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Review
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Thermophoresis or When Small Objects Meet Temperature Gradient: Numerous Applications

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Abstract
This mini review discusses the phenomenon of thermophoresis, also known as the thermophoretic effect. Thermophoretic effect arises from the combination of a temperature gradient and particles of very small dimensions, on the order of magnitude of the mean free path of the molecules of the surrounding gas. Despite being a little-known effect, it is critical to many physical and chemical processes and for characterising the properties of nanostructured materials that could be used in industry for sensing applications. A description and definition of otherwise very similar thermophoresis terms is provided, as well as a brief overview of the literature on this topic, with a focus on research in the twenty-first century.

Keywords
Thermophoresis, photophoresis, thermomechanical effects, thermophoretic force, temperature gradient

1 Introduction

The thermophoretic effect is a thermo-mechanical phenomenon that results from the absorption of electromagnetic radiation on a material (Fig. 1). Incident radiation creates a temperature gradient, which further fuels heating and material expansion.[1]

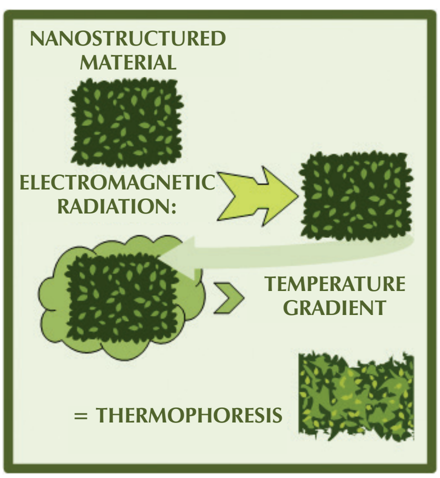

Fig. 1 – Schematic presentation of thermophoresis
Slika 1 – Shematski prikaz termoforeze

The physical properties of polymers and biophotonic structures are determined at the mesoscopic level by their internal architecture, and are not directly dependent on the chemical structure of the material. The correlation length specifies the average distance between two structural elements (polymer chain, nano-geometry, *etc.*). When the correlation length (structure dimension) corresponds to the molecule's mean free path and interacts with electromagnetic radiation, a force is formed, the effect of which is visible at the macroscopic level; this force is known as radiometric force.[2] As pointed out previously, the existence of a temperature gradient, in addition to the previously stated structural dimension, is crucial for producing the desired effect.[1] In this regard, a simple experiment was conducted: for the purpose of detecting the thermophoretic velocity in microgravity, an experimental setup was created.[3] This apparatus (see Fig. 2) had the ability to quickly replicate an

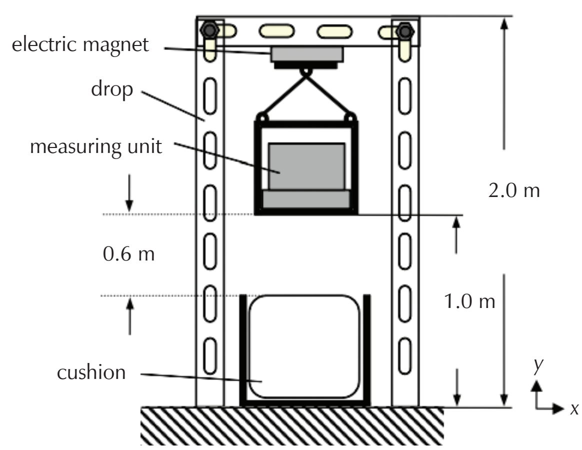

Fig. 2 – Schematic of apparatus for detection of thermophoretic velocity of particles in microgravity conditions[3] (CC BY 3.0)
Slika 2 – Shematski prikaz aparature za mjerenje termoforetske brzine čestica[3] (CC BY 3.0)

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experiment. Among regular thermophoretic experiments, one with no temperature gradient was carried out, which verified no particle movement.[3]

The method of holographic interferometry proved to be another interesting method for detecting the thermophoretic effect, that is, for monitoring deformations of nanostructures. This method is described in detail elsewhere.[4] The holographic approach is extremely useful since it monitors changes at the nano-level in real time. Furthermore, this approach has no effect on the deformation itself.

This effect will be crucial in the field of structured nanomaterials. It is especially important for materials that may be heated as part of their application. Understanding the behaviour of the corrugated structure under different conditions, such as interaction with light of different wavelengths, is critical for selecting suitable materials for various applications.[1]

When the nanodimensions of the observed effect are taken into account, we cannot rely with confidence on the laws of classical physics that apply at macro-levels[5]. For particles with dimensions on the order of magnitude of the mean free path of the molecules in the surrounding gas, the photophoretic force can be greater than the gravitational force, causing movement against the law of gravity.[6] Furthermore, in order to observe the specific structures of materials as well as their chemical and physical properties, thermophoresis and photophoresis must be related with intermolecular interactions within the material, and thus the studies enter the field of quantum physics.[7,8]

The thermophoretic effect has piqued the interest of scientists over the past two centuries because, on the one hand, it is enigmatic and unusual, and on the other, it is practically applicable for research and confirmation of other phenomena in the world around us, that is, in the world reduced to the micro- and nano-scale.[9] Theoretically, there are numerous applications for the studied phenomenon in the world of science. Scientists have already experimented in the fields of astronomy and avionics,[10–12] chemistry,[13–16] physics,[17–18] and this effect also contributes to pharmacy and medicine.[19–21] In practically any environment with extremely small object dimensions and a temperature gradient, one of the aforementioned cases of the effect will manifest.

2 Term definitions

Keeping in mind the many similar and closely related terms in this delicate area, as well as some inconsistencies, i.e., ambiguities in the literature, the differences and similarities between radiometric force, thermophoresis, thermophoretic effect and force, and photophoresis, photophoretic effect and force, will be clarified first.[1]

Radiometric force is a consequence of the non-uniform distribution of radiation energy, *i.e.,* the result of the interaction between the fluid and the unevenly illuminated/heated surface.[22] This force represents a component of the *photophoretic/thermophoretic force*, which depends solely on the temperature gradient and the characteristic dimension (nano, meso correlation length "η") of the material. *The characteristic dimension* of the nanostructure must match the size of the mean free path of the molecule to produce a force, and this relationship is characterised as the dimensionless Knudsen number, which is used in practice as a parameter for gas flow analysis.[1] This quantity is named after the Danish physicist, Martin Knudsen (1871–1949).[23]

It should be emphasized that *thermophoresis* is a process of movement of particles, typically in a gas around the considered material, which is conditioned by the existence of a temperature gradient (*e.g.,* due to different absorption of thermal radiation) on the material. On the other hand, *thermophoretic force* is a force that leads to the deformation of the material, and arises after the temperature gradient occurs on the same.[1]

A special case of thermophoresis is called *photophoresis*. The *photophoretic effect* is caused by the action of sufficient light intensity, which creates a non-uniform temperature distribution, and the *photophoretic force* causes the effect of material deformation due to the effect of light radiation on small-sized particles.[1]

Apart from the cause of the temperature gradient, the only difference between the described effects is that, due to photophoresis, the gradient does not spread over the entire material.

3 The very beginnings of the thermophoretic effect

The first hints of the thermophoretic effect were observed, in the form of material movement due to the appearance of a temperature gradient, by Augustin-Jean Fresnel (1788–1827), a French engineer and physicist known for his achievements in the field of optics, and William Crookes (1832–1919), an English physicist and chemist.[24] Later, the phenomenon was investigated and observed by numerous scientists, until Felix Ehrenhaft (1879–1952), an Austrian physicist, produced it with light radiation and defined it as photophoresis.[25]

In 2002, Passian experimentally demonstrated that by heating microcantilevers with laser beams, if they are at a distance comparable to the mean free path of the surrounding gas molecules, certain forces are created that act on the observed microcantilevers.[26]

Scientific research at the end of the twentieth century was mostly focused on the application of the thermophoretic effect in gas systems.[27,28] During this time, the effect was also mainly used to study channel/pipe flow.[29,30]

4 Thermophoretic effect: The twenty-first century

Since its discovery, the thermophoretic effect has not been thoroughly investigated. Until recently, the application of this effect was mostly limited to polymer solutions and other colloidal fluids. When discussing the process's nature, thermophoresis is described as a non-equilibrium transport process with the potential to play an important role in macromolecular fractionation as well as microfluidic manipulation.[31] When mentioning thermophoresis as a non-equilibrium effect, it is important to highlight that it can be observed in different systems. When it comes to applications in chemistry, *Talbot et al*.[13] used thermophoresis to exploit directed motion of liposomes. This is significant because it demonstrates the ability to segregate lipids based on their head group by utilising this effect (Fig. 3).[13]

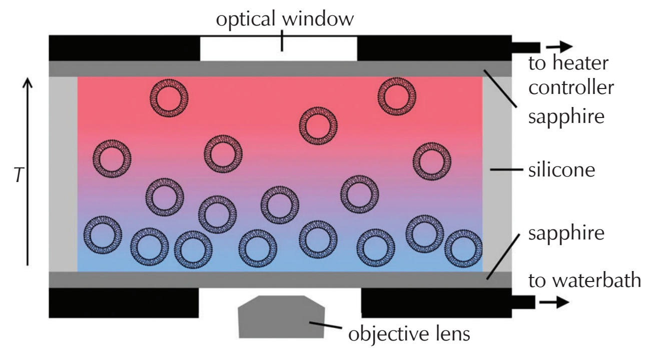

Fig. 3 – Schematic cross-section of the temperature cell used for monitoring thermophoresis of single-type lipid vesicles. An example of thermophoretic effect exploitation in chemistry.[13] (CC BY 4.0)

Slika 3 – Shematski prikaz poprečnog presjeka temperaturne ćelije upotrijebljene za praćenje termoforeze lipidnih mjehurića. Primjer primjene termoforetskog efekta u kemiji.[13] (CC BY 4.0)

As previously stated, it has a wide range of applications in chemistry, and can be applied to any chemical reaction system that is diffusing along a temperature gradient (see Fig. 4).[16] A group of authors[16] discussed how most microscopic systems exhibit thermophoretic behaviour, requiring heat to be transported from warm to cold regions. The conclusion is that thermophoresis occurs in all systems that have internal states, whether structural or chemical.[16]

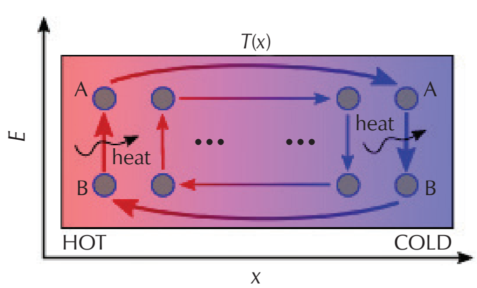

Fig. 4 – Schematic representation of temperature absorption on the warm side, followed by diffusion and particles moving to the cold side. After cooling the particles and their transition to low energy states, the cycle repeats.[16] (CC BY 4.0)

Slika 4 – Shematski prikaz apsorpcije temperature na toploj strani praćenoj difuzijom i pomicanjem čestica na hladnu stranu. Nakon hlađenja čestica taj ciklus se ponavlja.[16] (CC BY 4.0)

It is mostly used in aerosol systems, particularly as a tool for particle manipulation, such as particle deposition technology for particle collection or a thermal precipitator for sampling nanoparticles on two parallel plates.[32] Another example suggests that thermophoresis can be used to control the motion of water nanodroplets confined in carbon nanotubes. Because wetting the water-carbon interface has no effect on thermophoresis, this mechanism was demonstrated to be successful in this case.[33] These studies proposed thermophoresis as a method for controlling particle separation as an ideal process for use in microfluidic devices that employ appropriate temperature control strategies.[34]

A group of authors[35] conducted a numerical investigation into some factors influencing thermophoretic effect. They discovered the influence of particle density, airflow velocity, and thermal conductivity. However, the thermal conductivity ratio between the particle and the air had no effect.[35]

At one point, thermophoresis was represented as a rectification of Brownian motion. The observed microgel particles in this study demonstrated unusual thermophoretic properties.[36] Until recently, the most common application of the thermophoretic effect was as a mass transfer mechanism.[37] It can thus be used for chemical reactions with characteristic particle deposition, which is related to thermophoresis.[38] When discussing the influence on heat transfer such as through nanofluids, the thermophoretic effect is also important.[38,39] Some flow field studies consider micropolar nanofluids due to the thermohoretic effect.[40,41] There are numerous thermophoresis-related heat and mass transfer studies. One of these is investigated in an induced magnetic field by observing the flow through a vertical cone.[42]

The thermophoretic effect is mostly used in engineering to remove small particles.[43] This effect is also useful for other methods, such as holographic techniques.[4,44] Thermal Diffusion Forced Rayleigh Scattering is a technique for detecting thermophoretic behaviour in liquids. The point is that certain chemical interactions in liquids are causing the shifts in thermophoretic behaviour nature.[45] Aside from that, there is the holographic method, which uses the thermophoretic effect for various types of research, such as studying biophotonic materials or oscillatory reactions.[4,46]

This phenomenon was even investigated by NASA. As a result, when discussing the prospects for microgravity research, the scientists also considered the thermophoretic effect.[47]

A review of the thermophoretic effect in solid-state particles may be even more important in terms of technology, industry, and material sciences. To begin with, it is demonstrated that thermophoresis has an effect on solids. Of course, when discussing thermophoresis, only very small particles are taken into account.

While developing a new plasma spraying process it was discovered that small solid zirconia particles are quite sensitive to thermophoretic effect.[48] These particles move away from the plasma jet, which could represent the previously described solid-state model of fluid motion. Also proposed is a mechanism based on this effect for studying DNA translocations through plasmonic nanopores.[49]

Many scientists have observed thermophoretic motion of solid nanoparticles. Schoen's work described how thermophoretic force caused mass transport of gold nanoparticles inside carbon nanotubes.[50] It has also been demonstrated that the movement of graphene oxide nanosheets is dependent on thermophoresis, among other.[51] The investigation of nanotransportation and nanoparticle positioning identified thermophoresis as a suitable technique for manipulating solid nanoparticles. *Becton et al.* demonstrated how the motion of a nanoflake is controlled by thermal gradient; therefore, thermophoresis has been used to move nanoparticle on a solid surface.[52] The complex interaction of small oxide smoke driven by thermophoresis with aluminium particles again demonstrated thermophoretically affected nanoparticle motion.[53]

An example of thermophoretic effect application was studied to improve understanding of the behaviour of soot particles in combustion fields and devices. Carbon black particles were observed in this study, and the relationship between thermophoretic velocity and bulk density was discovered.[54,55] When studying soot formation in two-phase combustion, thermophoresis was revealed to be an important contributor to high volumetric fractions.[56] One concrete application is the use of the thermophoretic effect as a propulsion mechanism for polymeric nanomotors. These motors are designed to generate motion as a result of the aforementioned effect.[57]

At the turn of the century, an intriguing concept was proposed. Specifically, thermophoretic forces were used to form and position the dust structures. This was referred to as "thermophoretic traps".[58] This premise later evolved into the invention of optical tweezers. Thermophoretic manipulation of nanoscaled particles is obviously the foundation of opto-thermophoretic tweezers.[59] Because they do not use lasers, these thermophoretically based optical tweezers have a significant advantage in terms of not damaging biological samples. Instead, fluid flow fields are employed, for example, near the heated gold surface (as seen in Fig. 5).[60]

Because the presence of a temperature gradient is a necessary condition for the thermophoretic effect, the influence of this gradient on the effect must be investigated. Keeping this in mind, the thermophoretic effect was investigated at various levels, the most important of which was the investigation of solid-state nanopores.[61] This study predicted a successful outcome in biosensing applications. In this regard, microscale thermophoresis was proposed as a new biophysical method for studying the binding of large biomolecules and small molecules.[62] Our research group investigated the thermophoretic effect in biological samples.[1] The butterfly wing is an excellent example of structured materials that can be used in infrared radiation sensors.[63] This same thermophoretic effect was used to examine the impact of various wavelengths of light on heating and deformation of biological structures.[1] The thermophoretic effect is caused by heating the wing structure with laser

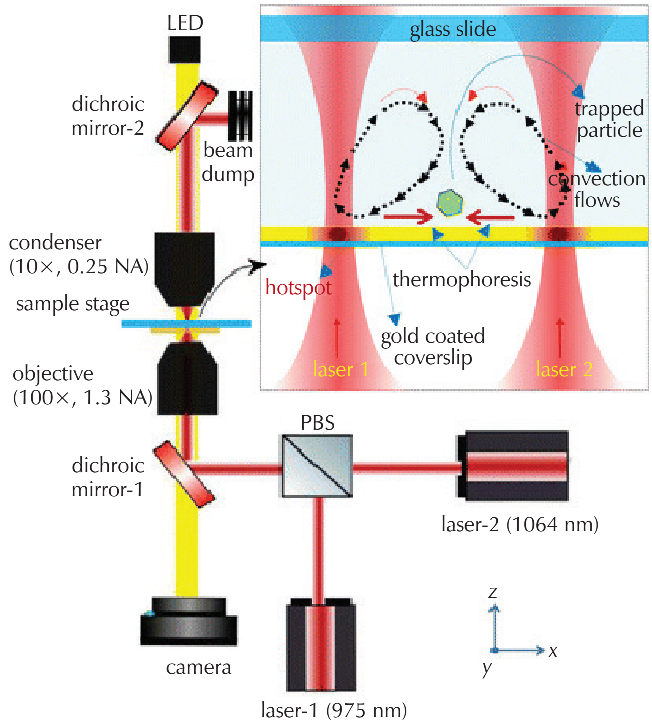

Fig. 5 – Schematic diagram of optical tweezers setup[60] (CC BY-NC 3.0)

Slika 5 – Dijagram postavke eksperimenta s "optičkim pincetama"[60] (CC BY-NC 3.0)

light. The influence of this effect on the observed microstructure was monitored holographically.[1,4] Fig. 6 depicts a hologram[4] recorded during the induced effect and its reconstruction.

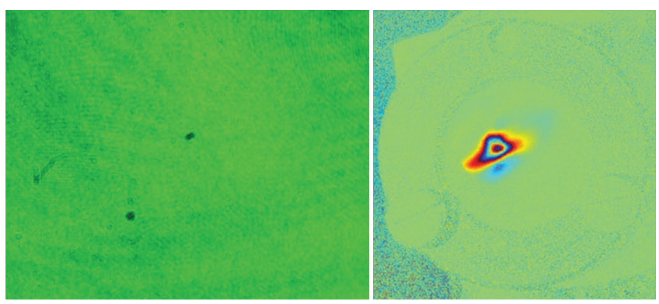

Fig. 6 – Images selected at random from holographic monitoring of the thermophoretic effect on a butterfly wing: hologram (left), and holographic reconstruction (right). Holographic analysis was carried out in order to discover new sensing applications.[1]

Slika 6 – Holografsko snimanje termoforetskog efekta na krilu leptira: hologram (lijevo) i holografska rekonstrukcija (desno). Holografska analiza izvedena s ciljem pronalaska novog mehanizma detekcije.[1]

The reconstruction recorded the resulting changes in microstructure. The dynamics of the effect and its influences were defined by comparing and analysing a large number of reconstructions that record different moments of the heating process, *i.e.,* deformation.[1] This is a good example of utilising the thermophoretic effect in a sensing application. It is crucial to highlight that thermophoresis is an unstudied phenomenon, and new discoveries and scientific research studies on the subject are anticipated.

5 Conclusion

The thermophoretic effect was introduced in this review. This is an effect in which the micro/nano-particles move under the influence of the thermophoretic force created by the temperature gradient on the material/fluid. This force arises due to the size of the particles, which are of the order of magnitude of the mean free path of the molecules of the surrounding gas, within which the temperature gradient is created. This effect plays a role in the processes within practically all micro/nano-systems, so it is of great importance for fundamental sciences, such as physics and chemistry, as well as many others. For the first time, terms related to this effect are precisely defined, and a thorough analysis of research on this topic over the past two decades is presented. However, thermophoresis is a relatively new concept and its applications have yet to be developed.

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SAŽETAK

Termoforeza – ili kad se susretnu mikroobjekti s gradijentom temperature: brojne primjene

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Ovaj pregled bavi se fenomeom termoforeze odnosno termoforetskog efekta. U pitanju je efekt koji nastaje kao posljedica kombinacije temperaturnog gradijenta i čestica veoma malih dimenzija, reda veličine srednjeg slobodnog puta molekula okolnog plina. Radi se o ne tako poznatom efektu, koji je od velikog značaja za mnoge fizikalne i kemijske procese, kao i za karakterizaciju svojstava nanostrukturiranih materijala koja može imati primjenu u industriji za različite detektore. Dan je i opis definicija inače veoma sličnih pojmova iz područja termoforeze, kao i sažet pregled literature na ovu temu, s posebnim osvrtom na istraživanja u dvadeset prvom stoljeću.

Ključne riječi
Termoforeza, fotoforeza, termomehanički efekti, termoforetska sila, temperaturni gradijent

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