Final publishable summary report

1.1.1 Executive summary

Although nanotechnology, and particularly the development of nanoparticles-based materials, has advanced rapidly in recent years, industrial production techniques have not kept pace. At this point there is a substantial ***need for safe production facilities, enabling the synthesis of large amounts of nanoparticles*** with controlled and uniform quality (particle size, particle size distribution, chemical composition, etc.). This project responded to this need by developing an industrial plasma production line including ***on-line monitoring*** systems, assuring at the same time a ***high quality*** of the synthesized product as well as ***safety*** for the operating personnel and surrounding environment.

The **process development** and optimization has first been performed at a **lab-scale**. These developments have been supported by both modelling and monitoring that allowed an efficient industrial scaling-up. In parallel to these technological and scientific tasks, HSE issues have been particularly investigated for guarantying safe conditions for the workers along all the processing and manipulating steps. '**Health, Safety and Environment**' dust measurements have been carried out, showing that emissions of nano- and micron-sized silicon into the workspace are very low and controlled. Good practical guidelines for the manipulation of nanoparticles have been written and are available on the project website. A training on HSE has been organized within the consortium. A Life Cycle Assessment has been carried out for the ICP synthesis of Si nanopowder.

Based on the know-how and research performed at lab-scale, a **scaling-up** has been done in order to improve the yield and optimise the powder quality. One of the items that have been investigated is the industrial precursor feeding allowing a high and stable throughput of precursor. Among the monitoring techniques **multi-wavelength laser extinction spectroscopy** has been used to analyse in-line the quality of the produced powder.

Finally the elaborated nanostructured powders have been evaluated for battery applications.

Dissemination was an important part of the project. A webpage has been created with a public and a private access (www.simba-project.eu). A list of the papers published out of Simba project results and conference attendances is given on the project website.

1.1.2 Summary description of project context and objectives

Although nanotechnology, and particularly the development of nanoparticles-based materials, has advanced rapidly in recent years, industrial production techniques have not kept pace. At this point there is a substantial need for safe production facilities, enabling the synthesis of large amounts of metallic nanoparticles with controlled and uniform quality (particle size, particle size distribution, chemical composition, etc.). This project responded to this need by developing an industrial production line including on-line monitoring systems, assuring at the same time a high quality of the synthesized product as well as safety for the operating personnel and surrounding environment. The nano-structured materials of interest for this project are silicon and silicon-based alloyed nanoparticles, which have a huge potential as anode material in battery applications.

The production technology proposed is the Inductively Coupled Plasma (ICP) technique, which generates a high temperature thermal plasma discharge at atmospheric pressure. Since most of the ICP processes currently available are batch processes, the core of this project is the transfer of the ICP towards an industrial scale permitting the continuous production of a wide range of semiconductor or metallic (alloyed) nanoparticles. In order to achieve this, major scientific breakthroughs are required such as the incorporation of a novel on-line functionalisation technique, the design of an industrial powder injection system to ensure a continuous production, on-line monitoring techniques to ensure quality and safety and advanced modelling of the particle trajectories in the ICP plasma. A schematic representation of the scope of this project is illustrated in Figure 1. The resulting industrial production line will provide maximum production efficiency by virtue of fully automated and controlled feeding of raw materials and optimal reactor processing including evacuation of the processed powders. An additional advantage of this industrial line will be the recovery and recycling of excess precursor components and gases.

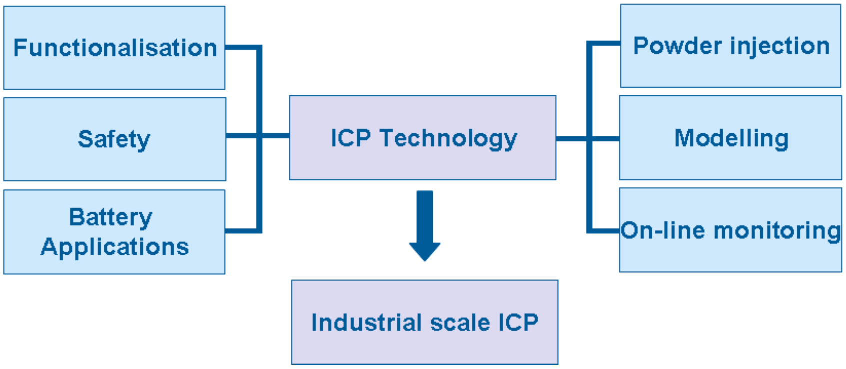

***Figure 1**. Schematic presentation of the scientific project breakthroughs*

The **overall objective** of this project is to **transfer the ICP processing knowledge and technology investigated at a lab-scale (10-100 g/h) to an industrial** scale apparatus for the **continuous production of tailored oxygen-free Si-based nanopowders at a production rate between *1 and 10 kg/hour*.**

The scientific objective with regard to the ICP processing is to synthesize nano-silicon based particles with an average particle size between 20 and 30 nm at a production rate of

1 to 10 kg/hour and a processing yield of at least 80%. The operational cost target at Umicore is 30€/kg.

The technical objective in terms of electrochemical performance for the developed Si based powders is to reach 1500 mAh/g at the nano-alloy scale and an energy increase at cell level of 25% and 40% for respectively portable and industrial applications contributed with long cycle and storage life (> 300 cycles for portable application and > 1000 cycles for industrial application). The different quantitative project objectives are summarised underneath:

Production of Si-based nanopowder
Powder characteristic: Nano Si ~20-30 nm
Production rate: 1-10 kg/h
Yield: 80%
Cost: 30-50 €/kg

Application to the production of new high performance anode material
Electrochemical capacity: 1500 mAh/g (+300% vs. graphite)
Cycle life: >300 cycles for portable application and >1000 cycles for industrial application

Different sub-objectives have been defined in each workpackage in order to meet the main objective. The interrelation between the different workpackage objectives is schematically illustrated in Figure 2.

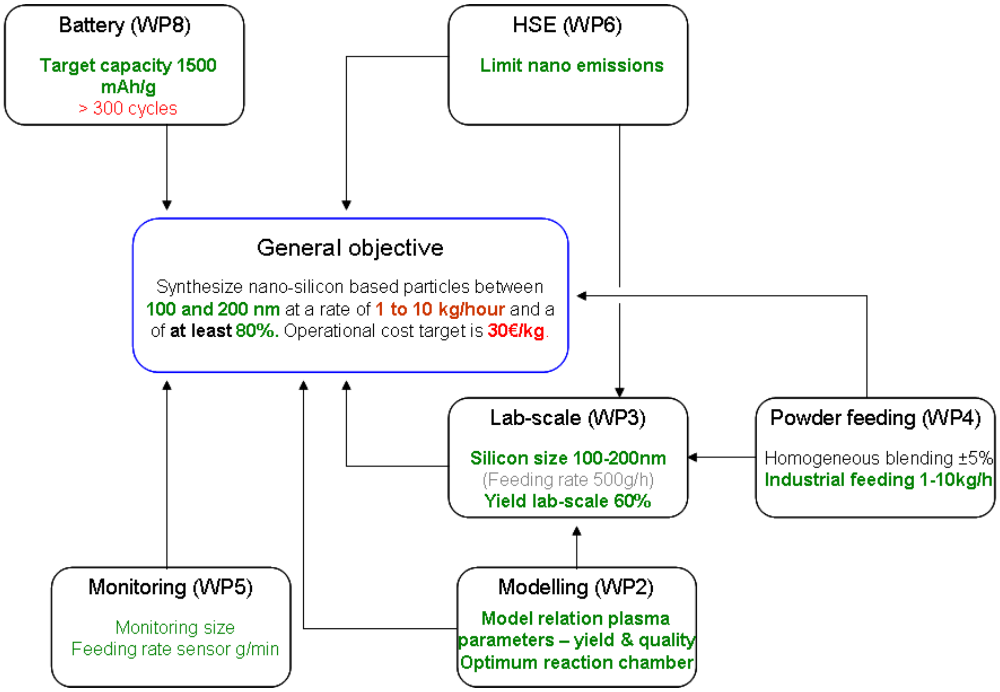

Figure 2. *Workpackage objectives in relation to each other and to the main objective*

1.1.3 Description of the main S&T results/foregrounds

During the three year project, most of the S&T objectives of the SIMBA project have been reached. The main results are then described in the following paragraphs.

WP2 – Modelling

In WP2, computational modelling modules for the Si nanoparticle synthesis process in ICP systems have been developed and exploited for the simulation of lab-scale plasma facilities available in the consortium and for the design of new optimized reaction chambers for increasing the process yield. Based on these modules and their validation, the improvements obtained at lab-scale could be transferred to the industrial scale process and a new design for the industrial reactor could be proposed. These modules are summarized below.

- Module for the **calculation of plasma properties** for the gas mixtures used in Si nanoparticle synthesis: computations have been carried out for different mixtures of gases and silicon vapour as a function of electron and gas temperatures as well as operating pressure.
- Module for the **simulation of plasma thermo-fluid-dynamics coupled with electromagnetic fields**: an equilibrium model has been developed taking into account the full 3D geometry of the torch and reaction chamber. Beside the equilibrium model that assume same temperature for the electrons and the gas phase, a **thermal non-equilibrium model** has been developed to estimate the electron and gas temperatures separately (*Deliverable 2.1 and Milestone 2*). Using this model, non-equilibrium regions have been obtained in case of strong quenching conditions.
- Module for **precursor tracking and interaction with plasma** thermo-fluid-dynamics: this model has been used to evaluate the evaporation efficiency of different commercial precursors and to optimize the lab-scale and industrial-scale operating conditions and geometries of the system.
- Module for **nanopowder nucleation, transport and growth**: a model based on the “method of moments” has been used to optimize and design the geometry of the reaction chamber of the ICP system with the final aim of increasing the process yield. Simulations have been done to optimize both the lab-scale reactor and the industrial scale process. A more accurate model based on “nodal method” has been used to characterize the lab-scale nanoparticle synthesis process (*Deliverable 2.2*).

These **models have been validated** by comparison with experimental measurements performed in WP3 of plasma properties (mainly the enthalpy) and nanoparticle properties (specific surface area and particle size distribution).

The developed models have been successively used for **designing a prototype optimized reaction chamber** for lab-scale processing based on the concept of “conical shape with injection of shroud gas” as well as for **designing a reaction chamber suitable for high production rates of nanopowders** at industrial scale based on the concept of the “porous wall” (*Deliverable 2.3 and Milestone 5*).

The prototype lab-scale reaction chamber has been designed with the support of plasma-thermo-fluid-dynamics models in order to guarantee a uniform gas injection and a sufficient cooling of chamber walls.
The **prototype has been manufactured and installed at UNIBO laboratories**: tests have been performed to check the decrease in powder deposition on chamber walls using shroud gas to protect the chamber walls (*Deliverable 2.4 and* Milestone 11).

WP3 – Lab-scale ICP processing

The first task (Task 3.1) was to identify and select potential precursors for Si, the alloying materials, the protective coating and the functionalization using a binder. European providers were then preferred when it was possible. Si could be synthesized in the lab-scale plasma facility at Empa leading to the achievement of *Milestone 1*. Typically, the average particle size of the crystalline Si nanoparticles can be tailor between 30 and 70 nm (BET values 25-70 m^2/g). An amorphous oxygen-rich layer of 1-2 nm is surrounding the nanoparticles while the total oxygen content is between 2.5 and 10 wt.%.

The second task (Task 3.2) was the investigation of in-flight alloying of Si nanoparticles as the alloys should present a better stability regarding their electrochemical performances in battery application. Mainly the **in-flight alloying of Si with Sn** has been investigated. After a few preliminary syntheses, the consortium decided to stop the study of the Si-Cu system for safety issues. Indeed the in-flight carbon coating could lead to the formation of explosive copper acetylide and therefore the functionalization of this alloy would have been critical. The Si-Sn alloys were produced at lab-scale using both premixed powders and in-line blending (separate feeding). Premixed inlet composition of **Si_{32}-Sn_{68}** (wt%) resulted in a product composition of Si_{21}-Sn_{65}-O_{14} to Si_{18}-$Sn_{72/75}$-$O_{10/7}$ (wt%), meaning a weight ratio in the product ranging from 1:3 to 1:4 depending on the feedrate. In the case of in-flight blending, starting with weight % ratio from 1:1.2 to 1:3 resulted in a product composition from 1:3 to 1:5. Even if the weight ratio expected was around 1:2, the product exhibited **initial good results regarding electrochemical** performance while mixed with carbon. The **reproducibility of the process** has been verified and **cycling using the products are still on-going to conclude on the lifetime**.

Pure Si nanopowders have been tested using either its full capacity or only a part of the capacity (between 1200 and 1800 mAh/g Si). The performance of pure silicon **reaches the objective** in term of **initial capacity** > 1500 mAh/g but is **below the cycle life objective** using this electrode technology.

The capacities of **Si-Cu and Si-Sn alloys** are **lower than objectives** (between 444 and 1066 mAh/g) and lower than expected. Cycle life is between 100 and 240 cycles, except for one sample (600 cycles) which exhibits the lowest capacity.

For all prepared Si alloys, high initial irreversible capacity has been measured (between 56% and 70%) probably due to oxide formation. Since reversible and irreversible capacities and cycle life are not improved compared to pure nano silicon, pure nano silicon had been selected for electrochemical test in prototype *(Milestone 9)*.

The main task of the work package 3 was the optimization of the lab-scale process (Task 3.3) with particular interest in **increasing the maximum evaporation rate** as well as **improving the powder quality**. First the plasma properties have been measured using mainly the enthalpy probe technique as well as optical emission spectroscopy. These measurements were used to develop and **validate the CFD modules** investigated in WP2.

To increase the maximum evaporation rate, efforts were put into three strategies: (i) development of a **new precursor injection probe**, (ii) systematic study of the impact of the **process parameters** (plasma power, pressure, composition of the plasma) and (iii) **prolongation of the evaporation zone** designed after modelling. Close collaboration with the modelling developed in WP2 has allowed focusing mainly on optimal solutions. Additionally, a study of the **impact of the precursor feeding** on the maximum evaporation rate was performed at lab-scale (frequency, pulse duration).

To evaluate the **maximum evaporation rate** into the plasma, a technique was developed based on the detection of the non-evaporated micro-particles using Optical Emission Spectroscopy. This method allows detecting the threshold above which the precursor micron sized particles are not totally evaporated. **Different injection probes** designs have been evaluated. The homemade designed solution is the most efficient, under lab-scale standard conditions, with improvement of up to **50%** of the maximum evaporation rate *(Deliverable 3.1)*. However the new dispersion probe has proven a more efficient evaporation at industrial scale.

The systematic study of **the process parameters impact** on the maximum evaporation rate concerned the change of the background pressure in the reactor from 400 to 700 mbar and sheath gas hydrogen flow from 3 to 10 l/min. It showed that increasing the pressure to **700 mba**r while decreasing auxiliary gas flow to **4 l/min** allows nearly **doubling** of the maximum evaporation rate compared to previous lab-scale conditions. However, even under such conditions that were non stable and then non compatible with a long term production, the maximum evaporation rate was about 200-250 g/min. Those observations provided validation for modelling in WP2.

The **prolongation of the evaporation zone** was done using inserts with two geometries based on modelling in WP2. High speed imaging showed a plasma tail that was less fluctuating spatially but **no improvement** regarding the evaporation rate of precursors was observed.

The **precursor injection parameters**, meaning the feeding frequency and duration of pulse, mainly, were varied to observe their impact and revealed that **pulsed regime with lower frequency** may result in **an increase** of the maximum evaporation rate.

For improving the **powder quality,** meaning to better control the size distribution of the Si nanoparticles, the aim was to **prevent recirculation** of the processed nanoparticles into the plasma tail as much as possible and to **separate** in-line the large non-fully evaporated particles from the nanoparticles (impactor). Recirculation of the processed nanoparticles in the plasma tail is responsible for most of the decrease in quality of the products.

Unfortunately, the efforts put in to reduce turbulences in the lab-scale reactor did not fully success, making it clear that such drawbacks can only be solved by designing a new reactor as planned in the actual project (WP2). In order to reduce recirculation, and based on modelling, two solutions have been carried out, namely the above mentioned inserts and a modification of the quench system. The first design did not lead to any improvement. The optimized version presented some advantages regarding the stability of the plasma tail but had no impact on the nanopowder quality. Experiments performed with high speed imaging showed clearly that recirculation in the "hot" zone of the plasma still occurred in the presence of the insert. The optimised quenching provided an improvement regarding the stability of the plasma tail but none concerning the recirculation and the nanopowders quality.

Separation of the nanoparticles in-flight during production was optimized using a homemade impactor and tested under standard conditions of process. Even if the design could still be improved by modelling the impact stage, this solution proved to be efficient in getting rid of the micron – submicron particles.

Task 3.4 was dealing with the **increase of the production yield**. The above mentioned designs were also evaluated to increase of the production yield but lead to the conclusion that a completely new reactor had to be designed in order to improve the yield. The actual lab-scale reactor presents too many turbulences and recirculation problems.

The aim of task 3.5: In-situ in-flight functionalization was to fulfil two main functions fundamentals for battery application: applying a **coating on the Si nanoparticles** with a thin conductive **film of carbon**, then depositing in-flight a **binder** at a lower position of the reactor (cooler zone).

Carbon coating of the processed nanoparticles was investigated under the following conditions for gas phase CH_4 as carbon precursor. Optimisation of the quench and recirculation solution was systematically investigated for flows of CH_4 ranging from 0.1 to 3.5 l/min. Whatever the considered solution it appears that it was **impossible to get a C-coating of the Si nanoparticles in the lab-scale reactor probably because of recirculations**. The Si:C inlet ratio was shown to result in similar ratio on the collection filter. The powders collected showed a combination of Si and SiC nanoparticles. Additionally, a solution based on the use of polymers as **solid precursor for C** were investigated, but was abandoned due to clogging issues inside the feeding line.

In-flight deposition of different binders was investigated after selection. A dedicated device for binder dispersion in the reactor during process was developed based on a Palas AGK 2000 nebulizer equipped with a homemade injector *(Deliverable 3.4)*. Different binders were selected. All binders were diluted in a solvent (water or ethanol) to the highest concentration that allowed injection using the nebulizer (too high viscosity preventing dispersion). The nebulizer was connected at two different positions namely the bottom of the conical part of the reactor and the exhaust line (lower temperature). Whatever the position for injection, it was not possible to quantitatively detect the presence of a binder at the surface of the nanoparticles, even if obviously the behaviour and aspect of the powder was different.

Amongst the 3 types of tested functionalized powders, 2 types show too low reversible capacity (between 400 and 500 mAh/g) and too high irreversible capacity. In the case of carbon coatings, the low capacity (400 mAh/g) is explained by formation of silicon carbide compounds. The highest capacity (1000 mAh/g) has been measured using type#1 functionalization for which the cycles are still running (>40 cycles).

As a conclusion, it has been demonstrated experimentally that with the present power level and configuration of the lab-scale setup (optimized injection probe and process parameters, prolongation of the evaporation zone), the **maximum evaporation rate** achieved under stable conditions is around **150 g/h** and the process **yield** is less than **15%**. According to modelling results however, the yield should be drastically increased by the new reactor geometry (up to determined objective at 50%) but the low power available will not allow reaching the targeted 500 g/h evaporation rate.

WP4 – Industrial powder feeding

At the beginning of the project, both powder feeders from DACS available at Empa and Umicore have been updated. Additionally, t**wo feeding systems** allowing also **blending** of two different powders for in-flight alloying purposes were delivered at lab-scale and industrial scale facilities (*Deliverables 4.1 & 4.4*).

Feeding parameters were developed for testing different evaluated precursors. For long-term stability feeding, a **back-pulse** has been introduced to prevent clogging of the sucking line. The silo has been redesigned with an **over-pressure control valve for improving safety** and reducing contamination with air. The **long-term accuracy** of powder feeding was assessed at Umicore during a long test over one week and it lies within the **specified 5%.**

A **simple unit** has been delivered to IWS for the development of the monitoring sensor for which nanoparticles had to be transported (*Deliverable 4.3*). A high volume feeding system has been also developed and delivered to Umicore for **recycling** experiments.

A **new industrial concept** for powder feeding has been worked out (*Milestone 7*). The feeding rate control is then done using a flow rate sensor install on the vacuum line and no more only using the pressure in the cells. It is now more sensitive to membrane clogging effects or problem on the cells. Moreover, for a **longer maintenance interval**, larger filter is built on the vacuum line, and an additional filter has been implemented to protect the flow rate sensor. Higher flexibility is also achievable for the silo size. Silos from 2 to 20 liters can be adapted on the same design. The system is easy to clean and moveable. Indeed the control unit as well as the feeding module and silo are set on tracks that can be separated or moved together. Finally, the **feeding sensor** investigated in WP5 can be easily integrated in the system.

WP5 – Process monitoring

In WP5 two main objectives were defined: developing a **nanoparticle size** measuring system usable directly in the generation process and, a precursor **particle feeding rate sensor**.

In the beginning of the project, the presently available **nanoparticle characterization** technologies have been investigated and rated concerning in situ, gas borne nanoparticle (< 100 nm) and vacuum applicability (*Deliverable 5.1*). **Multi-wavelength light extinction spectroscopy** (LES) as only method with the capability to meet the requirements in general has been selected. The physical effect, which is taken advantage of, is light weakening **absorption** and **scattering** as a function of particle size. To extensively test the extinction spectroscopy setup, dedicated **test equipment** was designed and built up (*Deliverable 5.2*). It enabled to ad hoc generate gas borne nanoparticles by laser ablation and variable monitoring through a variety of viewports and interfaces. Additionally, it was necessary to design proper **viewport elements** to **ensure sedimentation-free, long-term stable** optical access.

After engineering the testing equipment and the viewports, a dedicated **nanoparticle sensor concept** has been developed (*Deliverable 5.3*). To meet all demands and with respect to the knowledge already gathered, the developed sensor concept consists of:

- long-term-stable **process integration** variant: process exhaust line between raw particle separation (cyclone) and nanoparticle filter,
- **variable** design of **light – particle interaction zone** to cover various, non-predictive particle concentrations,
- optical setup of **three geometrically combined laser beams** with wavelengths of **405, 532 and 635 nm** due to expected usability with the consortium's material systems,
- **integrated software** for system hardware control, extinction simulation (including open port for external simulation data import), **self-calibration**, measurement, data calculation/presentation and data export.

All knowledge, up-to-then measurement results and demands were integrated in the R&D subcontracting work, so the constructed **extinction sensor setup in final state** is in first evaluation state at the subcontractor's site.

With this first design of the light extinction nanoparticle sensor detecting nanoparticles in a laboratory setup**,** the milestone **"Concept of on-line monitoring of Si nanoparticles"** was even experimentally proven to be achieved **(***Milestone 6***).**

Regarding the second task, a **sensor for raw precursor particle feedrate** characterization was developed and initially evaluated (*Deliverable 5.4*). The working principle is the utilization of weakening of the transmitted electromagnetic intensity by particles in a material system dependent wavelength range, while the **light intensity attenuation is proportional to the amount of material**. For evaluation the sensor was integrated between powder feeder and exhaust in the powder line. Sensor raw signals acquisition (voltage, range: 0…10V) was performed by a storage oscilloscope.

High-speed **semi-industrial evaluation of the feeding rate sensor** (*Deliverable 5.5*) was performed with a KEITHLEY K-USB 3102 device (max. recording speed 100 kHz), which was adapted to the feed rate sensor. The feed rate is evaluated every 10 seconds by integration of the recorded data. The integrated signal (with 5 % error bars) against the feed rate could then be measured. In conclusion, the **prediction of the feed rate for the requested feeding range is possible**. A **lab-scale evaluation of the nanoparticle sensor** was successfully performed(*Deliverable 5.5*).

Scientific and technical culmination of the WP 5 was the **integration** of the nanoparticle sensor in an **industrial nanoparticle ICP generation process** (Deliverable 5.6).Integratability, functionality, reasonability of the results and the general sensor behaviour and long-term stability were convincingly demonstrated during the **on-site evaluation at the production-scale reactor**. An industrial measurement campaign has been carried out (*Deliverable 7.2*). Particle sizes of about **50…60 nm were reliably detected** and confirmed afterwards via TEM reference.

WP6 – Health, Safety & Environment HSE

The main aim of this work package was to **guarantee a safe manipulation** of the nanopowders during processing at lab and industrial scale as well as during characterization and battery material preparation. Therefore, the safety guidelines regarding nanopowders of each partner were first collected and analysed. A specific meeting has been organized to define **handling safety guidelines for the consortium** *(Deliverable 6.2)*. These guidelines taking into account the **packaging, reception and manipulation** of the produced Silicon based nanopowders have been validated. The guidelines have been published onto the website in the public access zone (www.simba-project.eu). In parallel a specific and internal **training on HSE** (*Deliverable 6.3*) has been organized for the project consortium during the first 6 month project meeting.

Particle emission measurement campaigns have been performed at different partners for assessing the emission potential during the whole plasma processing from precursor manipulation to packaging and cleaning steps (*Deliverable 6.1*). The measurements were performed using particle counters able to measure micro- and nano-scale particle concentrations in the environment. In general, the particle concentration measured around the plasma setups are significantly lower (by a factor 5 to 10) than outside. According to the results of this WP, the partners **adapted their laboratories and facilities**.

A **life cycle analysis (LCA)** has been also performed taking into account the processing of 1 kg Si nanopowders from the inlet materials to the packaging. Different models were compared and all showed that the process yield is the most important factor impacting on the environment. A comparison with graphite has been presented.

An evaluation has been done on the feasibility to recycle course 'waste' powder from the reactor and from the cyclone for precursor injection (*Deliverable 6.4*). It has been shown that this recycled powder allows easy and stable feeding behavior. Furthermore, no accretions are formed on the quench ring/torch. When an ICP experiment is carried out starting from the recycled material, a nanopowder is obtained in the filter with the same characteristics as the standard powder. It is clear that by the recycling of the coarse waste fraction, the overall environmental impact of the process is reduced.

WP7 – Industrial scaling-up of the ICP

An evaluation at pilot scale illustrated that the specific procedure for **precursor injection** (developed in WP4) allowed for the continuous synthesis of Si nanopowder. Stable operation was possible, with constant feeding rate and without clogging of the feeding tubes. On top of that a new **injection probe** was designed (*Deliverable 7.1*). By combining the optimal settings for precursor injection with the standard set of ICP parameters (defined in WP3), 'production-like' runs could be carried out where **larger quantities of Si powder** with a targeted BET value were obtained (*Deliverable 7.5*). The selected process settings resulted in a good stability, which enables the large quantity synthesis of Si powders with reproducible product properties. The resulting powders could be used to supply the partners for further evaluation (*Deliverable 7.3*).

The overall **process yield** was increased by the installation of a modified reactor concept (developed in WP2 using extensive modelling): an improvement in yield (in the collector) was observed with a factor of 5. First industrial tests confirmed high yields. Today the continuous production of 900g/h silicon with yield 70-80% and BET 20-40m^2/g has been achieved.

The **on-line monitoring sensor** developed by IWS was successfully evaluated on the industrial ICP installation during the synthesis of Si (*Deliverable 7.2*). Several measurements have been carried out at different injection rates. The intensities measured during the experiments showed quite stable values. A clear identification and differentiation could be made between powder feeding on, feed rate alteration and powder feeding off. Despite transmission alteration a stable particle size calculation could be made. The 3λ extinction sensor results are plausible and showed correlations with TEM results as explained in WP5. A consistent measurement has been obtained, however, so far only for a limited number of conditions.

A **new high throughput filter** has been installed on the large-scale ICP installation (*Deliverable 7.4*). This allows for the synthesis of Si nanopowder with high throughput at stable pressure when working at high feed rates. The influence of the back pulse is eliminated and moreover a safer cleaning manipulation is achieved (HSE improvement).

WP8 – Battery evaluation

Based on literature survey, **Cu and Sn alloying elements** were chosen since they were the most studied for Si composite or alloy. Based on metallurgical considerations, several compositions have been proposed for the synthesis of Si-M alloys by ICP.

For electrode technology and agglomeration, **different types of carbon and binders** have been selected.

Task 8.3 was dealing with the development of the electrode technology and electrochemical evaluation in **laboratory cell** *(Milestones 4 & 10 and Deliverables 8.1 - 8.3)*. First test using low carbon and binder content led to poor cycle life results. Therefore, to achieve the 300 cycle objective, an optimization of the electrode technology has been carried out to increase cycle life using high amount of additives. Many process parameters have been explored and finally an **improved electrode technology with optimized process has led to stable performance for more than 400 cycles**.

Meanwhile efforts have been allocated to reduce additive amount (to 20%) in the electrode composition. By modifying process and the nature of carbon, **high capacity (1200 mAh/g) and good cycle life has been achieved (>320 cycles without capacity loss)**.

In Task 8.4, **full cell evaluation** has been done in cylindrical cell using NCA lamellar oxide as positive electrode material. For this final evaluation, pure Si nanopowder produced on the pilot plant has been selected. Since the cycling results of low additive content electrode technology was not known at the time to launch the prototype cells, the 50% additive electrode technology solution was applied *(Milestone 16 and Deliverables 8.2 - 8.4*).

The comparison between the standard graphite /-/ cell and nano Si anode cell expressed per gram of positive material reveals a higher irreversible capacity of silicon anode compared to graphite which results in a loss of utilized capacity of the positive electrode. As a consequence, the cell capacity was equivalent between graphite and Si anodes even if it was possible to introduce longer electrodes in the cell. Nevertheless, **good performances** have been measured at high discharge rates (10C) as well as at low temperature (down to -10°C).

Finally, the cell capacity during cycling for Si nanopowder is not as stable as for graphite. The end of life criteria (20% capacity loss) is achieved after around 70 cycles. The lower cycle life observed in prototype cell compared to the measurement in half cell is due to the lithium consumption during electrolyte decomposition on the negative electrode. This phenomenon does not affect the performance in half cell but causes a capacity fading in full cell.

WP9 – Dissemination, Exploitation and Training

Dissemination and exploitation (WP9) was an important topic of the Simba project. At the beginning of the project, **a project website** (*Deliverable 9.1*) has been created with a private and a public access (**www.simba-project.eu**). Some relevant information like for instance the guideline for a safe manipulation of nanopowders as well as some presentations are available on this website.

At most of the project meetings every 6 months, a **specific training** has been organized. In total 7 meetings took place and, each partner organized at least one. Trainings were performed on ICP processing, safety, gas phase monitoring and characterization of nanopowders, batteries and modelling.

Beside these mandatory project meetings, 2 phone conferences were organized with all the partners at month 3 and month 15. After each meeting, a report was written (*Deliverable 1.1*).

The scientific results obtained mainly in WP2 and through the fruitful collaboration between the modelling and experimental activities led to the publication of **10 papers** in international recognized papers, and to the participation to **18 conferences**. Also the electrochemical performances could be presented by SAFT at **two more international conferences** (ECS 2011 and IMLB 2012).

1.1.4 Potential impact

The core objective of the SIMBA project was the transfer of the ICP technique from lab-scale towards an **industrial scale**, permitting the continuous production of a wide range of semiconductor or metallic (alloyed) nanoparticles. Therefore, major scientific breakthroughs were needed in the fields of **on-line functionalization**, **industrial powder injection**, **on-line monitoring** and advanced **modelling** of the particle trajectories. In order to link the scientific findings as much as possible to real industrial applications, all these activities were focused on the ICP processing of high added value **silicon-based nanomaterials** for battery applications. Potential **health, safety and environmental** issues of the developed technology were studied in detail as well.

With regard to **industrial plasma synthesis**, the new concept of porous wall has significantly increased production yield. We noted at least a factor of 4 increase in production of Si-nanoparticles. Umicore and Empa can and will use this concept in their research on similar reactor geometries. It will also be a major advantage for the industrial synthesis of nanomaterials in general.

The **modelling** codes and software developed by Unibo in SIMBA can serve as a basis for other projects and collaborations on plasma synthesis of other nanomaterials as well as on high temperature gas phase synthesis modelling. Current impact and dissemination activities are:

- submission FP7 project IMMENSE – “Insight via Multiscale Modelling Environment for Nanoparticle Synthesis and Engineering”; call: FP7-NMP-2013-1.4-1
- training students
- dissemination of SIMBA results on international scientific journals (J. Phys. D: Appl. Phys., Plasma Sources Sci. Technol., etc.)

The new **plasma** reactor concept designed for lab-scale experiments will allow the elaboration of much more controlled nanoparticles regarding their particle size distribution but also potentially their stoichiometry and composition. This could open new application field for plasma processed nanoparticles.

With regard to the **powder injection**, DACS has developed and further improved within SIMBA his industrial powder feeder with monitoring sensor. Thanks to the developments within SIMBA, the powder feeder design was essentially optimised and the product range was expanded by integrating an optical powder flow sensor. These optimisations resulted in several impacts and further dissemination activities:

- A first sale of a unit based on the newly developed industrial powder feeder concept. This system will be installed in December this year at a customer in France.
- As a further result of the approved increased reliability of the powder feeder DACS signed a new co-operation contract with a Swiss technology company

active in the pharmaceutical industry that wants to promote the new feeder technology in his market.
- A third co-operation with another Swiss company is concerning further research and development, where the new powder feeder generation is an essential part to realise new coatings for the automotive industry.

An **on-line monitoring** sensor concept has been developed by IWS that could potentially been used for other technologies, powders and applications. Current impact and dissemination activities are:

- improvement of the sensor for the use as situ characterization tool in carbon black generation processes
- cooperation with the Technical University Dresden within the framework of a student traineeship,
- in case of success, a cooperation with a particle characterization systems involved company to bring the system to market is planned.

In SIMBA, great importance was given to **health, safety and environmental** issues. A methodology for dust measurements, combining continuous and ad-hoc dust measurements has been developed for nanopowders. This methodology is based on using two different dust measuring techniques to determine the weight fraction of dust in combination with the volume of ultrafine nanoparticles. This concept is currently applied at Umicore and could eventually be implemented in other environments. The project partners have put the procedure on how to deal with and to handle nanopowders on the public part of the SIMBA-website to make it available to the broader nanomaterials community. On the basis of this procedure, an internal training has been organised within the SIMBA project and a presentation to the broader community has been given during the SIMBA workshop organised in June 2012 during the HTPP12 conference in Bologna. Recently, Umicore has also given training on how to deal with nanoparticles in industrial environment to external companies, mostly in bi-lateral meeting. A Life-Cycle-Analysis has been carried out for the ICP plasma synthesis of nano-Si for battery applications. Moreover, a benchmark was made with current anode materials. The knowledge obtained on the methodology as well as on the specific aspects for silicon based battery materials can be used in further projects to improve technologies and products.

The results of the SIMBA-project were published in a series of scientific publications in international journals (J. Phys. D : Appl. Phys., Plasma Sources Sci. Technol., IEEE Trans. Plasma. Sci.). Up to now these articles were cited at least 45 times. The SIMBA-researchers participated in several national and international conferences and presented 10 articles on the SIMBA-results for publication in peer-reviewed journals.

To disseminate the results of the project, the project team organised a workshop in Bologna in June 2012. This workshop, was linked to an international conference on high temperature plasma processing. This gave the SIMBA partners the opportunity to promote their results to the scientific plasma community as a whole. Several institutes, companies and projects have afterwards expressed their interest in the nano-silicon powders resulting from the project. Up to now contacts were established with another

FP7-project SOMABAT (University Kiev) and LABOHR (Tel-Aviv University). Silicon nanopowder has been supplied by Umicore to both projects. In the next months additional contacts are expected.

1.1.5 Public website

1.1.5.1 Project logo

Figure 3. *Project logo*

1.1.5.2 Project website

www.simba-project.eu