ABSTRACT: Flame spray pyrolysis of precursor–solvent combinations with high enthalpy density allows the design of functional nanoscale materials. Within the last two decades, flame spray pyrolysis was utilized to produce more than 500 metal oxide particulate materials for R&D and commercial applications. In this short review, the particle formation mechanism is described based on the micro-explosions observed in single droplet experiments for various precursor–solvent combinations. While layer fabrication is a key to successful industrial applications toward gas sensors, catalysis, and energy storage, the state-of-the-art technology of innovative in situ thermophoretic particle production and deposition technology is described. In addition, noble metal stabilized oxide matrices with tight chemical contact catalyze surface reactions for enhanced catalytic performance. The metal–support interaction that is vital for redox catalytic performance for various surface reactions is presented.

INTRODUCTION

The value of the global nanoparticle market at $1.1 billion in 2018 is expected to rise to $2.2 billion by 2025, growing at a compound annual growth rate (CAGR) of 10.5% from 2019 to 2025.1 Fundamental research and industrial R&D of these materials focus on their implementation in devices such as new catalysts, gas-to-fuel energy conversion systems, catalytic converters in vehicles and power plants, electrode fabrication for batteries, fuel cells, solar converters, and chemical sensors. In all of these advanced applications, materials and/or fabricated layers require sufficient specific surface area for fast and efficient mass/charge transfer. The materials in their metastable states are key to many above-mentioned applications due to unique inherent properties. The realization of the high material performance depends on the specific chemical nature of the material, the material–material interface at the nanoscale, and the device fabrication technology. Apart from the functional properties of the materials and/or device configurations, the nanoparticle layer is required to be robust. Considering all the properties of these designed materials (homologous materials with different doping, functionalization or mixtures) and layers, the key to the industrial realization is the cost-effectiveness and the viability of the synthesis, e.g., production volume and usage of different metal salts and/or organometallics either as a source of the parent matrix (oxide support utilized for the catalytic and/or sensor investigation) or the active site in the material.

Different syntheses pathways including solid state reactions, precipitation, and impregnation in aqueous and/or non-aqueous liquids, aerosol and plasma synthesis produce materials that are specific to their physicochemical properties (e.g., shapes, sizes, surface functionalization, and doping) and performance (see Table 1 for details). The gas-phase synthesis technique flame spray pyrolysis (FSP), developed in the last two decades, is versatile in nanoparticle synthesis. The spectrum of nanoparticles that could be produced was greatly enhanced by flame spray pyrolysis (FSP) that relies on the direct introduction of the liquid and non/volatile precursors into the premix flame producing highly dispersed, ultrafine, and single crystalline powders.2,3 Because of the broad range of precursors available, FSP is one of the most useful single-step techniques for the synthesis of binary, mixed binary, ternary, and mixed ternary metal oxide NPs.2,4 The particles formed following two different routes: (1) crystal seed formation within the liquid droplet at low temperature (droplet-to-particle formation) and (2) nucleation in the gas phase at high temperatures (gas-to-particle formation) (Figure 1A,B).

In flame aerosol synthesis, gas-to-particle is the dominant route to ultrafine single and/or multicomponent particles provided that the precursor–solvent combinations have high combustion enthalpy density5–9 (Figure 1B). Metal organic precursors, namely, alkoxides, acetylacetonates, and naphthenates, with high combustion enthalpy density vaporizes at the flame operation temperature, while metal nitrates, chlorides, and/or carbonates take up energy from the flame for combustion (endothermic combustion) giving rise to larger/hollow metal oxide particles (Figure 2A–D). During flame spray pyrolysis, low combustion enthalpy density (<4.7 kJ g–1 gas) and higher melting/decomposition temperature of the metal precursor compared to the boiling point of the solvent results in an inhomogeneous particle mixture via droplet-to-particle (incomplete droplet evaporation) and/or gas-to-
| Property | Description |
|----------|-------------|
| Primary particle size | The particle sizes are in the range of 3 nm − 80 nm. |
| Purity | Low temperatures in hydrothermal, sol-gel, and precipitation methods might give impurity phases due to the high molecular organic solvents. Hydrothermal or sol-gel processes are extraordinary in size and morphology control. Metastability is difficult. Morphology control is possible via polymer/surfactants in precipitation, sol-gel, and hydrothermal processes. Metastability, defect density, kinetic control are also possible in the chemical routes of particle synthesis. Size control is difficult. Site control is possible through precursor feed, concentration, dispersant O2 flow, and the combination of chemistries and the particle formation model using gas phase chemical kinetics. |
| Capping | Noble metal can be stabilized on the metal oxide matrix (Pt, Pd, Ag/CeO2, Al2O3, NiO, Co3O4, ZrO2, CuO, SnO2, SrFe12O19, pure and Fe doped TiO2). Doping: All the particles can be doped through mixing precursors before combustion or by doping metal oxide NPs, e.g., Al and/or CeO2 doped YSZ, Ln doped Y3Al5O12, e.g., La doped SnO2, Gd doped CeO2, mullite formation (3Al2O3-2SiO2), Sn3+ doped Sr2CeO4, Cr3+ doped hydroxyapatite, Eu doped Y2O3, Sn-doped In2O3, Fe doped TiO2, Co doped TiO2, Fe2O3 doped ZnO, Al doped ZnO, Zn doped Bi2O3. Doping with PEI (MSNPs modified with exposed –NH2), capped SiO2, microencapsulation with metal oxides. |
| Porosity | The particles are nonporous. However, there are reports of porous powders, e.g., La2Ti2O7/CuO. In a separate report, the crystal morphology of In-doped ZnO is known to change from spheroidal (pure ZnO), to cuboid, and to rodlike with an increase in the indium content. In spray pyrolysis, particles are microporous (d< 2 nm) spherical solids. |
| Morphology | Morphology control is possible via polymer/surfactants in precipitation, sol-gel, and hydrothermal techniques. Morphology control from combustion techniques is rather rare and known for SiO2, TiO2, and Al2O3 obtained via vapor phase synthesis and adapting various precursor feed, concentration, dispersant O2 flow, and the combination of chemistries and the particle formation model using gas phase chemical kinetics. Applications of these particles include: (1) catalysis (NOx catalyst, selective CO oxidation, oxidative hydrogen production, Fischer-Tropsch) (2) photocatalysis (5) optics, electronics and phosphors. |
| Size control | Particle size control is possible through precursor concentration during combustion. Size control is possible through precursor feed, concentration, dispersant O2 flow, and the combination of chemistries and the particle formation model using gas phase chemical kinetics. Applications of these particles include: (1) catalysis (NOx catalyst, selective CO oxidation, oxidative hydrogen production, Fischer-Tropsch) (2) photocatalysis (5) optics, electronics and phosphors. |
| Metastability, defect density, kinetic control | Usually metastable particles can be designed using the gas phase synthesis route with an appropriate soft-chemical route at low temperatures. Defect density, metastability, kinetic control are also possible in the chemical routes of particle synthesis. Size control is difficult. Site control is possible through precursor feed, concentration, dispersant O2 flow, and the combination of chemistries and the particle formation model using gas phase chemical kinetics. Applications of these particles include: (1) catalysis (NOx catalyst, selective CO oxidation, oxidative hydrogen production, Fischer-Tropsch) (2) photocatalysis (5) optics, electronics and phosphors. |

The physical properties of the particles obtained via chemical routes are similar to those obtained from the gas phase synthesis method. While specific properties such as wide varieties of morphologies and solid state reactions might give impurity phases due to the high molecular organic solvents. Hydrothermal or sol-gel processes are extraordinary in size and morphology control. Metastability is difficult. Morphology control is possible via polymer/surfactants in precipitation, sol-gel, and hydrothermal reactions. Metastability, defect density, kinetic control are also possible in the chemical routes of particle synthesis. Size control is difficult. Site control is possible through precursor feed, concentration, dispersant O2 flow, and the combination of chemistries and the particle formation model using gas phase chemical kinetics. Applications of these particles include: (1) catalysis (NOx catalyst, selective CO oxidation, oxidative hydrogen production, Fischer-Tropsch) (2) photocatalysis (5) optics, electronics and phosphors.
particle routes (supersaturation of the metal vapor in the gas phase). In addition, the precipitation of the metal precursor at the surface of the droplet followed by fast evaporation gives rise to hollow particles (Figure 2A–H). When the solutions with high combustion enthalpy densities are flame sprayed, the exothermic reaction in the gas phase yields ultrafine homogeneous particles (Figure 2K–T). Such particles are obtained when the pure metal precursors are (1) in liquid phase at ambient temperature, (2) readily soluble in combustible organic solvents, (3) volatile and with low viscosity, and (4) commercially available.3

To obtain homogeneous particles (Table 1), either the process temperature is increased (e.g., using reactive gases with high combustion enthalpy density) or via selection of exothermic precursor–solvent systems. The possibility of doping beyond solubility limits, surface modifications in the gas phase, scalable production of binary, mixed binary, ternary, and/or mixed ternary composites makes the gas phase particle synthesis unique.

## PARTICLE SYNTHESIS FROM FLAME SPRAY PYROLYSIS

During the flame aerosol synthesis, the liquid precursors that are introduced to the flame subsequently vaporize followed by atomization and chemical reactions in the gas phase forming NP cluster seeds.40 The atomization is achieved with a fluid nozzle that utilizes high gas velocities to disintegrate the liquid into fine droplets.41 Rosebrock et al. reported the single droplet combustion of the precursor droplets, where the nature of the precursor influences the decomposition and the droplet burning properties. During combustion, the volatile solvent vaporizes from the droplet surface leaving behind the low volatile components leading to the surface heat-up as the droplet moves closer to the flame (mass diffusion is lower than the heat).42 The high boiling point of the precursor on the droplet surface causes an increase in the surface temperature and eventually precursor decomposition.43 The decomposition gives rise to gaseous, liquid, and/or solid intermediates where the liquid/solid intermediates form a viscous shell, hindering vaporization. Such hindrance triggers superheating of the low volatile shell and causes vapor bubble formation at the shell interface via heterogeneous nucleation. The droplet disruption takes place when the vapor pressure in the bubble is greater than in the shell.42 While the report described that the combustion of the precursor using single droplet significantly differs from the common droplet-to-particle formation model, additional investigation was necessary to validate the model based on the shell formation and breakage.44 In another report, Rosebrock et al. described that the occurrence of spontaneous microexplosions despite the precursor volatility or nature of the solvents in flame spray pyrolysis and/or single droplet combustion gave rise to homogeneous nanoparticles. Such explosions are possible for precursors decomposing in multiple steps over a wide range of temperatures and boiling points higher than those of the solvents. In contrast, nonhomogeneous particles are expected during droplet-to-particle conversion using precursors and water miscible solvents that induce flame extinction (endothermic precursors reduces the flame temperature significantly). However, homogeneous particles can be realized if the decomposition/volatilization temperature of the precursor-solvent is lower than the flame temperature.42 Hence, the strategic design of precursor–solvent combinations (even metal salts such as metal nitrates, chlorides) that follow microexplosion during combustion would allow homogeneous nanoparticle production. Li et al. reported single droplet combustion of metal organic precursors such as tin ethylhexanoic acid dissolved in xylene (Figure 3). The onset of droplet combustion is immediately after the ignition, followed by the first microexplosion.44 The occurrence of multiple explosions gives rise to well-formed particles without any end residues. The SnO₂ particles had mixed sizes ranging from <10 nm to >20 nm. The larger particles were probably due to the first microexplosion where the fuel is atomized into small fragments carrying the precursor from the inner part of the droplet shell and the combusted intermediates around the shell in the flame front.45 Such fragments spread on the remaining surface and are entirely exposed to the available near surface oxygen to burn from outside to inside (observed through large bright white flame) producing microexplosions (see Figure 3).
The droplet explosion event is accelerated due to (1) fast evaporation of the liquid fuel into the gas phase and (2) the fast reaction progress during secondary atomization. The exothermic heat release during the microexplosion events causes fast sintering, promoting homogeneous nanoparticle formation via gas-to-particle conversion as shown for volatile nitrate precursors. In a similar report, Li et al. studied single droplet combustion of xylene, isobutanol, and ethanol in dry and wet atmospheres at different oxygen concentrations. The decrease in oxygen in the presence of dry and wet atmospheres decreases the burning constant and increases the lifetime of the droplets in the flame. The water vapor diffusing from the atmosphere had a negligible effect on xylene, a slight effect on isobutanol, and a significant effect on ethanol due to differences of water solubility in these solvents (solubility_{xylene} < solubility_{isobutanol} < solubility_{ethanol}). The burning ethanol droplets absorb water vapor from the flame and/or flame vicinity resulting in premature droplet combustion to give a C-rich residue. While suitable liquid fuels can avoid the residual formation, the presence of an oxidizing environment accelerates complete droplet combustion. The investigation of single droplet combustion using a variety of precursor–solvent combinations: (1) with high combustion enthalpy density, (2) producing a low amount of water vapor (as byproduct of the reaction) during combustion, and (3) with lower decomposition/volatilization temperature leads to highly crystalline and homogeneous particles. To validate similar chemistries taking place in the single droplet combustion and flame spray pyrolysis, single droplets of different lithium and titanium precursors and solvents were tested. The solutions with highly volatile component vaporizes from a thin layer of the droplet causing a linear decrease of the spherical droplet diameter. The accumulation of low volatile components hinders further evaporation inducing rapid heating of the droplet, followed by decomposition. The further increase in droplet temperature superheats the low-volatile components trapped inside the droplet forming vapor bubbles via...
heterogeneous nucleation and final explosion of the droplet giving phase pure Li4Ti5O12.7 The combustion phenomenon for more complex ternary systems such as Y4Al2O9 was also studied. The addition of 2-ethylhexanoic acid in the solution of Al and Y precursors dissolved in xylene and toluene, respectively, allowed COO2−Y3+ complexation as observed by Meierhofer et al. to form a stable solution. The accelerated mass transfer in the gas phase during FSP of volatile precursors is responsible for microexplosions (gas-to-particle) during the formation of pure Y4Al2O9.5,48

■ LAYER FABRICATION PROCEDURE

The layer fabrication requires a strong contact between the coated layer and the substrate. Such fabrication technologies including drop coating, dip coating, spin-coating, and doctor blading involve liquid pastes designed using a defined volume of the material and organic binders spread over the substrates. Such methods have both advantages and shortcomings over the nature of the coating—substrate contact for performance evaluation. In these traditional coating techniques, the layer is created via particle rearrangements during sintering, i.e., evaporating the organic components from the paste triggering in and/or out-of-plane stress on the substrate inducing either the substrate bending and/or cracks on the layer. Hence, a crack-free and homogeneous surface coating is difficult to obtain in a single experimental step via chemical routes.50 In gas-phase deposition, thermophoresis transports particles from the high velocity aerosol stream to the substrate.50 Such layer fabrication technique (1) avoids post treatments of the layers, e.g., drying the liquid organic binders, (2) the absence of capillary forces, and (3) in situ particle arrangements leading to a crack-free layer. The flame deposition is directly related to the temperature gradient of the gas-particle stream and the temperature of the substrate. The thermophoretic deposition rate is influenced by the increased temperature at the deposition layer leading to a reduced temperature gradient resulting in a decreased rate of film growth with increasing layer thickness.50 The disadvantage of this technology is (1) insufficient particle—substrate contact strength. The nanoparticles follow the aerosol stream and collide with the
substrate. This makes it challenging to coat temperature sensitive substrates, and (3) insufficient mechanical stability of the porous nanoparticle layers (degree of particle-substrate strength). While particle-substrate bonding is weaker for the thermophoretically deposited layers, Schopf et al. reported a new process capable of transferring a porous layer of TiO₂ nanoparticles (as a model example) to various substrates using pressure-based role-to-role lamination at room temperature. The thin/thick film fabrication process was feasible for powders obtained from gas phase synthesis on various substrates including temperature sensitive organic polypropylene foil (Figure 4a,b).

The results of the thermophoresis deposition followed by role-to-role particle transfer showed a mechanically stabilized nanoparticle layer. While the porosity of the thermophoretically deposited layer is almost 98%, the restructuring of the layer during role-to-role transfer was responsible for the homogeneous porosity over the layer. This effect is a key factor for gas sensing applications, while the optimized porosity enhances the electrical conductivity (Table 1). Hence, the development of the layer fabrication technology could have an immediate impact due to (1) porosity adjustment via lamination pressure monitoring, (2) abrasion resistant and layer stability in the liquid media. In another report, Baric et al. validated the experimental data for nanoparticle aggregates film compaction effect using the discrete element method (DEM). The simulated data were in excellent agreement with experimental data observed by Schopf et al. when elastic sinter bridges between primary particles were considered. The simulation of the single particle films assuming noncovalence, adhesion, very rigid, and strong agglomerates resulted in a deviation from the experimentally determined porosity and pore size distribution against the applied pressure (Figure 4c). While it is unlikely that the particles of this size could produce brittle fractures, the stable sinter bridges prevent breakage of the aggregates at 3.4 MPa compaction pressure.

**PARTICLES AND/OR LAYERS FOR GAS SENSORS**

The experimental and theoretical investigation (using the discrete element method (DEM)) indicates the compaction of the nanoparticle films with homogeneous porosity is a key factor for enhanced sensing and energy storage application. On the basis of the acquired knowledge of the layer fabrication procedure and film properties, thermophoretically deposited...
particles and/or layers were tested for their gas sensing properties. The in situ thermophoretic deposition of the sensor substrates was realized using a FSP system coupled with a coating unit where multiple sensors could be in situ deposited in a single spray. The sensors based on 0.2 and 2.0% Pt doped SnO2 were directly deposited onto interdigitated Pt-electrodes to form a porous (~98% porosity) thin film. These sensors exhibited high CO sensitivity, good reproducibility, and a remarkably low detection limit (1 ppm of CO in dry air at 350 °C). The knowledge acquired from a single film deposition was extended to multiple layer deposition on the sensing substrate. Two consecutive sensing layers were realized on ceramic substrates, pure SnO2 and Pd/SnO2 with a top filter layer of Pd/Al2O3 and tested for CH4, CO, and C2H5OH (Figure 5A). In the case of CH4, the pure SnO2 sensor with Pd/Al2O3 filter showed higher sensor signals and improved selectivity in relative humidity compared to pure SnO2 films. At higher temperatures, the high catalytic activity of the Pd/Al2O3 filter significantly influenced the sensitivity of the Pd/SnO2 sensor (Figure 5B–D). The results showed the potential of FSP to generate commercial sensors of high standards.

In the search for new sensing materials, Kemmler et al. designed a library of flame-made Sn-doped In2O3 NPs. At 43% of Sn in In, a new ternary metastable InSn0.1O0.9 was obtained (Table 1, Figure 6A–E). The thermophoretically deposited sensors tested using this metastable InSn0.1O0.9 outperformed the state-of-the-art metal oxide devices for air pollutants such as formaldehyde (Figure 6F). The InSnO12 phase was stable beyond the operation time and temperature, demonstrating its largely undiscovered potential. Recently, a review on gas sensing at the nanoscale described the state-of-the-art of the gas sensing FSP materials and their performance including both the p- as well as n-type material.

Apart from direct thermophoretic deposition of the sensors, Flame synthesized crystalline SnO2 NPs (using tin ethylhexanoate precursor in ethanol) were screen printed as thick film on the Pt-interdigitated substrates. The sensors showed high and fast response to both reducing and oxidizing/reducing gases. The work function change and conduction measurements of these thick film sensors in the presence of dry and humid CO showed significant variations in the electron affinity and caused band bending in the solid state for efficient charge transfer for sensing transduction, clearly suggesting the influence of humidity on the sensing properties. In a similar report, 0.2 and 2.0% wt % of Pt was doped in SnO2 to obtain 10 nm homogeneous powders. The sensing performance was reported to be outstanding with CO concentration as low as 5 ppm. The low Pt doped SnO2 outperformed the state-of-the-art with high stability up to 20 days. Recently, thermophoretically deposited chemoresistive Si doped ε-WO3 sensors were tested for acetone in the human breath to develop a marker for diabetes diagnosis. The results showed that metastable ε-WO3 was sensitive and selective to 20 ppb of acetone at an optimal doping concentration of 10%. These cost-effective solid state sensors were further developed to portable devices allowing fast detection of acetone in the breath. Analogous to ε-WO3, flame-made Si-doped α-MoO3 phase was also selective for NH3 in the human breath.

In another report, screen printed WO3 sensors were tested simultaneously for oxidizing (NO2) and reducing (CO) gases. The results showed that sensors with excellent baseline stability had very different CO response kinetics (2) tuning the morphology of the particles; the gas sensors based on WO3 could be enhanced due to different surface stabilities and reactivities.

The screen printed sensors from the particles obtained from chemical routes have been extensively studied compared to thermophoretically deposited layer and/or screen printed sensors from gas phase synthesized particles. The sensors have been tested for almost all reducing and oxidizing gases including volatile organic species. Srivastava et al. reported the development of an “electronic nose” printed with Pd, Pt, and/or Au functionalized SnO2 based sensor-arrays and an artificial system for the detection of volatile components including alkane, alcohol, ketone, benzene, and xylene. The data transformation and the ability of an electronic network via
differently sized sensor-arrays were applied to allow gas detection accuracy. Recently, electrospun SnO$_2$/MO$_x$ (i.e., M = Zn, Ga, and W) based heterostructured nanotubes and nanofibers were tested for ethanol, acetone, and xylene. These sensors were very selective against interfering gases such as formaldehyde, benzene, and toluene with an accuracy of 90% at a concentration of 10 and 20 ppm. Hydrothermally synthesized SnO$_2$ was molecularly impregnated on $5 \mu$m X 500 nm microsheets and tested for NH$_3$, acetone, LPG, O$_2$, and benzene. The sensors responded to 600 and 280 ppb of NH$_3$ and acetone respectively, with the sensitivity of 77%. In another report, a mesoporous SnO$_2$ sensor allowed precise detection of 0.5–1000 ppb of H$_2$S at an operating temperature of 92 °C and 85% relative humidity (RH). Such first sensors able to detect H$_2$S at such a low concentration had a large specific surface area for gas detection at a low concentration. Tournier et al. used Pd doped SnO$_2$ from Coreci Company to detect CH$_4$ aliphatic hydrocarbons and CO. The selectivity of the sensors for CH$_4$ and hydrocarbons was at an operating temperature of 400–450 °C, whereas CO selectivity was realized at a low temperature of 50 °C. These two sensors were recommended for domestic alarm and car pollution monitoring. In addition to binary oxides, ultrafine Bi$_2$WO$_6$ perovskite nanoparticles made by flame spray pyrolysis (FSP) offered a
fast response of 3.7 s to 2000 ppm pulse of acetone at 350 °C. It was suggested that further doping of such perovskites might significantly improve the response time of those sensors. In another report, flame spray pyrolysis was utilized to prepare BaTiO₃ perovskite thin film sensors with a fast response and selectivity to liquid petroleum. While gas phase synthesized particles and/or thermophoretically designed sensors are still in their early stage, they are reasonably similar in terms of sensitivity and selectivity to those observed from particles obtained via chemical synthesis routes.

**PARTICLES AND/OR LAYERS FOR ENERGY STORAGE**

The ESP-based thermophoretic layer fabrication procedure was also exploited to rechargeable lithium-ion batteries (LIBs). Conventional syntheses of Li₄Ti₅O₁₂ including solid-state, sol–gel, hydro-/solvothermal, combustion, and gas phase synthesis have been reviewed by Sun et al. These techniques involve multiple complex processes, long batch times (up to 24 h), and expensive post treatment to obtain the required crystal structure with specific particle sizes, making the synthesis process challenging especially toward industrial-scaled production (Table 1). While Li-ion batteries are attractive and promising both in terms of storage capacity and performance, the large scale production of the particles is quite expensive. A new innovative production, process optimization, and automation engineering of the large-scale battery production are important for economic viability. The current industrial Li-ion battery production route carries three major steps including (1) synthesis, (2) slurry-based electrode preparation, (3) cell assembly, and (4) formation and aging. The role-to-role layer transfer (described earlier) is one of the best techniques for layer fabrication. The traditional electrode manufacturing procedure is a very complex and labor-intensive process requiring a new fabrication procedure that fulfills the market demand. In this section, the state-of-the-art of Li₄Ti₅O₁₂ synthesis and its use in energy storage application are discussed. However, the gas phase Li₄Ti₅O₁₂ production is rather new. Ernst et al. synthesized LTO nanoparticles from a liquid precursor mixture consisting of lithium tert-butoxide dissolved in THF and titanium (IV) isopropoxide (TTIP) dissolved in xylene. The initial molar ratio of Li/Ti in the spray solution was investigated in the range of 0.5–1.0 at a total metal concentration of 0.8 and 1.6 M. The highest yield of the Li₄Ti₅O₁₂ phase (90 wt %) was reported for the spray solution with initial molar ratio of 1.0 and a total metal concentration of 0.8 M. Similarly, Bresser et al. synthesized and investigated flame sprayed LTO nanoparticles. Along with spinel Li₄Ti₅O₁₂, several impurity phases including rutile and anatase TiO₂, as well as electrochemically inactive lithium titanate phase (Li₁.14Ti₁.8O₄) were obtained. The charging and discharging μ- and nanosized LTO showed specific capacities of 1.5 and 70 mAh g⁻¹, respectively, revealing that size reduction has a significant effect on rate capability of LTO materials. In another report, Kim et al. prepared Li₄Ti₅O₁₂ materials using lithium nitrate (LNT) and TTIP in an ethyl alcohol/water mixture. Initial Li/Ti molar ratios at 4/5 and 4.6/5 (stoichiometric and nonstoichiometric conditions) in the precursor were studied to synthesize Li₄Ti₅O₁₂. The ultrasonic spray generated droplets were carried to a diffusion flame by 10 L/min of O₂ flow. The obtained Li₄Ti₅O₁₂ had anatase and rutile TiO₂ impurity phases due to a very short residence time within the flame. The post treatment of the powders at the temperature range of 600–800 °C gave phase pure Li₄Ti₅O₁₂. The particle size ranges (23 nm) were much lower at a lower temperature (600 °C) compared to the size of 300 nm at 800 °C. It is clear that the precursor–solvent combinations are crucial to avoid expensive post treatment of the powders. In addition, the extensive characterization and influence of impurity phases needs to be acquired to describe electrochemical performance of the sole Li₄Ti₅O₁₂ phase from the mixtures of the multiple phases. As described by Ernst et al., the spray solutions with a molar ratio of 0.5 < Li/Ti < 1 produced two spinel phases, namely, Li₄Ti₅O₁₂ and Li₁.14Ti₁.8O₄ along with anatase and rutile TiO₂. The highest mass % of pure Li₄Ti₅O₁₂ was 87% at a Li/Ti ratio of 1.74. While high energy storage performance is mostly realized for the phase pure material, the redox performance of these mixtures was much below the theoretical specific charge (170 mAh/g). To successfully obtain phase pure Li₄Ti₅O₁₂, Meierhofer et al. used five different solvents, three different lithium precursors, and a single titanium isopropoxide precursor (in total 25 solutions were ideal for the flame spray) to synthesize phase pure Li₄Ti₅O₁₂. Out of 25 flame sprayed powders, 13 precursor–solvent combinations gave Li₄Ti₅O₁₂ with a purity of ≥95 mass%. The use of 2-ethylhexanoic acid as a solvent for these 13 combinations producing high mass% of LTO purity was due to a strong M–COO⁻ chelating property in the solution prior to flame spray. Moreover, it was discovered that the presence of carboxylic acid transforms inexpensive low-volatile lithium nitrate into a more volatile organometallic lithium complex, which enabled gas-to-particle formation during flame synthesis and turned out to be an important criterion to reach high-quality LTO nanoparticles. The electrochemical performance to evaluate the energy-storage potential for three Li₄Ti₅O₁₂ obtained from different precursor solvent combinations showed almost the same discharge/charge reaction kinetics during cycling and rate tests. The doctor-bladed half-cells were stable for 450 full cycles at 1 C with a discharge capacity of 146.5 mAh/g and 170 mAh/g at C/5 (Figure 7A–D). The chemistry of nitrate carboxylation (see IR investigation of the spray solutions) observed during the screening process and prior to flame spray is vital for strategic designing of the other energy materials. The traditional doctor-blading technique requires carbon additives and organic binder to create a paste with the active material for the layer fabrication. This is an intensive procedure that requires time, energy, and high processing cost.

Gockel et al. used cost efficient in situ C-mixing via thermophoretic double flame spray pyrolysis followed by pressure-based role-to-role lamination technique for layer transfer. During flame spray pyrolysis, one independent flame was used for combusting xylene (C-source) and the second one was for combusting precursor–solvent (Li₄Ti₅O₁₂ source) placed at 20° with the horizontal (Figure 7A). The mixing of two aerosol streams produced homogeneous C/Li₄Ti₅O₁₂ mixtures. The particles were eventually transferred to the Cu-electrode via a pressurized lamination technique avoiding any organic/inorganic impurities that would otherwise affect the electrochemical performance (Figure 7A). The electrochemical performance of the laminated electrodes showed significantly lowered cell polarization for electrodes prepared at higher laminating pressures. All the laminated electrodes were able to withstand cycling for 200 cycles at 1 C. The performance of the compacted laminated electrodes compared to the doctor-bladed ones exhibited higher capacity.
retention upon long-term and high rate testing\textsuperscript{79} (Figure 7A). An innovative electrode fabrication procedure is a key to most of the large-scale battery production technology which requires flexible electrodes to implement them in electronic devices. To enable portability and easy use, Li-ion batteries development involves light, thin, flexible, and miniaturization against current thick, rigid, and bulky batteries. The strong consumer market demands innovative battery and flexible devices including roll-up displays, touch screens, and active radio frequency identification tags (RFID).\textsuperscript{80} Hence, fundamental investigation of the electrode flexibility is significant. Recently, Gockeln et al. used a fast, dry, and scalable flame aerosol technique capable of depositing pure and 6 nm crystalline Li4Ti5O12 nanoparticles in thin (0.55 μm) and smooth layers, directly onto flexible polyimide substrates (Figure 7D). The flexible electrodes assembled into all-solid-state Li-ion battery cells exhibited excellent stable dis-/charging for 30 cycles at 2 μA (1 C) without capacity loss.\textsuperscript{78} High stability was observed even after cycling at higher rates (≤10 μA, ≤5 C) (Figure 7E,F). Investigation on the static cell bending showed a capacity drop due to the loss of contacts via mechanical stress. In postbending condition, cycling was found to remain stable and capacity drop recovered\textsuperscript{78} (Figure 7F).

The perovskites are immerging materials for energy storage application. Recently, the gas phase synthesized BaTi\textsubscript{1−x}Zr\textsubscript{x}O\textsubscript{3} (x = 0−0.2) perovskites (∼10 nm) were able to shift the Curie temperature toward a lower temperature range with increasing Zr thereby decreasing the dielectric permittivity. The observed frequency dispersion of the permittivity is explained by Maxwell–Wagner polarization for ceramic particles at the
In a recently published review article, the progress in various functional materials including perovskites synthesized via the gas phase is discussed for their potential applications in energy storage and conversion.\(^6\)

### PARTICLES FOR CATALYTIC APPLICATION

The catalysts containing two or more metal oxide components as support, promoter, or catalytically active species play an important role in heterogeneous catalysis. The synthetic routes for designing such catalysts include precipitation or coprecipitation for a base material and the subsequent impregnation of catalyst promoter. However, recently, flame spray pyrolysis has emerged as a potential technique for the synthesis of such doped and/or mixed multicomponent materials for catalytic application (see Table 2 for details). Gas phase synthesis especially the flame spray pyrolysis avoids complex chemical reactions and introduction of catalytic promoters such as Ru, Pt, Pd, or Au in liquid feed prior to flame spray.

The in situ functionalization of the promoters in multicomponent catalytic systems is possible using double flame configuration (mixing of two aerosol streams during synthesis, Table 1). Schulz et al. described rapid quenching of the particles during FSP synthesis was a key for independent control of support (TiO\(_2\)) and noble metal (Pt) dispersion on the surface. The cooling of the flame temperature was able to reduce the Pt diameter significantly.\(^5\) Teoh et al. synthesized complex mixed metal oxides based on Ru-doped cobalt-zirconia (20 wt % Co) for catalytic application. The trace amounts of Ru (0.04 and 0.4 wt %) in Co\(_2\)O\(_4\) were able to lower significantly the two-step reduction temperature of Co\(_2\)O\(_4\) to metallic Co resulting in 4-fold CO chemisorption. The Fischer–Tropsch (FT) synthesis for CO conversion improved proportionally with the increase in metallic Co active sites.\(^6\) Minnermann et al. showed the promoting effect of Pd on the structural properties, and the catalytic behavior was determined mainly focusing on the understanding of structure–composition–performance relationships. The FT catalysis of the FSP synthesized Pd doped Fe\(_2\)O\(_4\) nanoparticles showed 1–2 nm Pd clusters evolve initially from homogeneously distributed Pd, and the iron oxide transforms into iron carbides depending on the Pd content. While the activity was enhanced with increasing Pd content, the selectivity was shifted toward long-chain hydrocarbons, mainly paraffins.\(^8\) While multiple flame spray allows for specific control of the homogeneous mixture of the multicomponent systems, double flame configuration was used to prepare Co/Al\(_2\)O\(_3\) based Fischer–Tropsch catalysts. The catalysts were stable on the Al\(_2\)O\(_3\) support during the FT reaction with enhanced performance unlike Pd/Fe\(_2\)O\(_4\) catalysts obtained using single flame setting.\(^5\) Piacentini et al. synthesized a library of Pt–Ba/Al\(_2\)O\(_3\) multicomponent catalysts using a similar multiflame reactor with various Ba loadings (4.5–33 wt %) and tested for NOx storage capacity.\(^6,8\) The absence of the HT-BaCO\(_3\) phase led to improved NOx storage potential at higher Ba loadings. Høj et al. also used a double flame system to prepare catalytic material (CoMo) in Al\(_2\)O\(_3\) support. The sulfidation and hydro-denitrogenation activity of the catalysts without heat treatments showed improved performance from 71% (single flame) to 91% (double flame) similar to the commercial sample, suggesting better promotion of the active molybdenum sulfide with reduced spinel phase (CoAl\(_2\)O\(_4\)) formation.\(^8\) For the stable and reasonably performing catalysts, enriched surface Al\(^{IV}\) species (in the case of Al\(_2\)O\(_3\) as a support) dispersed on the surface with metal promoters including Ru, Pt, Pd, and/or Au atomically dispersed on Al\(^{IV}\) surface allowing catalytic reactions. Considering Pd/SiO\(_2–\)Al\(_2\)O\(_3\) catalysts, Al\(_2\)O\(_3\) support provides an increased Al\(^{IV}\) number density (especially for the catalysts obtained in the gas phase) as active metal sites based on surface acidity that can be tuned from weak to very strong sites. To verify this effect, Wang et al. used double flame-generated Pd/SiO\(_2–\)Al\(_2\)O\(_3\) catalysts for chemoselective hydrogenation of aromatic ketones (Figure 8A–D).
The Pd particles with identical electronic properties on the Pd surface showed similar chemoselectivity for the hydrogeneration of carbonyl groups on all double-flame-derived catalysts. The hydrogeneration of the aromatic ketones was strongly enhanced by tuning the density of the surface Bronsted acid sites on the catalytic support by varying Si/Al ratios\(^9\) (Figure 8E–G). In another similar work, Wang et al. showed a synergistic effect due to two neighboring Al centers interacting with the silanol group in flame-synthesized Si/Al particles with high Al content. The two neighboring Al centers cause a decrease in the electron density of the silanol oxygen enriching the surface acidity responsible for C–H bond activation of benzene and glucose dehydration to 5-hydroxymethylfurfural.\(^89\) The double flame setup was also used to control the deposition of the catalytically active material (Co) on the Al\(_2\)O\(_3\)/La\(_2\)O\(_3\) support to obtain a defined particle size and tuned surface Bronsted acid sites density using different intersection distances of the particle streams from the independent nozzles for high catalytic activity.\(^90,91\) Apart from the mixed oxides, flame-made ternary oxides particles such as spinel (general form, AB\(_2\)O\(_4\)) are known to be catalytically active. The Pd promoted MgAl\(_2\)O\(_4\) spinel showed outstanding thermal stability compared to Pd promoted TiO\(_2\), CeO\(_2\), or ZrO\(_2\). The catalytic performance for methane oxidation (methane conversion) was less than 100% compared to Co/Al\(_2\)O\(_3\)/La\(_2\)O\(_3\) multicomponent catalysts.\(^90,92\) As a sequel to the work, Vegten et al. reported transition metal doped MgAl\(_2\)O\(_4\) spinel [Mg\(_{1-x}\)Al\(_x\)M\(_{1-x}\)O\(_4\) (x = 0.1, 0.5, 1 and 2, M = Mn, Fe, or Co) for methane combustion. The results showed that the performance increased in the order Fe < Co < Mn. The aluminum free spinel showed reasonable initial activity, and due to low thermal stability the catalytic activity was significantly reduced. The report also shows that the noble metals free (promotion free) catalysts could be industrially important as they significantly reduce the cost of production.\(^93\)

In another report, Mutz et al. investigated methanation redox catalysis using bimetallic Ni\(_x\)Fe/Al\(_2\)O\(_3\) and single metal Ni/Al\(_2\)O\(_3\) catalysts obtained via a homogeneous deposition–precipitation reaction. The redox catalysis in the industrial scale setting showed a Ni\(_x\)Fe/Al\(_2\)O\(_3\) catalyst revealed 71% CO\(_2\) conversion with 98% selectivity toward CH\(_4\). In addition, 17 wt% Ni\(_x\)Fe/Al\(_2\)O\(_3\) catalysts were stable above 350 °C compared to the commercially available catalysts.\(^94\) In the similar methanation reaction (under stationary and dynamic reaction conditions) using a commercial Ni/CaO–Al\(_2\)O\(_3\) catalyst, Mutz et al. reported 81% CO\(_2\) conversion and selectivity of 99% toward CH\(_4\) with a turnover frequency of 19.6 × 10\(^{-3}\) s\(^{-1}\) at 250 °C. The Operando XAS characterization of the catalysts showed rapid bulk oxidation of the Ni after the removal of H\(_2\) from the catalytic gas stream.\(^95\)

The other catalytic materials obtained from gas phase synthesis are perovskites. The photocatalytically active perovskite-slab-type La\(_2\)Ti\(_2\)O\(_7\) (∼26 nm) (using methyl orange decolorization via UV irradiation) showed a ∼30% improvement in the activity. The synthetic technique involving flame spray pyrolysis offers high yield as well as superior catalytic activity.\(^96\) Recently, perovskites (6–7 nm in size) based on LaCo\(_{1−x}\)Fe\(_x\)O\(_3\) were successfully reported using different Fe (0–60 wt %) substitution with Co against total metal concentration. The data showed that higher Fe substitution had a lower catalytic activity (ethanol oxidation), while lower Fe substitution offered higher electrocatalytic performance.\(^97\) In another report, cinnamyl alcohol oxidation kinetics with tertiary butyl hydroperoxide (TBHP) over the same LaCo\(_{1−x}\)Fe\(_x\)O\(_3\) perovskites showed beneficial effects of trivalent Fe in the perovskites. The reaction between liquid-organic moieties was catalyzed heterogeneously by the perovskite produced reactive oxygen species for surface kinetic reaction.\(^98\)

### THE CHALLENGES AND PROSPECT OF FSP

The gas phase nanoparticle synthesis for their application, especially flame spray pyrolysis, is a fascinating production route for many oxide materials due to the possibility or availability of (1) a wide range and variety of precursor solvent combinations, (2) particle designing (multicomponent mixing and/or doping beyond the solubility limits) using versatile flame configurations (symmetric and asymmetric, see Table 2 for details), (3) flame combustion at desired precursor and/or dispersant gas flow conditions, and (4) surface modified particles in the gas phase. Owing to these attractive properties and possibilities, the technique has produced more than 500 different materials, and it is widely used in academia (more than 30 groups working with FSP), adapted by industry (officially Johnson Matthey has announced the use of an FSP facility),\(^99\) and the largest public reactor installed in Spain (http://www.advance-fsp.eu/) producing several kilograms/day. While the synthesis of a highly performing material is possible and that the gas phase reactor allows implementation of the different reaction conditions, the successful synthesis of such materials lies in the judicial selection of expensive precursor–solvent combinations with an inherent high enthalpy density. The preparation of such a solution prior to flame spray is also challenging due to the absence of hydrolytic reaction with metal alkoxides and water molecules either present in the solution or from the atmosphere. The future prospect lies in designing such high enthalpy density spray combinations using economic metal salts (nitrates, chlorides, acetates) that react with the solvent producing in situ metal alkoxides and water molecules either present in the solution or from the atmosphere. The future performances in the gas phase (in situ) may have a significant impact on the crystallinity of the materials.

### CONCLUSION

The versatility of the flame aerosol technique shows the production of functional materials applicable but not limited to gas sensors, energy storage, and homo-/heterogeneous catalysts. The gas phase reaction at a high temperature environment, high thermal gradient, and multicomponent mixing at nanoscale with unique physicochemical properties makes FSP a key technique for the researchers working in the field of materials science. Apart from the gas phase particle production, facile in situ thermophoretic deposition of the particles in different flexible and/or nonflexible substrates
makes this technique even more attractive. In addition, the possibility of efficient layer transfer of in situ deposited particles using a role-to-role pressure based lamination process (flame spray pyrolysis combined with lamination) might even see large-scale industrial applicability in the field. It is clear that the potential of flame synthesis is far from complete exploitation compared to the wet-chemistry-based synthesis and layer transfer technology (spin-coating, dip coating doctor blading, etc.). Nevertheless, the literature has demonstrated that the flame synthesis and layer fabrication techniques offer opportunities that are unique on their own, which are not realized in the other traditional methods. Therefore, it is expected that the use of flame methods for the production of materials and layer fabrication procedure will contribute significantly to the state-of-the-art and beyond with wide scientific acceptance.

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Notes

The authors declare no competing financial interest.

Acknowledgments

Authors would like to thank Deutsche Forschungsgemeinschaft (DFG) for funding this work within the priority program SPP 1980 SPRAYSYN under the Grant MA 3333/14-1 and the Gottfried Wilhelm Leibniz Prize. Authors would also like to thank European Research Council (ERC) under Grant Agreement ReSuNiCo 786487. The authors declare no competing financial interest.

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