diffusivity, the surface tension of the fluid is γ, and H is the height of the drop. So, hydrophobicity or high contact angle of the liquid on the substrate is needed for getting higher H to induce stronger Marangoni flow inside the droplet.

Once the factors deciding the dominance of capillary flow or Marangoni effect have been identified, it becomes possible to control the deposition pattern and choose whether a thin ring or a uniformly covered circular patch or a small spot at the center of the drop is desired. Besides physical and chemical properties of the fluid and substrate, manipulating the temperature distribution using heated/cooled substrates or microheaters has also been used to tailor the deposition pattern [119–121] for various applications. The effect of particle shape has also been found to play a role in the final desiccation pattern; ellipsoid-shaped particles suppress the coffee-ring effect as shown in several works [109, 117].

2.2.2. Evaporation of a Salt Solution. In Section 2.2.1, we considered a fluid drop containing suspended solid particles of nm to μm size, which do not interact with each other. Now we discuss the case of a *dissolved salt* in a suitable liquid. So here we have dissociated ions uniformly distributed in a liquid drop. Unlike the colloid particles in Section 2.2.1,

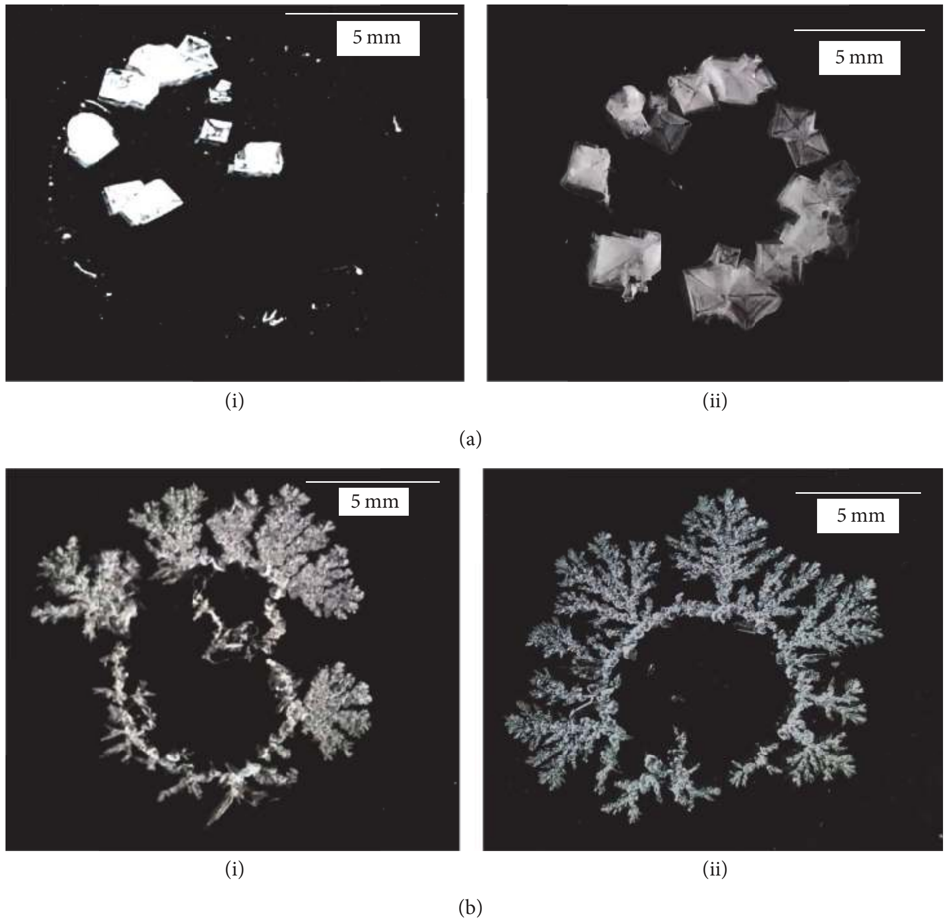


Figure 7: (a) and (b) are dried drops (50 μl) of NaCl and Na_2SO_4 solutions on Piranha treated glass substrates. (i) and (ii) represent different concentrations (0.05 M and 0.1 M, resp.) of salts in the solutions.

the ions tend to form *crystals* as the drop dries and salt concentration increases.

Dutta Choudhury et al. [24, 26] evaporated droplets of aqueous NaCl solution of different concentrations on a hydrophilic substrate. They found that the capillary flow dominates here and the salt collects along the pinned TPCL in the form of a ring of cubic crystals. The size and structure of the crystals depend on salt concentration and evaporation rate. For large concentrations, there is a tendency to form *hopper crystals*. Hopper crystals [122, 123] are somewhat like empty boxes and form when growth at the corners and edges is faster compared to the faces (Figure 7(a)). Droplets of aqueous copper sulphate solutions also display the coffee-ring effect, with crystals forming along the periphery of the drop [124–126]. However, Shahidzadeh et al. [124] did not find coffee rings on drying aqueous NaCl. Rather they found that the salt accumulates near the center of the dried drop. In their experiments, the crystals formed initially at the periphery but were later pushed towards the center. The conflicting findings of Dutta Choudhury et al. [24] and Shahidzadeh et al. may be due to finer details of the experiment. It is possible that fine imperfections/roughness on the surface in the case of the former group pinned the NaCl crystals at the drop periphery, without allowing them to be pushed inward.

Other interesting forms of crystallization have been reported as well. Sodium sulphate crystallizes in two forms—thenardite and mirabilite. At high relative humidity (RH), a drop of aqueous sodium sulphate shows on drying tree-like growth of mirabilite on the substrate *outside the drop* in addition to crystal formation within the drop boundary (Figure 7(b)). This peculiar growth has been explained as follows: crystals formed along the TPCL release water of crystallization which seeps out from the drying droplet and the dendritic trees grow from these. At low RH, thenardite crystals grow within the drop boundary [125, 127].

2.2.3. Evaporation of a Complex Fluid Drop Containing Salt. If the host liquid is a complex non-Newtonian fluid and salt is added to it, the solid residue after evaporation exhibits most beautiful patterns. Such patterns on dried biological fluids have long been studied [103, 104, 128–137], particularly as a tool for diagnosis of certain diseases [3, 62, 138–141]. Before discussing the salt added complex fluid, we should examine what happens in absence of the salt.

If a drop of gelatinized starch or gelatin or an aqueous solution of a polymer like polyethylene oxide is allowed to dry, usually a uniform circular film is produced. Depending on the nature of the fluid and substrate, the film either sticks to the substrate or can be cleanly pulled off [142].

Gelatin, starch, or a similar medium which increases the viscosity of the solution by orders of magnitude can change pattern formation of salts drastically. The flow fields within the drop get suppressed and salt crystals no longer show their normal morphology. The role of gelatin in tuning crystal growth has been known for a long time [26]. Goto et al. [143] demonstrate how tuning concentration of a drying gelatin and NaCl drop and manipulating concentration gradients create unique morphologies such as orthogonal or oblique lattices and curving patterns. They show that nonequilibrium growth conditions lead to various dendritic patterns.

If salts are present in the droplet, they usually crystallize during drying. Morphology of the salt crystals is very sensitive to the kind of salt, concentration, and type of colloidal particles, as well as the rate of evaporation [24–26, 29, 142, 144–148]. This sensitiveness allows using the morphology of salt crystals as an indicator, for example, to diagnose different diseases by drying drops of biological fluids [139, 149].

On addition of a salt, the salt crystallizes in different forms, often several different crystallization modes appear as drying proceeds, and video recordings observed under an optical microscope are fascinating to watch (see videos; a drop of aqueous gelatin solution with a little NaCl salt is allowed to dry; large crystals of salt form, followed by multifractal dendrites in-between and a drop of gelatin containing sodium sulfate forms patterns on drying; concentric rings and dendrites growing from them can be seen). Sodium chloride in gelatinized potato starch solution forms dendrites or hopper crystals of different morphology, depending on experimental conditions [24]. In gelatin, two distinct modes of pattern formation were observed [26, 142]. Formation of initial faceted rectilinear crystals of macroscopic dimensions (of ~mm) size was followed by a fine dendritic network observable only under a microscope. The faceted crystals appear to consist of NaCl only, while the dendrites may be a composite; further analysis is needed here. The dendritic pattern consists of fine self-similar branches meeting at right angles and has been shown to be multifractal [26]. Other salts also form interesting patterns when dried in a gelatin drop. Sodium sulphate forms a series of rings, which are grouped in bands, and dendritic crystals grow from the rings [126]. Copper sulphate forms feather-like patterns, with anisotropy evident from images under crossed polarizers [126]. These patterns also reveal fractal characteristics [150]. Effect of albumin concentrations on sodium chloride crystallization from drying drops of albumin-salt solutions has been experimentally investigated [147].

The processes occurring during the desiccation of the sessile colloidal droplets and morphology of the resulting precipitate depend on many different factors, for example, the nature and shape of the colloidal particles [109] and their initial volume fraction [147], the presence of admixtures (e.g., surfactants) in the solution [50, 107, 151, 152], ionic strength and pH of the solution [28], the properties of substrate (thermal conductivity, whether hydrophilic/hydrophobic) [24, 85, 153, 154], and evaporation mode [32, 36, 155–157].

Surfactants have a strong effect on Marangoni flow as already discussed. A particle-surfactant mixture thus can modulate the capillary flow [18, 107, 152]. Colloidal sulphur with salt self-assembles into a wide variety of patterns, depending on conditions [158]. Different sodium salts are formed in situ, using different acids—monobasic, dibasic, and tribasic. For HCl, the typical NaCl aggregation pattern was observed (similar to [26]). For dibasic sulphuric acid, morphology similar to the two crystalline forms of sodium sulphate was observed and tribasic, citric acid produced a uniform deposition without any definite pattern.

Classification of possible desiccation modes may be done using two characteristic times, namely, the drying time, t_d, and the gelation time, t_g [28]. There are three different modes of colloidal sessile droplet desiccation [28]:

(1) $t_g \gg t_d$, where t_g is the gelation time and t_d is the desiccation time. The gelled phase occurs near the droplet edge and moves inward, while the central area of the droplet remains liquid.

(2) $t_g \approx t_d$. The gelled skin covers the free droplet surfaces. This thin shell cannot prevent evaporation of the solvent. The buckling instability occurs [48].

(3) $t_g \ll t_d$. The phase transition from sol to gel in the whole bulk of the droplet is almost instantaneous. The gelled droplet loses solvent via evaporation very slowly.

When $t_g \gg t_d$, the desiccation process can be divided into several stages (see, e.g., [63, 159, 160]):

(1) Initial single-phase liquid stage: the whole droplet is a sol. The outward flow carries suspended particles to the droplet edge until the volume fraction of the suspended particles, Φ, reaches the critical value, Φ_g. Note that particle-enriched region is extremely narrow, whereas the particle volume fraction in the central area of the droplet is almost constant along its radius. This stage was simulated in [161–163] and in [159].

(2) Intermediate two-phase stage: a gelled ring appears near the droplet edge and grows towards the droplet center. The volume fraction of the colloidal particles is constant inside the *foot*, that is, the outer gelled band, Φ_g, and almost constant in the sol, Φ, except for a rather narrow area near the phase front. This stage was simulated in [164–166] and in [159, 167].

(3) Final single-phase solid stage: the gelled deposit loses the remaining moisture very slowly. Some real fluids of interest (e.g., biological fluids) can contain both suspended particles and dissolved substances. In this case, the dendritic crystals can occur in the central area of a sample [25, 29, 60]. Finally, the desiccation crack patterns appear [8, 28–31, 33, 36, 38].

2.2.4. Biological Fluids. Most biological fluids can be regarded as a complex fluid containing salts. But we devote a

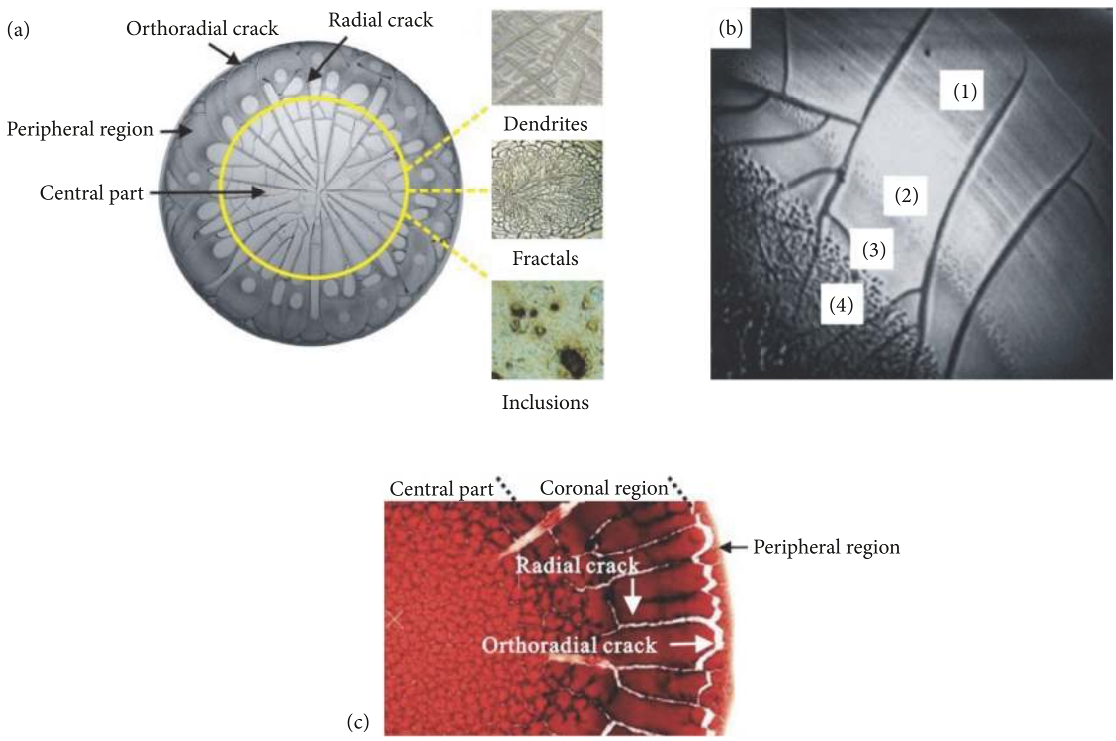


Figure 8: Desiccation patterns in the sessile drop of (a) blood plasma from a healthy adult (adopted from [61] with the permission of Springer and from [62] with the permission of Hindawi); (b) morphological details of four regional patterns in the sessile drop of BSA saline solution: (1) homogeneous protein film, (2) protein precipitates, (3) protein gel, and (4) salt crystal [63] (reproduced with permission of Elsevier); (c) whole blood droplet drying on a glass substrate (adopted from [64] with the permission of Cambridge University Press). The whole figure is reproduced from [65].

separate section to these liquids, because the complexity in composition and the bioactive nature of the constituents of body fluids make the evaporation of such liquid drops quite different from laboratory prepared micro or nanofluid drops.

Although pattern formation can be observed during drying of both inorganic and organic colloids, the case of biological fluids is increasingly attracting attention from the scientific community in recent years [64, 168, 169] as this study promises to be a simple and cost-effective technique for diagnosis [20]. We shall discuss further details of application in medicine in Section 4. In this section, we will review some of the experimental observations of drying on drops of human blood serum, whole blood, and other biofluids such as urine, saliva, and tears.

While performing the experiments seems very simple (table top experiments producing interesting patterns can be done even at home), understanding the physics behind the pattern formation phenomena turns out to be extremely complicated and involves a number of interrelated processes of different nature [10, 12]. During desiccation of biological fluids, a sequence of various physical and physicochemical processes can be observed [63, 170]. For example, redistribution of the components occurs. Protein molecules are carried by flows to the edge of the droplet and accumulate to form a gel. The salt is distributed over the whole area of the droplet almost uniformly. After complete drying of the droplet, a protein precipitate remains on the substrate in the form of a ring; the width of the ring depends on concentrations of the protein and the salt [3, 171, 172]. Salt crystals can form fractal (dendritic) structures [24, 29, 142, 173, 174]. In the later stages of drying, a sample may crack [8, 28, 29]; the characteristic pattern of the cracks also helps in diagnosing diseases from which the subject may be suffering [175].

Blood serum mainly contains 90% (by mass) of water, 6% of macromolecular proteins, 1% of inorganic electrolytes, and other minor components [20]. Although its composition is complicated, blood serum behaves like a Newtonian fluid. Desiccation patterns of a sessile drop of blood serum drying on a solid substrate are generally characterized by two distinguishable regions: a peripheral region and a central part. Cracking patterns in orthoradial and radial directions are observed throughout the whole sessile drop, and crystal patterns with different morphologies accumulate in the central part [61], as shown in Figure 8(a).

Three major factors influence the morphologies of these serum patterns: concentration of inorganic salts, concentration of macromolecular proteins, and the wettability of the droplet on the substrate. Inorganic salts are essential for formation of crystal patterns in the central part [50]. High concentration of salts promotes the aggregation of macromolecular proteins, thus changing the morphologies of crystal patterns [176]. The wettability of the substrate plays an important role in determining the apparent contact angle of the sessile drop [177]. This may further influence the TPCL motion and the mass transportation during drying and in turn the desiccation patterns.

Whole human blood behaves like a non-Newtonian fluid unlike blood serum. It is composed of plasma (55% by volume) and cellular components (45% by volume) (i.e., red blood cells (RBCs), white blood cells (WBCs), and platelets); RBCs, WBCs, and platelets represent 97%, 2%, and 1% of the total volume of these cellular components, respectively. Desiccation patterns in the dried sessile drop of whole human blood are significantly different from those of blood plasma without cellular components [64, 153]. Patterns in the blood droplet from healthy adults dried on glass substrates consist of three distinct zones with different characteristic cracking patterns (Figure 8(b)), namely, a fine peripheral region adhering to the substrates, a coronal region with regularly ordered radial cracks and large-sized deposit plaques, and a central part with disordered chaotic cracks and small-sized deposit plaques [64]. Desiccation patterns of the blood droplet are significantly influenced by the external drying conditions, such as the RH and the wettability of the blood droplet on the substrates [36, 38, 157].

Characteristic desiccation patterns are also formed in the sessile drops of other biofluids (e.g., urine, saliva, and tear fluids) [11, 178, 179]. Yakhno et al. investigated the drying of sessile drops of urine and saliva from the healthy adults and divided it into three stages: the redistribution of materials leads to the continuous flattening of the droplet; the deposited macromolecular proteins aggregate to form the gel matrix; the inorganic salts induce phase transition of macromolecular proteins to form desiccation patterns [11]. The desiccation patterns of the tear droplet are characterized by a thin amorphous film in the peripheral region, with fern-like patterns in the central part. The thin amorphous film in the peripheral region has crack patterns as observed by SEM. The fern-like patterns in the central part are composed of cubic crystals and dendritic patterns adjacent to them. Energy dispersive X-ray analysis (EDXA) results revealed that dendritic patterns were predominantly made up of sodium and chloride, while cubic crystals were potassium and chloride [180].

Despite the application of the phenomena for practical purposes and considerable progress in understanding of the phenomena [10, 12], the theoretical description of the pattern formation in desiccating biological fluids is still incomplete. The physical, biophysical, biochemical, biological, and physicochemical processes occurring in the dehydration of biological fluids remain largely to be clarified.

Analysis based on a visual comparison of the structures formed by drying a liquid drop [3, 4, 81, 181, 182] has significant drawbacks. Conclusions are liable to be subjective, without techniques for defining quantitative parameters to characterize the structures. Computer pattern recognition may be tried to eliminate this shortcoming [36, 61, 183].

Other samples of biological origin which form liquid crystal phases have shown interesting patterns on drying droplets. For example, DNA [184] forms a ring-like deposit with zigzag patterns [185]. Cetyltrimethylammonium Bromide drops with salt were also found to form concentric rings and crystalline aggregates near the center [186].

2.3. Evaporation of Drops under Levitation. So far we discussed sessile droplets that sit on a solid substrate during drying. Even for the superhydrophobic case with contact angles approaching 180°, the dried pattern left on the substrate is two-dimensional or quasi-two-dimensional (when the deposits pile up forming rings or aggregates). The aspect ratio of the deposit, that is, the maximum height, divided by the drop radius is much less than 1. Is it possible for a drying drop to reduce to a spherical shell after evaporation? It turns out that this can be achieved by drying a levitated drop floating in air with zero contact to any surface. Suspending the drop by properly adjusted acoustic fields is the most convenient technique for this study. Radiative heating leads to evaporation of the solvent.

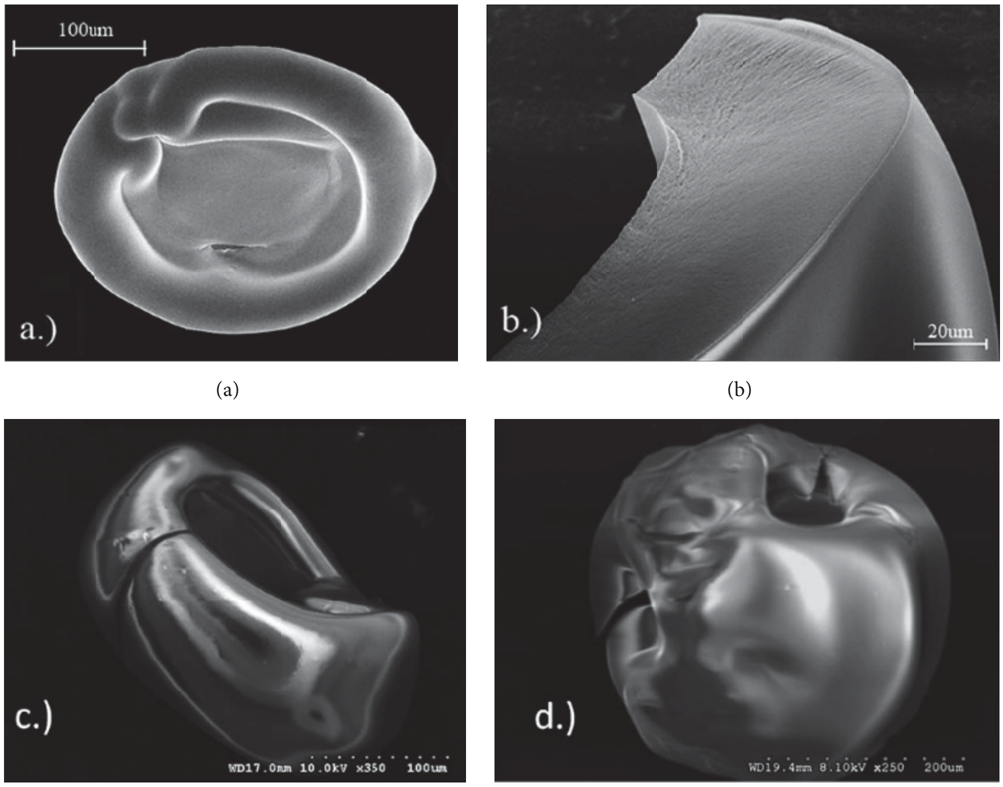
Tijerino et al. [66] evaporated droplets with nanosilica suspensions under different conditions of acoustic amplitudes and solid concentration to obtain residues with varied shapes like rings, bowls, and spheroids (Figure 9). Wulsten et al. [70] used acoustic levitation to investigate the effect of a solvent containing different polymers and the drug itraconazole on drying. The morphology of the residue was found to depend on the drug, while the polymer determined the drying rate. Single-phase droplets [70, 187] and binary or multiphase droplet [188, 189] have been studied using similar techniques.

Although liquid drops can be levitated by various levitation techniques ranging from electrostatic to diamagnetic [190], the acoustic technique is more suitable for evaporation experiments as the electromagnetic properties of the materials are not relevant here. The drying patterns of colloidal droplets depend on the particle-particle interactions and are significantly affected by the substrates as well. With the increase of substrate contact angle, the final residue pattern can be coffee-ring-like stains (Figure 10(a)) on hydrophilic substrate [37] and bowl-shaped residue (Figure 10(b)) on a hydrophobic substrate ($\Theta \sim 90^\circ$) [67]. Under acoustic levitation, a ring-shaped residue has been obtained (Figure 10(c)) [68], which exhibits a geometrical similarity with the initial dog-bone shape of the acoustically levitated droplet [191]. These results highlight the important role played by the drop profile in drying pattern formation, and acoustic levitation provides the possibility to study drop evaporation at zero-contact condition for different initial shapes.

2.4. Desiccation Crack Patterns on Drying Drops. As a colloidal drop dries, viscosity increases and a sol-gel transition may occur. Stress accumulates nonuniformly due to the droplet shape and inhomogeneity of the fluid. This often leads to formation of cracks with characteristic patterns. In addition to the *crack patterns*, other quasi-3D effects are of interest such as wrinkling, buckling, and skin formation.

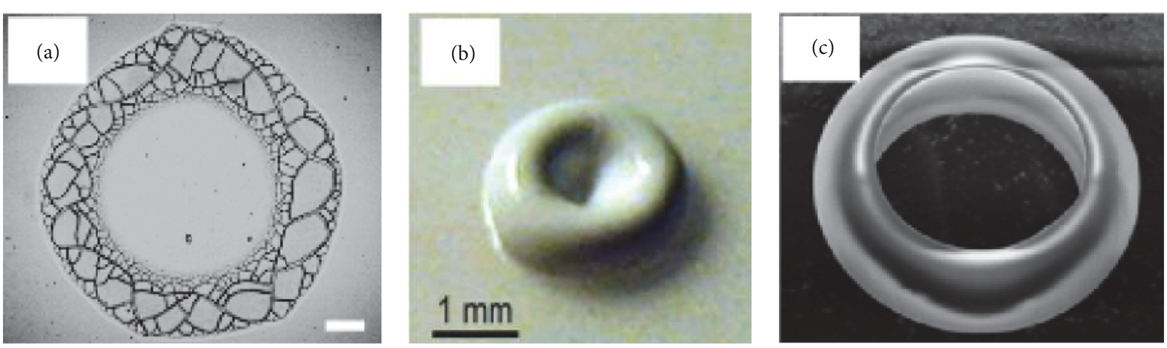
Desiccation crack patterns have been intensively investigated both experimentally and theoretically [28–32, 34, 36–41, 192]. State of the art may be found in the recently published book [8]. We discuss here some studies on crack formation in drying drops [16, 24, 29, 193].

The final drying pattern and crack nucleation vary with the kinetics of the evaporation rate. During solvent evaporation, curvature of the solvent-air meniscus is responsible for



(c) (d)

Figure 9: Different residues obtained from levitating droplets (reproduced from [66], with the permission of AIP Publishing).



(a) (b) (c)

Figure 10: Different drying patterns obtained from varied conditions. (a) Coffee ring-like stain on hydrophilic substrate, reprinted with permission from [37]; (b) bowl-shaped relic on hydrophobic surface, reprinted with permission from [67]; (c) ring-shaped residue obtained from acoustic levitation, reprinted with permission from [68].

a capillary pressure in the liquid phase. The capillary pressure induces shrinkage of the porous matrix, which is constrained by the adhesion of the deposit to the glass substrate. As tensile stresses build up, the internal stresses become too great and fractures appear to release mechanical energy. The differences in pattern formation arise due to the competition between the drying process and the adhesion of the matrix on the substrate.

Annarelli et al. [29] worked on the evaporation, gelling, and the cracking behavior of a deposited drop of protein solution, bovine serum albumin. They observed that the cracks appearing at the gelling edge were regularly spaced and were a result of the competition between evaporation-induced evolution and relaxation-induced evolution. When the crack evolution is only evaporation-induced, the mean crack spacing is proportional to the layer thickness. However, in the case of a drop of bovine serum albumin, the evolution of cracks has been described in relation to the change with time of the average shrinkage stress. In this case, the mean crack spacing was observed to be inversely proportional to the deposit thickness. This is unexpected as normally crack spacing increases with thickness.

Brutin and his group worked on the pattern formation of desiccating droplets of human blood from which the coagulation protein had been removed [7, 64]. They studied the dynamics of the process of evaporation of a blood droplet using top-view visualization and the drop mass evolution during the drying process. Brutin et al. [64] showed that there are two distinct regimes of evaporation during the drying of whole blood. The first regime is driven by convection, diffusion, and gelation, while the second regime is only diffusive in nature. A diffusion model of the drying process allows a prediction of the transition between these two regimes of evaporation. Concentration of the solid mass in the drop was important and fracture occurred at a critical mass concentration of solid in a drying drop of blood. They showed that the final crack patterns formed on drying droplets of blood collected from a healthy person, anemic person, and hyperlipidemic person are quite different. But drawing conclusions for definite diagnosis is not so straightforward as the crack patterns are strongly affected by external conditions such as the ambient relative humidity and the nature of the substrate.

Brutin et al. [64] conclude that the final drying pattern and crack nucleation vary with the kinetics of the evaporation rate. The transfer of water to air is limited by diffusion and is controlled by the relative humidity in the surrounding air. The drying process of a sessile drop of blood is characterized by an evolution of the solution into a gel saturated with solvent. When the gel is formed, the new porous matrix formed by the aggregation of particles continues to dry by evaporation of the solvent, which causes the gel to consolidate.

Carle and Brutin [154] studied the influence of surface functional groups and substrate surface energy on the formation of crack patterns and on the dry-out shape in drying a water-based droplet of nanofluid. They have also studied desiccation of blood droplets [153] on different substrates such as glass and glass coated with gold or aluminium. They measured the rate of heat transfer from the substrate to the fluid drop. They show that wettability of the substrate by the fluid is the decisive factor, which can account for the differences in the morphology of the desiccated blood drop on different surfaces, rather than the thermal diffusivity which determines rate of heat transfer from the substrate to the drop. On metallic surfaces, where the drop is nearly hemispherical and a glassy skin forms on the fluid-air interface, there are hardly any cracks. On a glass surface, on the other hand, where the drop is more or less flat, an intricate pattern of cracks form. Figure 11 shows spiral cracks typical of albumin; these can be easily observed by drying egg white [8].

The study of desiccation crack patterns on clay-gel droplets dried in a static electric field [35] led to some interesting results. The number of cracks formed and the time of first appearance of the cracks could be related to the field strength through exponential relations. In a further set of observations, the field was applied for a very short finite duration and then switched off. Now the time required for crack appearance after switching off could be related to the field strength and exposure time, when all quantities were appropriately scaled.

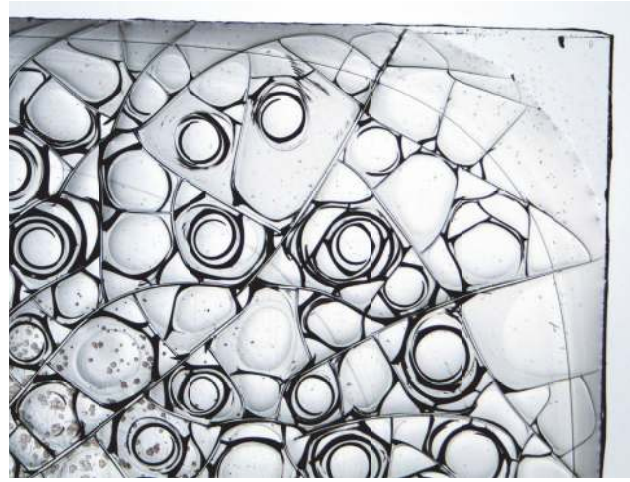


Figure 11: Crack patterns in a dried sample of albumin (courtesy of N.A. Koltovoi).

In a follow-up of this work [194], where platinum electrodes were used to avoid chemical interactions during drying, it was shown that the clay drop behaves like a leaky capacitor and could be modelled using methods of generalized calculus [195].

Effect of the drop constituents on *crack speed* has also been studied. Zhang et al. show that a colloidal solution of polytetrafluoroethylene particles cracks radially as it dries [196]. Crack speed varies as a power law with thickness and surfactants such as sodium dodecyl sulfate can be used to tune the cracking.

2.5. Desiccation under Perturbation. While there are several works on desiccating droplets done by different groups, there are very few studies on drying droplets in the presence of a perturbation. The contact angle of a conductive aqueous laden drop with organic or inorganic solutes or ambient oils changes with the application of alternating current (AC) voltage during drying. Banpurkar et al. [197] studied the above effects in experiments to demonstrate the potential of electrowetting-based tensiometry. Contact angle (θ) decreases with increasing amplitude (V_{AC}) of AC voltage following the linear relation of $\cos\theta$ with V_{AC}. They applied low frequency AC voltage and obtained interfacial tensions from $5\,mJ/m^2$ to $72\,mJ/m^2$, in close agreement with the macroscopic tensiometry for drop volumes between 20 and 2000 nL. Vancauwenberghe et al. [198] reviewed the effect of an electric field on a sessile drop. They observed that an external electric field can change the contact angle and shape of a droplet. The electric field also affects the evaporation rate during drying. The contact angle is not always an increasing function of the magnitude of the applied electric field but may be a decreasing function for some liquid droplets as well.

Studies have also been done on the effect of external perturbations during drying. The effect of electric fields has been studied on cracks formed in drops of drying clay gels

[35, 194]. Khatun et al. [35] and Hazra et al. [194] investigated desiccation cracks on drying droplets of aqueous Laponite solution in the presence of a static electric field (DC). The electric field had cylindrical geometry, with the peripheral electrode being an aluminium wire [35] or platinum wire [194] bent into a circular form with diameter of ~1.8 cm. A drop of Laponite gel was deposited inside this wire loop. Another aluminium/platinum wire with its tip touching the lower substrate through the centre of the drop acted as the central electrode. Typical cracks had radial symmetry and were found to emerge always from the positive electrode. The cracks formed even when the field was applied for a few seconds and then switched off; this was interpreted as a *memory effect*.

Sanyal et al. [199] experimentally investigated nanoparticle aggregation and structure alteration of evaporating sessile colloidal droplets when subjected to low frequency vibrations of the substrate. Low frequencies perturbed the droplet when the corresponding vibrational wavelength was comparable to the size. For forcing frequencies in the resonance band of lowest allowable mode, the change in the overall morphology of the deposit structure from natural droplet drying was pronounced, with a sharper wedge at the periphery. Recirculation and subsequent outer flow near the droplet edges created higher particle concentration, leading to faster growth of the peripheral wedge. For frequencies away from resonance, the internal flow was mostly uniform, leading to less enhanced wedge structure, similar in appearance to the case of natural evaporation. They demonstrated that, by using forced vibration, desired control of particle deposition could be achieved for various applications.

2.6. Interacting Droplets. We briefly mention another recent interesting line of research here. There are studies showing that a drying drop influences the pattern formation on another drying drop, provided that the distance between them does not exceed a certain limit. This is because the presence of another drop enhances the RH on the side nearer the other drop. So evaporation at the near side is less than that at the far side when two drops are placed side by side. Pradhan and Panigrahi [200] give an experimental as well as a simulation study of this problem.

Cira et al. [201] showed experimentally that two-component droplets of well-chosen miscible liquids such as propylene glycol and water deposited on clean glass at distances of up to several radii apart moved towards each other. This occurred over a wide range of concentrations and even when both droplets had the same concentration. The droplets increased speed as they approached each other. These long-range interactions were preserved even across a break in the glass slide. They explained that the two neighbouring droplets each lie in a gradient of water vapor produced by the other. This gradient causes a local increase in RH and thus decreased evaporation of the thin film on the adjacent portions of the droplets, breaking symmetry. The decreased evaporation leads to an increased water fraction in the thin film, hence increasing the interface tension between liquid and vapor for the film, denoted by $\gamma_{\text{LV,film}}$ locally. Asymmetric $\gamma_{\text{LV,film}}$ around the droplet causes a net force that drives the droplets towards each other. Cira et al. [201] proposed a mathematical model to obtain the net force acting on each droplet as follows:

$$F = 2\gamma_{\text{LV,droplets}} m R \int_0^{\pi} \frac{\left(1 - RH_{\text{room}}\right) R \cos\psi}{\sqrt{d^2 + R^2 + 2Rd\cos\psi}}\, d\psi, \quad (4)$$

where m is the slope of a plot of apparent contact angle versus the relative humidity, RH; d is the distance between the droplets; ψ is a parameter of integration; RH_{room} is the ambient humidity far from the droplets. This net force causes droplet motion and is balanced by a viscous drag.

If droplets of different surface tension but equal concentration coalesce upon contact, fluid is directly exchanged between the droplets. This exchange of fluid leads to a surface tension gradient and Marangoni flow across both droplets, where the droplet of lower surface tension chases the droplet of higher surface tension, which in turn flees away. They explained the observed phenomena of droplets of sufficiently different concentrations exhibiting a "chasing phase" indicative of a repulsive force that comes into play as the gradient of the vapor pressure decreases $\gamma_{\text{LV,film}}$ around the droplets causing them to move away.

Using their understanding of "self-fuelled surface tension driven fluidic machines," the authors have explored several applications in food coloring, glass slides, and permanent Sharpie markers.

3. Theoretical Description and Modelling

3.1. Models of Evaporation. Any model of mass and heat transfer inside an evaporating drop and any model of deposit pattern formation are based on models of evaporation from free surface of the drop. The functional form of the evaporation rate depends on the rate-limiting step, which can be either the transfer rate across the liquid-vapor interface or the diffusive relaxation of the saturated vapor layer immediately above the free surface of the drop [202]. Hence, two main approaches should be mentioned.

The first one is based on the assumption that evaporation from free surface of the drop is steady-state and diffusion-limited. Analysis of this assumption validity can be performed in detail by Popov [202]. In the case of diffusion-limiting quasi-steady process, the vapor density above the liquid-vapor interface obeys the Laplace equation. When droplet shape is governed by surface tension, it can be treated as a spherical cap (see Section 1) and Laplace equation can be solved analytically. This solution has one essential drawback; namely, vapor flux is singular at TPCL. To suppress this physically senseless singularity, a correcting factor may be introduced [203–205].

The second one is based on the assumption that the rate-limiting factor is heat transfer [206]. In this case, a singularity is missing in analytical formula for the vapor rate except for the case of highly volatile droplet.

3.2. 2D Models of Mass Transfer. Modelling of the processes occurring during the drying of colloidal droplet solutions is very complicated, because these processes are extraordinarily varied and complex [63]. The authors have different views about the driving mechanisms that lead to the formation of the solid phase [22, 23, 90, 91, 202, 207]. For example, [208] considered competition of convection and sedimentation, but [159] considered competition of convection and diffusion. Numerous models were proposed during the last two decades. Several models describe some particular processes occurring during the colloidal droplet desiccation (e.g., capillary flow and mass transport processes) [23, 90, 105, 167, 202, 207–215]. Generally, models are developed for systems with low concentrations of the colloidal particles.

Two very different situations are possible when a colloidal sessile droplet desiccates. In the first case, the particles inside a droplet can interact with each other only mechanically (impacts). In this case, the deposit forms a porous medium. Such a medium prevents neither bulk flow inside it nor evaporation from its surface. Moreover, such a porous medium can enhance evaporation from its surface due to drainage effect [209]. In the second case, the colloidal particles can form strong interparticle bonds. In this case, hydrodynamic flows, particle diffusion, and solvent evaporation are restricted. The proposed theoretical models mainly deal with the first situation [23, 90, 105, 167, 202, 207–214]. Only a few models treat the deposit as impenetrable for flows and preventing evaporation [159, 165–167]. Nevertheless, the simulation of desiccated colloidal droplets with phase transition is extremely important for high-throughput drug screening [60], biostabilization [216], identification of fluids [183], and medical tests [139, 217–219]. The models in [159, 165–167] utilize sets of rather complicated partial differential equations (PDE).

Other approaches for simulating pattern formation in drying drops have also been tried. A simple Monte Carlo algorithm for evaporation and pattern formation has been developed by Dutta Choudhury et al. to reproduce the formation of faceted salt crystals and dendritic aggregates in drying droplets of aqueous gelatin containing NaCl [142]. The pattern formation can be correlated to the topological concept of the Euler number [220].

Several models describing desiccated sessile colloidal droplets have been reported recently [117, 159, 165, 167, 221]. They are based on the lubrication approximation [222]. This approach has several serious shortcomings [223] as enumerated below:

(1) Only thin films can be considered; all quantities are supposed to be dependent only on one radial coordinate.

(2) In fact, a two-phase system is considered as one-phase system; the gel is assumed to be a liquid with very high viscosity; the hydrodynamic equations are written for the whole droplet desiccation.

(3) The mathematical expression for evaporation flux above the free surface is speculative rather than being supported by experiments. To the best of our knowledge, measurements of the vapor flux above a system with sol-gel phase transition are not published yet.

(4) Knowledge of the effect of particle concentration on viscosity is needed for calculations. This dependence can be obtained from experiments with rather large volumes of colloid. Viscosity of a small droplet with a large free surface and large contact area with a substrate can deviate from this in a rather complex manner.

(5) It is assumed that all the molecules that get to the edge of the droplet pass into the solid phase. Generally, this assumption can be wrong in the presence of convection of any nature in a droplet. An inward flux of particles due to diffusion may also exist.

To overcome the limitations of the listed models, three-dimensional (3D) models need to be developed and utilized.

3.3. Modelling Flow in 3 Dimensions. A number of papers devoted to 3D models of processes inside evaporating droplets were published during the last few years. Mostly, the articles consider droplets of pure liquids and simulate flows within them [114, 224–229]. The analytical solutions of the Laplace equation which describe the velocity field inside evaporating droplets of a nonviscous liquid were obtained for the contact angle of 90° by Tarasevich [226] and for a case of arbitrary contact angle by Masoud and Felske [230].

Flow inside the boundary line of an evaporating liquid for any contact angle was found using Stokes approach [231]. Numerical calculations of the velocity field within evaporating droplets were performed using finite element method [54, 110, 225]. Presence of dissolved substances or suspended particles inside the droplets and deposit formation were not taken into account in these models.

3.4. 3D Models of Mass Transfer. 3D models describing the processes inside the particle-laden droplets were developed using both the continuum and discrete approaches. Development of discrete models was initiated by the requirements of modelling of evaporation-driven self-assembly (EDSA) or evaporation-induced self-assembly (EISA) [214, 232–239]. Additional references can be found in [240]. Recently published models considered the Brownian motion of particles inside the droplets. For instance, in the work of Petsi et al. [213], the Brownian motion of the particles is superimposed on the hydrodynamic flow calculated previously [231]. A continuum approach has also been applied in some works [59, 208, 209].

Conflicting results from experiments imply that available models may be too simplistic and more realistic theories need to be developed. For example, some experimental data indicate that transfer of the suspended particles to the edge of the drying droplets is possible only when the Marangoni effect is suppressed [111]. However, other experimental studies consider the Marangoni effect to be the driving force for the formation of a new phase on the edge of a drop. It appears therefore that the theoretical models are incomplete, or too drastic approximations/assumptions have been made while formulating them.

There is also some confusion due to nonuniform terminology used by different research groups. For example, [209] reports *depinning* for large contact angle and no depinning for small contact angle. But it refers to receding of the fluid from the solid deposited at the TPCL as "depinning." But many other researchers use the term "depinning" as the inward motion of the TPLC as a whole, leaving no deposit behind.

Another such instance is the fact that the direction of flow can be opposite to a direction that is predicted by calculations for the pure solvent [241]. Independent experiments confirmed that the flows in pure liquids and in liquids with admixtures go in different directions [242, 243]. In the multicomponent liquids of biological origin, the thermocapillary and solutocapillary effects can eliminate each other [60]. Calculations of various research groups have shown that during evaporation of the droplet of a pure liquid there are circular flows caused by the Marangoni stress. The flow is directed along the droplets base to its edge and along its surface towards to the center of the drop [54, 114, 225, 228, 244]. At the same time, experiments conducted with biological fluids exhibit opposite direction of flow [241–243]. According to Kistovich et al., Marangoni flow cannot generally occur during drying of the droplets of biological fluids; they suggest that the observed circular currents are caused by buoyant convection [105].

Most of the earlier models for simulating evaporation of droplets have been developed for single solvent droplets. Recently, a finite element model has been formulated by Diddens et al. [245, 246], which explains results of interesting experimental phenomena such as self-wrapping of ouzo drops on a superamphiphobic substrate [247].

Evaporation of a multicomponent drop is interesting because the solvent contains different liquids with different volatility, so the composition changes during evaporation. For example, in a water-glycerol mixture, where evaporation of glycerol can be neglected compared to water, a large contact angle is shown to lead to a reversed Marangoni flow. If the relative humidity is high, when water content of the drop is reduced due to evaporation, water vapor from the surroundings condenses onto the drop. The problem now becomes nonlinear and chaotic vortices form [246]. This has been observed experimentally as well [248].

On the other hand, for a water-ethanol mixture, where both components evaporate fast, the substrate cools rapidly and thermal transport has to be taken into account. Chaotic behavior is predicted by the model [246] and has also been observed experimentally [249–251]. There are, in addition, models focusing on a particular aspect of desiccation such as skin formation [252, 253].

3.5. Crystal Growth. The sensitivity of crystal morphology on various parameters impedes modelling because a lot of different effects have to be taken into account. In fact, all used models should be treated as semiempirical. The models often utilize the lattice approach [142, 254–257] and diffusion equation [25, 258]. Adequacy of some models [254–256] has been questioned [258]. Mainly, dendritic crystal growth can be observed at the final stages of drop desiccation. Both nonequilibrium growth and presence of impurities may produce dendritic shape of crystals [259]; these effects can be reproduced in a simple model [260]. The semiempirical Monte Carlo approach by Dutta Choudhury et al. [142] qualitatively reproduces the crossover in faceted and dendritic crystal growth and shows the relevance of statistical methods in this problem through the Euler number [220].

The phase-field method [261] looks extremely promising for modelling crystal growth in desiccated colloidal droplets with salt admixtures, but it requires a lot of additional information, which is difficult to obtain experimentally.

4. Applications and Perspectives

Initially, the negative effect of the coffee ring was of concern to scientists and engineers, since it precludes uniform deposition in processes such as ink-jet printing. So methods to reverse it were in demand. We have discussed in Section 2.2.1 that this is possible by enhancing Marangoni flow or imposing temperature gradients. Another method is to use electrowetting [96, 262, 263]. This can be done by applying voltage though the drop by the technique known as eMALDI, introduced by Eral et al. [96]. Varying voltages give different deposition of salt patterns on substrates [262].

In recent times, however, the picture has changed; increasingly various patterns formed by evaporating drops are being put to good use, instead of being considered a hindrance. We briefly mention below some fields where the *nonuniform distribution* of solute in the drying drop has been helpful.

4.1. Application in Functional Materials. The group led by Shimoni et al. [264] created a connected network of coffee rings by deposition of conducting micro/nanoparticles on a transparent substrate. This produces a transparent conductor that is extremely important in today's technology for various photovoltaic devices. The droplet technique provides a much cheaper alternative to indium tin oxide or indium tin fluoride coated glass, traditionally used as transparent conducting material. Drying a drop of some specific solvents with CNT (carbon nanotubes) on a cooled substrate similarly produces a polygonal connected, self-assembled network, which is transparent as well as conducting [27]. Moreover, the coffee-ring effect is used for the separation of two different sized particles [265, 266].

4.2. Biomedical Application. The utility of studying patterns of dried biological fluids has been well known in medical diagnosis [138]. The structures observed after drying biological fluids on a horizontal impenetrable substrate attracted the attention of researchers as early as the 1950s [103, 104, 128–132, 134–137]. In the 1980s–1990s, doctors of the former Soviet Union began to use the appearance of structures formed by drying droplets of biological fluids for the diagnosis of various diseases [3, 4, 81, 181, 267]. Unfortunately, very few of these articles were published in English [62, 138, 140, 149, 182, 217].

Many constituents of biological fluids crystallize on drying. The presence of these crystals, their morphology, size, and abundance are of great help in pathological investigations. Denisov describes crystallization patterns observed in saliva of patients with gastrointestinal diseases [268]. He shows that box-shaped, cross-shaped, and dendritic crystals with multiple-level branching are observed in various samples. The shapes can be classified in a phase diagram with nonoverlapping groups to identify problems such as peptic ulcer, chronic gastritis, and other such diseases in patients.

The potential for diagnosis by using the evaporation patterns of whole blood or blood serum lies in the fact that blood composition may vary due to diseases, which in turn results in changes in the evaporation patterns. Researchers had used dried human serum patterns to diagnose metastatic carcinoma [269] and also found that various interesting patterns could be used to reveal different pathological information [61]. The dried drop patterns of blood serum were also suggested to be useful for disease diagnosis because some featured morphologies of blood serum patterns could be used to acquire information about the health state of human organisms [138]. However, the use of whole blood patterns for medical diagnosis was rarely reported [42, 153], and more systematic experiments are expected.

The coffee-ring effect has been used as low-resource diagnostics for detection of the malarial biomarker *Plasmodium falciparum* [270] and also as a biomarker elsewhere [138, 271–273]. Blood drop patterns are used extensively for diagnostic purposes [20]. *Crack patterns* in dried drops of biofluids are also used to extract valuable information in medical diagnosis. Dried droplets of blood and blood serum show characteristic crack patterns for patients suffering from anemia, hyperlipidemia, and other disorders [7, 21]. A related field where droplet patterns are of importance is forensics related to crime investigation [7]. More such areas are biopreservation [216, 274] and high-throughput drug screening, where pattern formation in the drying sample is not desirable [60].

Some other applications have come up too; patterns on dried droplets may be used for quality analysis of food grains [275], as well as alcoholic drinks [276], fast identification of fluid and substrate chemistry based on automatic pattern recognition of stains [183], assessment of quality of products [275], and Raman spectroscopy [180, 277–286].

4.3. Droplet Levitation. The idea behind drying a droplet under levitation is to eliminate the effect of gravity during drying and observe desiccation under *no contact* condition. However, the droplet levitation technology is a promising candidate for generating novel applications. For example, the technique of manipulating levitated drops used on liquid marbles [287] may be applied to insert desired components in dried shell structures [66] for drug delivery or other applications.

There is, however, one concern with evaporation under levitation. This is that although one can avoid substrate contact and maintain the evaporating drops in a quasi-spherical shape, the levitation techniques, for instance, acoustic levitation, often lead to an additional boundary layer to the levitated drop, which may influence the evaporation process. In addition, the levitation force could also influence the morphology of dried residues. To investigate the mass transfer and evaporation-driven assembly in a truly undisturbed manner, drop evaporation in a space station is highly desirable. This will be of great help in understanding the emergence of crust, formation of cavity, buckling of crust, and elucidating the effect of gravity on these processes.

5. Conclusion

There obviously remains much more work to be done in this interesting and useful area of research. At this juncture, some tasks can be specially emphasized:

(i) Obtaining new experimental data critically needed for the design and development of adequate models

(ii) The development of 3D models describing the redistribution of the components, the movement of the phase front, and the evolution of the profile of the drying colloidal droplets with salt admixtures. In these systems, phase transition from sol to gel is concentration-driven. The thermal phase transition from liquid to vapor also takes place in this system. This phase transition leads to a movement of the liquid-vapor phase boundary (i.e., the droplet volume decreases and droplet profile changes)

(iii) Considering additional effects that may be crucial to understanding the processes of pattern formation but have not yet been included in the models (e.g., variations of the viscosity of a colloid with time and concentration of salts and changes of the vapor flux above the free surface of the droplets when the phase boundary (sol-gel) is moving). Time-varying interactions between the components of the droplet (e.g., particles and ions) also need to be considered

(iv) Studying the effect of external fields such as electromagnetic and acoustic fields on the droplet evaporation process

(v) Analyzing the final pattern through tools such as fractal and multifractal characterization

To conclude, the simple but effective process of drying a fluid drop and observing it under a microscope (preferably with video recording) is rapidly developing into a new and exciting field of research. Several reviews [15–17, 20, 288, 289] and books [5–8] based on pattern formation during desiccation published within the short span of just four years confirm the intensely growing interest in this subject. Exploiting the full potential of this topic in basic science research and applications needs involvement and interaction between scientists and engineers from disciplines of physics, chemistry, biology, medicine, and other related fields.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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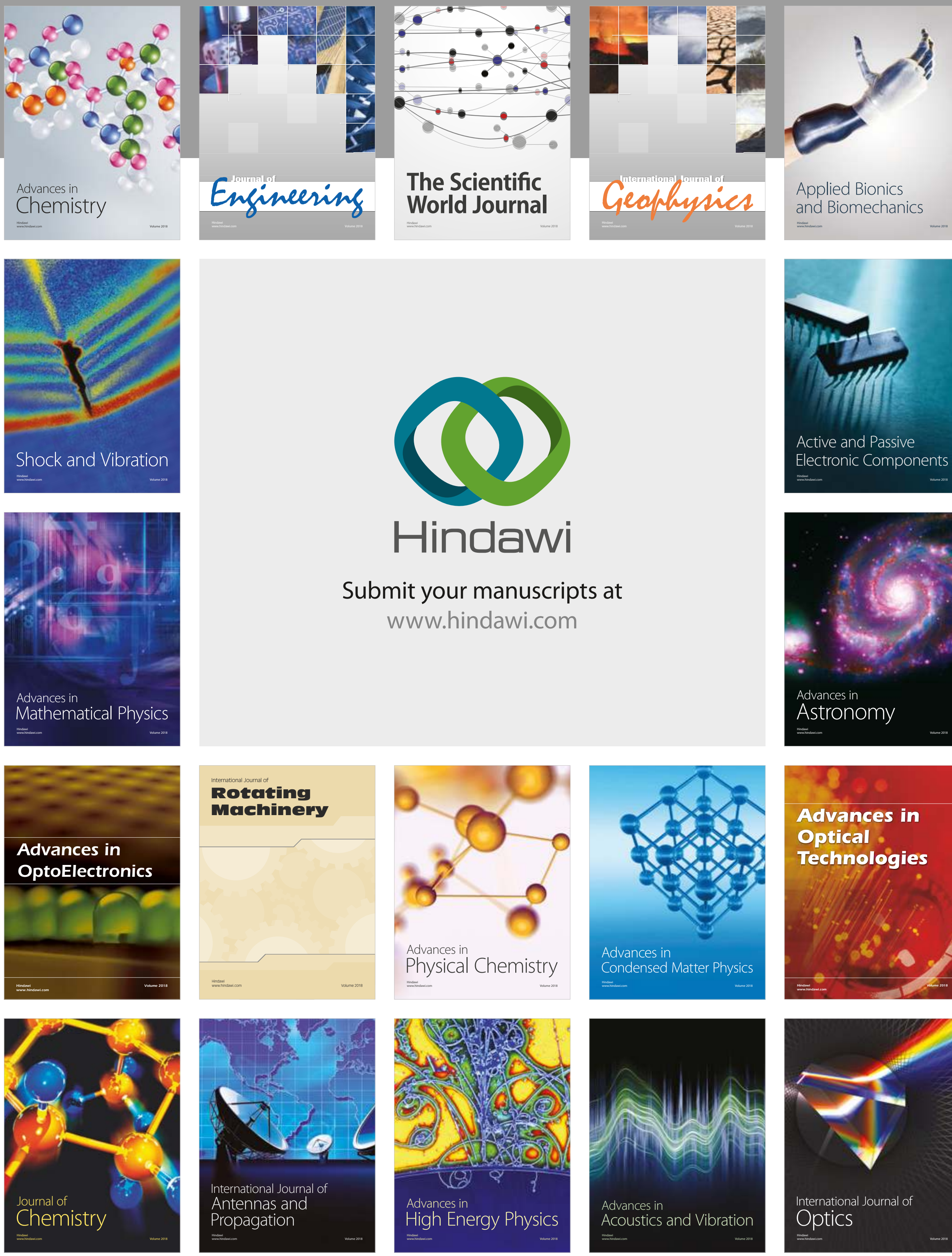
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