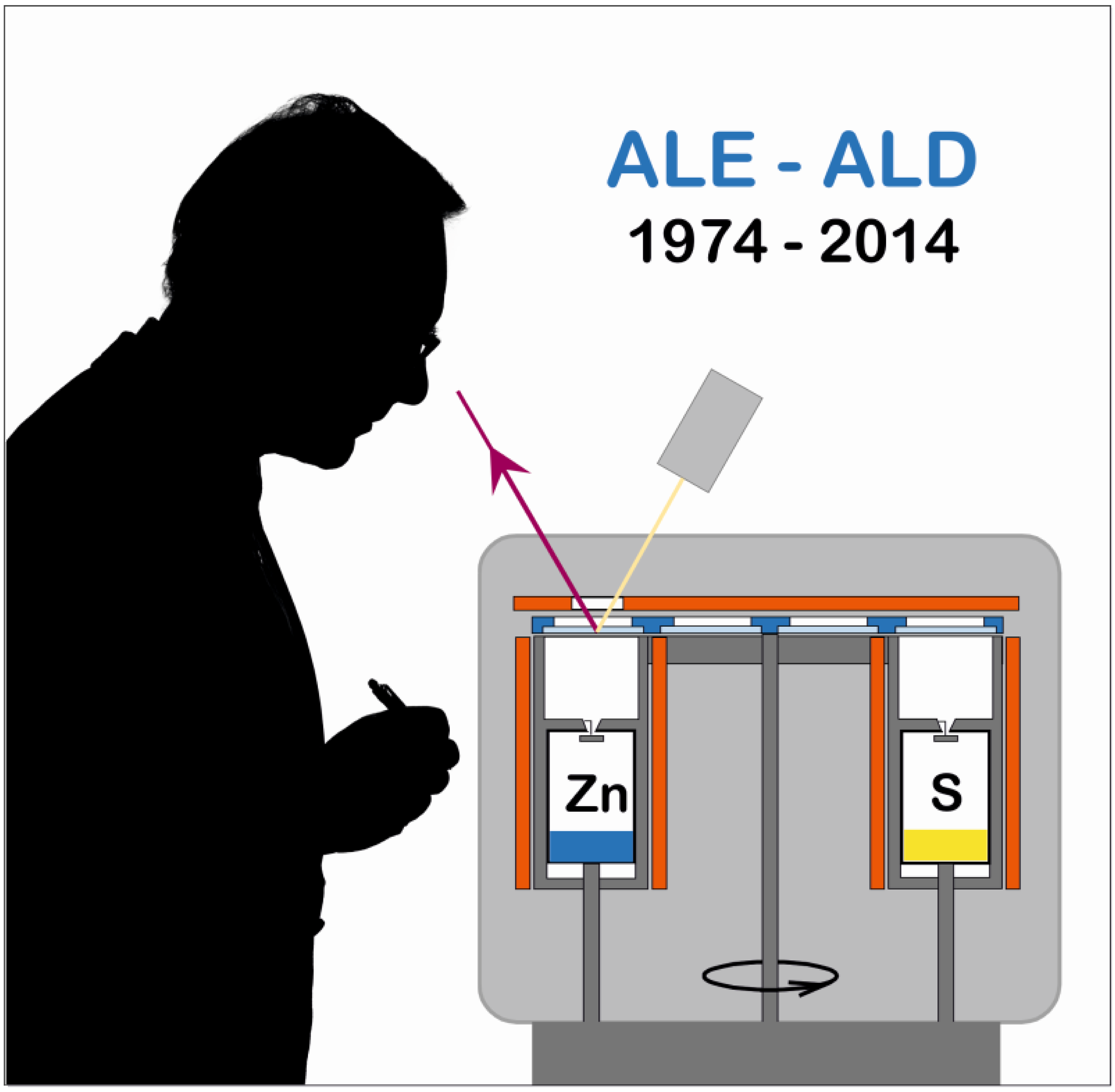
40 Years of ALD in Finland – Photos, Stories

“FinALD40”

Exhibition by the Finnish Centre of Excellence in Atomic Layer Deposition



PREFACE

To exhibition material released in the internet, Nov. 29, 2014

Atomic Layer Deposition (ALD) is not a new technique, since the first experiments in Finland were carried out over 40 years ago. Industrial use began in 1983 with the production of thin film electroluminescent (EL) displays. The ALD (ALE called that time) research in 1970s and in Finnish universities since 1981 was focused on materials needed in EL displays. In the EL displays, the dielectric-luminescent layer-dielectric thin films stack is made in a continuous process. ALD is commonly considered as a method suitable only for deposition of very thin films. Interestingly, in EL stack the total layer thickness is 1–1.5 micron. Other industrial applications of ALD remained limited for 20 years, but during the past 10–15 years microelectronics has been the major driver for ALD technology.

In the late 1990s, it became obvious that the continuation of Moore's law would require introduction of new materials into microelectronics. Additionally, new deposition methods were needed in IC technology since materials had to be deposited with atomic level accuracy as very thin films uniformly over the increasing wafer sizes and conformally over the increasingly demanding three-dimensional (3D) device structures. New interest was paid to ALD and enormous research activities went into the development of processes to manufacture high-k dielectric materials, metals, and materials for barrier layers. High-k dielectric materials for both gate oxides in metal–oxide–semiconductor field effect transistors (MOSFET) and capacitor dielectrics in Dynamic Random Access Memories (DRAM) have been the most important topics in ALD research during the last 10–15 years. Seven years ago, Intel announced that the first microprocessors using MOSFETs containing high-k dielectrics were coming to the market. These revolutionary new devices contained ALD-made hafnium-based oxides as dielectrics and metal gate electrodes. The third application area where ALD has been extensively studied in microelectronics is interconnects. Other areas in microelectronics for which ALD have been explored include non-volatile memories such as ferroelectric RAM, flash, and phase change memories.

During the years ALD has been studied in many other areas outside from microelectronics such as magnetic read-write heads, optics, protective coatings, and deposition of materials for different energy applications. ALD is also industrially applied in many of these areas. The recent developments in speeding the ALD process such as large batch reactors, spatial or roll-to-roll ALD have enabled utilization of ALD in deposition of protecting layers in LEDs and OLEDs and passivation layers on crystalline silicon solar cells.

ALD research has increased significantly within the years while in mid 80s annually less than 50 ALD (ALE) publications were published the number was more than 2500 in 2013. It should be noticed that almost 40 % from the publications are patents. The worldwide activity in ALD can also be seen in conferences. The annual AVS-ALD meetings collect 600-700 people and in addition there are several local ALD meetings and ALD sessions in many big conferences.

ALD has been and is very important for Finland. The technology development and first industrial applications were born here in 1970s. Especially the ALD tool manufacturing has been important in Finland and at the moment here are three companies in that business: ASM Microchemistry, Beneq and Picosun. The number of industrial users of ALD in Finland is limited, however, and at the moment the following companies are using ALD in their production: Inficon, Kalevala Koru, Lumineq, Vaisala.

Academic studies of ALD have long traditions in Finland. The first experiments were made in Tampere University of Technology and Helsinki University of Technology in early 80s. The number of scientific publications begun to increase after mid-80s when the first Academy of Finland and TEKES funded university projects were carried out. The ALD activities then expanded in 80s to University of Joensuu and VTT and in early 90s in University of Helsinki. Later expansion continued to Mikkeli (Lappeenranta University of Technology), and University of Jyväskylä. In scientific activities in both publications and conference participations Finland has been one of the top three countries together with USA and Korea.

ALD seems to be still a growing technology. Year by year the number of publications has increased as has done the number of participants in conferences. Only in number of patents small saturation can be seen. There are several reasons for the growth one important being that new countries such as China, Taiwan, Singapore, Turkey, and Mexico where volume and level of science is rapidly increasing have started ALD research. Industry is an important driving force. The family of reactor manufacturers is expanding and includes now more than 60 companies. The growth of ALD tool production is originating from the expanding user industry and application areas. It seems that there are no reasons why the growth of industrial use of ALD, ALD tool manufacturing industry and scientific research of ALD could not continue.

Celebrating the 40 years history of the Finnish ALD the Academy of Finland's Finnish Centre of Excellence on Atomic Layer Deposition, organized an exhibition: "40 Years of ALD in Finland: Photos, Stories". Initially, the exhibition was organized for the international Baltic ALD conference, May 12-13, 2014, Helsinki (http://www.aldcoe.fi/bald2014/). The main organizers of the exhibition have been Dr. Riikka Puurunen (VTT) and Dr. Jaakko Niinistö (University of Helsinki).
The exhibition material can be viewed at
- VTT, Micronova, Tietotie 3, Espoo (Mon-Fri 8:00 - 16:30), from May 12 until the end of 2014 and it was exposed at
- University of Helsinki, Chemicum, A.I. Virtasenaukio 1, Helsinki August 15 - October 15, 2014
- The exhibition has also visited ALD 2014 conference in Kyoto, Japan, in June (http://www.ald2014.org/).

The famous patent on Atomic Layer Epitaxy (FIN 52359) was filed on November 29, 1974 and this history material is now released exactly 40 years later.

Markku Leskelä
Professor of Inorganic Chemistry, University of Helsinki
Leader of the Finnish Centre of Excellence on Atomic Layer Deposition

Image, previous page:
Reconstruction of the first ALE experiment in 1974. Tuomo Suntola standing next to the ALE reactor, observing the appearance of purple interference colour in the growing film. The scheme has been modified from a figure in the story of Tuomo Suntola's ALE in Short. The silhouette photograph has been taken by Riikka Puurunen of Tuomo Suntola on August 23, 2014 - almost exactly 40 years after the first ALE experiment was made. (Image by Riikka Puurunen)

Tuomo Suntola's ALE in Short – Photos

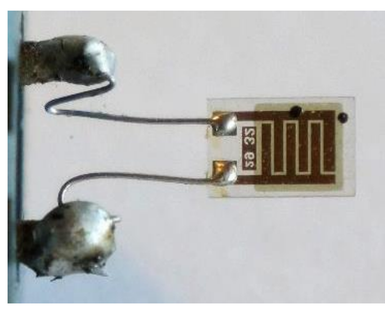
By Riikka Puurunen, VTT Technical Research Centre of Finland

These two posters contain an image summary of the full story

Prologue

Tuomo Suntola was born in 1943 in Tampere. He went to school in Turku and completed his Matriculation examinations (nation-wide examinations, which qualify to university studies) in 1962.

In 1963, Suntola started his studies on electrical engineering at Helsinki University of Technology (HUT - Teknillinen korkeakoulu, TKK). In 1967, he completed his Master's thesis ("diplomityö" in Finnish) on Schottky diodes for radar detectors (metal-semiconductor diodes for microwave frequencies) and in 1971 his doctoral thesis on picosecond "Ovonic" switches.



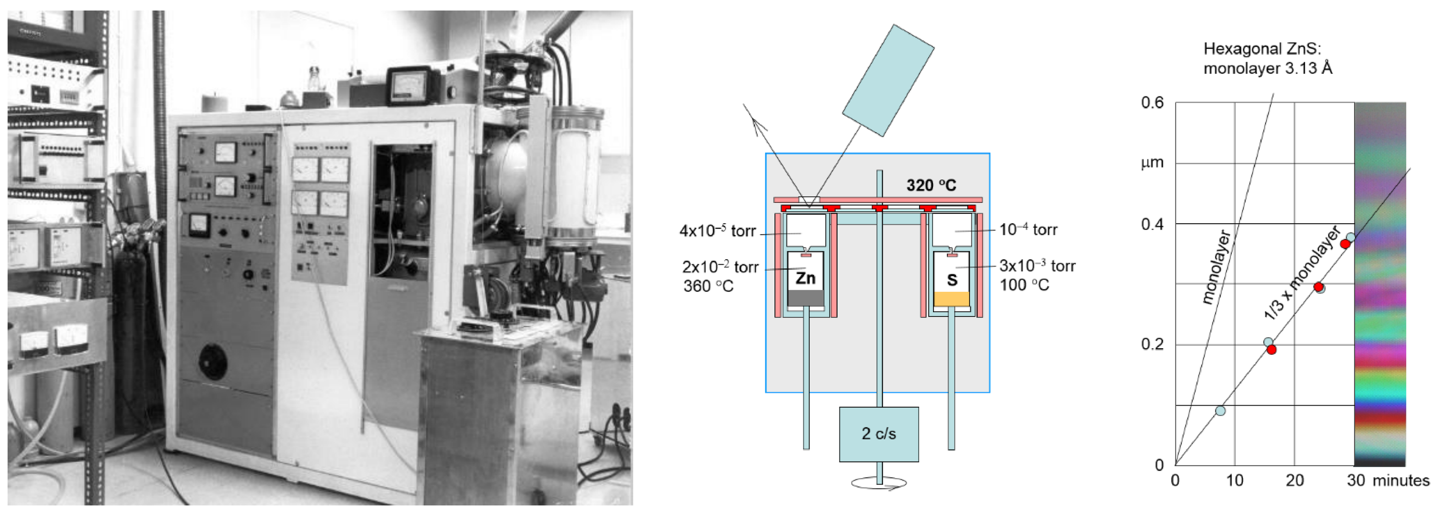
Picture: A prototype of the Humicap humidity sensor from 1973, designed by Suntola in the first industrial project of the VTT Semiconductor Laboratory. Vaisala's instruments using the Humicap sensor, based on the original patent 1972-11-12/1979-08-21, US 4,164,868 by Suntola, are still (2014) the world market leaders in humidity measurements The prototype sensor was fully functional after 41 years when photographed in 2014.

1974 - Initiation of the EL development and the invention of ALE

Late in 1973, Suntola was invited to establish a research group in Instrumentarium, with an open request to "suggest and find out something". After a market/technology mapping study, Suntola's proposal to Instrumentarium's management was to start working for two goals, for ion-selective sensors and for flat panel display. Suntola directed himself to solve the flat panel objectives.

The actual invention of ALE matured in early June 1974, Suntola describes it as follows:

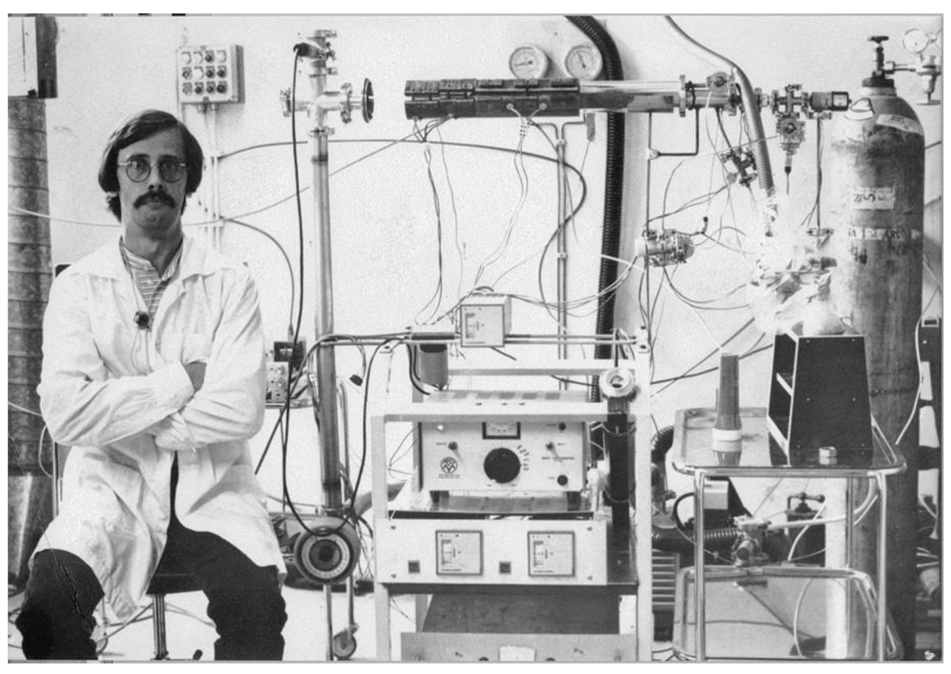
> *"We had still an empty laboratory with just tables and chairs and a Periodic Table of the Elements hanging on the wall. Looking at the Periodic Table, and thinking of the overall symmetry in nature, to me came the idea of "serving" the complementary elements of a compound sequentially, one at a time, onto a surface. Monoatomic layers may be obtained if the complementary elements make a stronger bond with each other than they do with their own kind of atoms".*



Pictures: The first ALE-reactor was built in the T-piece on right of the pumping unit. It consisted of a rotating carousel holding the substrates, and the sources for the reactants. Windows in the top flange allowed observation of the interference color of the film building up in the course of the process. The equipment operated at a low pressure of about 10^{-6} Torr. The sources for elemental Zn and S, reasonably isolated from the vacuum environment, were heated to obtain the desired vapor pressures. By comparing the color to an interference color map the increase of the thickness could be monitored during the process. The first growth experiments were made in August-September 1974. The very first experiment was successful; a film was observed to grow – not, however at the expected rate, one monolayer in a cycle, but only a third of a monolayer in a cycle. It took quite some time before it was understood, how the surface rearrangement accounted for the less-than-monolayer growth.

Steps towards practical processing of EL displays

In 1978, the ALE-EL project with the personnel and the facility was sold to the building material company Lohja Oy.



The very first experiment for a chloride process was tried by combining $ZnCl_2$ and elemental S, then by adding H_2 flow over the heated sulphur source. The experiments were not successful. The famous picture of Sven Lindfors sitting next to the glass tube ALD reactor was taken after the first successful $ZnCl_2/H_2S$ process – what is missing in the picture was Arto Pakkala and his first reaction to the success: "Siinä se on!" ("That's it!")

ALE-EL displays showed outstanding electrical and optical characteristics. The high dielectric strength of the dielectrics (then above 4 MV/cm, nowadays close to 10 MV/cm), with an intrinsic pinhole-free feature, allowed effective excitation of the light emitting ZnS(Mn) layer. Due to the hexagonal crystalline structure of the ALE-ZnS, the color of the light emitted was beautiful yellow, it was less orange than the EL devices based on sputtered ZnS with cubic crystalline structure. The fully transparent structure allowed a black background layer resulting in an outstanding contrast also in bright ambient light. Picture: Arto Pakkala operates reactors used for early ALE-EL prototypes.

1980 - The first public disclosure of TF-EL display

In the 1970s, the ALE-EL development was carried out strictly confidentially – until the first disclosure at the Society for Information Display (SID) conference in San Diego, California, in April 29 to May 1, 1980.

Picture: SID representative and Tuomo Suntola and his key colleagues, Jorma Antson, Sven Lindfors and Arto Pakkala (from the left to the right) received the 1980 SID Outstanding Paper Award for the EL work. The Award was handed to the group in the next year SID conference in 1981.

Commercialization of ALE-EL

In 1983, the pilot production of TF-EL displays started in Lohja, Kunnarla plant. The first real proof-of-concept of the ALE-EL displays was the large information boards installed in the departure hall at Helsinki-Vantaa airport in 1983.

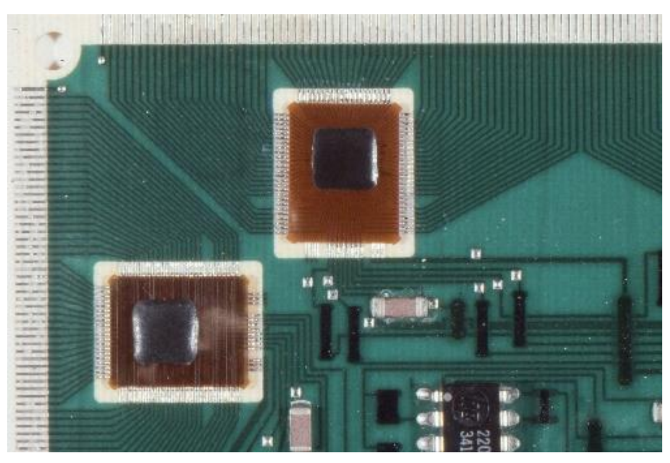
Final testing of the alternatives for dielectric layers in the thin film structure – Al_2O_3, ATO (Aluminum-Tantalum Oxide nano-laminate) or Ta_2O_5 – was carried out in a prototype board installed in an underground cave under the airport. Test modules with ATO dielectrics showed the best performance and reliability. Picture: Ralf Graeffe inspects the display modules in the test assembly in airport underground cave in 1983.

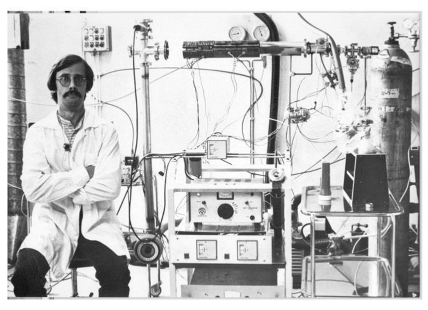
Picture: Olarinluoma plant in 2014, built for EL production in 1983-1984 by Lohja. EL-production continues, operated by Beneq Oy since 2012.

The production of the data modules started gradually in the Olarinluoma facility in 1985 and distribution channels were opened in the USA and in some other major industrialized countries. Following Sharp in Japan and Planar in the US, Lohja became the third EL manufacturer in the world.

In the picture: Tuomo Suntola (right) and Ulf Ström, the managing director of Finlux Inc. responsible for marketing the ALE-EL panels in the USA.

Picture: A corner of the EL panel circuit board with high voltage IC drivers.





40 Years of ALD in Finland – Photos, Stories

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1987 - 1998 Microchemistry Ltd

In 1987, Tuomo Suntola was offered the chance to set up Microchemistry Ltd, as a subsidiary of Neste, for the development of ALE-based solar panels. Suntola saw also the possibility of applying ALE to heterogeneous catalyst, which complemented the objectives decided for the new research unit.

As the first task in Microchemistry, Tuomo Suntola and Sven Lindfors designed a quick, compact research reactor for the PV development. The outcome was the F-120 reactor which also became the first commercial ALD reactor. The F-120 was optimized for fast cycling and easy operation and maintenance.

In the picture, the F-120 reactor. The heavy bars on each side of the reactor were meant to let the researcher lean and ponder while looking at the progress of the process.

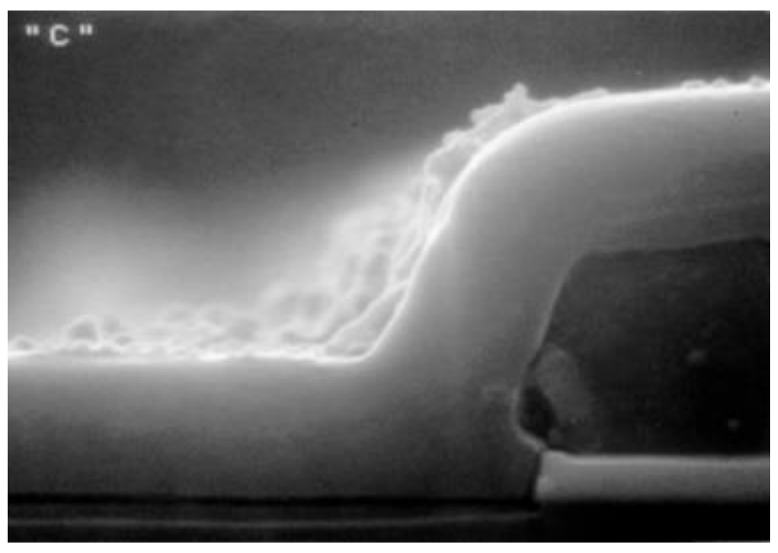
Microchemistry focuses on ALE

Suntola had seen the large potential of ALE from the very beginning of the invention, especially in semiconductor manufacturing. The knowhow in the ALE process and the reactors, not disclosed in the patents, had been kept strictly confidential throughout the development and commercialization of the EL-technology. The new situation offered Microchemistry the possibility to start working on ALE and the reactors for new applications.

Since the SID conference in the 1980, and from numerous other conferences and meetings, Suntola had contacts with key people in the display and semiconductor fields. Thanks to the confidence acquired with ALE-EL Suntola was offered the possibility for a visible introduction of ALE in the MRS 1994 Annual Meeting in Boston. The talk "ALE for Semiconductor Applications" was complemented by Microchemistry's ALE-booth in the exhibition held parallel to the conference.

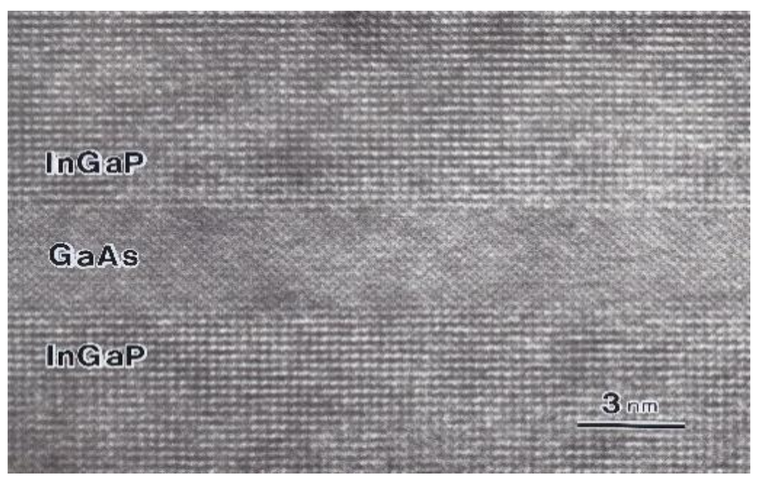
In the picture (left) Heli Vaara, assistant to Tuomo Suntola (right).

The Boston MRS presentation demonstrated the excellent uniformity and conformal coating characteristics of ALE technology. Based on the measurements of the refractive index, the high density of the Al_2O_3 films was also introduced. Actual 8" wafers were shown at the exhibition booth. Gradually, major semiconductor manufacturers and equipment manufacturers became interested in the technology. Picture: Demonstration of the conformality of the ALE-Al_2O_3 passivation layer at the edge of a contact pad in an integrated circuit.



In 1997, there was already a clear indication of the commercial potential of ALE. Microchemistry had sales representatives in the US, Japan and South Korea and several reactors had been delivered to customers. The most impressive contract consisted of several large ALE reactors capable of handling big batches of 500x500 mm^2 substrates (picture).

Spreading of ALE by year 1990



For his 1989 review paper, Suntola mapped known laboratories working on ALE

Suntola recalls that the Japanese III-V activity in mid 1980s was the most visible research effort on ALE. One of the most impressive demonstrations, introduced by Nippon Electric Company (NEC), was the InGaP-GaAs-InGaP superlattice, where the GaAs layer is obtained by applying 11 ALE cycles (the center layer in the picture).

The first International Symposium on Atomic Layer Epitaxy was held in Espoo, June 11-13, 1990. The conference chair was Prof Lauri Niinistö. In the picture, participants at the conference dinner in Herttoniemi, Helsinki, restaurant Vanha Mylly: Dr. Aoyagi, RIKEN; Erja Nykänen, HUT; Tuomo Suntola, Microchemistry Ltd.; Prof. Niinistö, HUT; Prof. Nishizawa, Semiconductor Laboratory, Sendai; Prof. Bedair, North Carolina University.

Epilogue

When handing Microchemistry Ltd. over to his successor Matti Ervasti in the beginning of 1998, Suntola left behind his active role in ALE, now renamed as ALD.

As a research fellow in Neste, now in Fortum Oyj, after the fusion of the state oil company Neste and the electric utility company IVO, Suntola worked for long-term research and global energy issues with main emphasis on renewable energies. He was a member of World Energy Council and served as the attorney of Fortum Foundation until part-time retirement in 2000 and full retirement in the beginning of 2004. In the same year, Suntola joined Picosun Oy, first as the scientific adviser and since 2007 as a board member.

The year 2004 brought a delightful surprise to Suntola in the form of the European SEMI Award 2004 "Honoring the Pioneer in Atomic Layer Deposition Techniques ... that paved the way for the development of nanoscale semiconductor devices". The Award was handed to Suntola at the Munich Electronics Show 2004 by Stanley Myers, the President & CEO of SEMI (USA).

Acknowledgements

About the author

Riikka Puurunen, who has just arrived to start writing the story of ALE. Photo taken on March 6, 2014, by Tuomo Suntola.

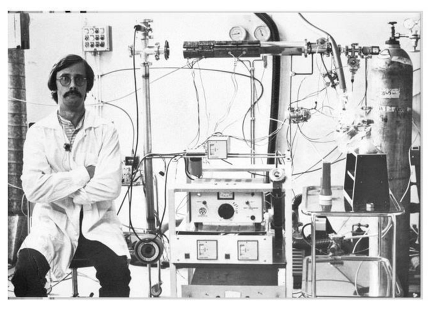
The full story behind these photos and other stories collected in a booklet at the exhibition:

Tuomo Suntola's ALE in Short
by Dr. Riikka Puurunen

Atomic layer deposition - Source materials enabling thin film depositions
by Dr. Marja Tiitta

Remembering Milja Mäkelä
by Dr. Marja-Leena Kääriäinen

Story on ZyALD
by Dr. Jonas Sundqvist



40 Years of ALD in Finland – Photos, Stories

Exhibition by the Finnish Centre of Excellence in Atomic Layer Deposition

By Jaakko Niinistö, Markku Leskelä, Mikko Ritala, Laboratory of Inorganic Chemistry, University of Helsinki, Helsinki, Finland

Atomic Layer Deposition Research at Helsinki University of Technology (Aalto University)

Prof. Lauri Niinistö, head of the Laboratory of Inorganic and Analytical Chemistry at Helsinki University of Technology (TKK, now Aalto University) was working on luminescent materials with his graduate student Markku Leskelä in the late 1970s. In 1981 Runar Törnqvist from Lohja Oy contacted Prof. Niinistö in order to get help to analyze the Na ion diffusion in ALD (that time ALE) grown EL display structures which Lohja Oy was about to commercialize. In addition, Lohja Oy was interested in the luminescent studies published in the doctoral thesis of Markku Leskelä (1980). As a result Lohja Oy offered research collaboration and donated the first ALD reactor to LIAC in 1981 (Figure 1), called with the code JASKA. The JASKA reactor was installed and the first ALD film (Mn-doped ZnS) was grown at LIAC in 1982.

The ALD research at LIAC in the early 1980's was concentrating strictly on the development of EL structures. The first M.Sc. student was Markku Tammenmaa. Markku Leskelä was supervising together with Lauri Niinistö his thesis. Other key persons involved were Tarja Koskinen and Lassi Hiltunen. The main task in the early years was to solve problems in product manufacturing, not to publish in scientific journals. Hence, the first publications from the ALD studies at LIAC appeared in 1984 (First symposium on ALE) and in 1985 (Thin Solid Films). During the first years research collaboration with Lohja Oy (later Planar Ltd.) was confidential. This collaboration enabled funding from the Finnish Technology Agency (TEKES) and provided invaluable start for the future studies.

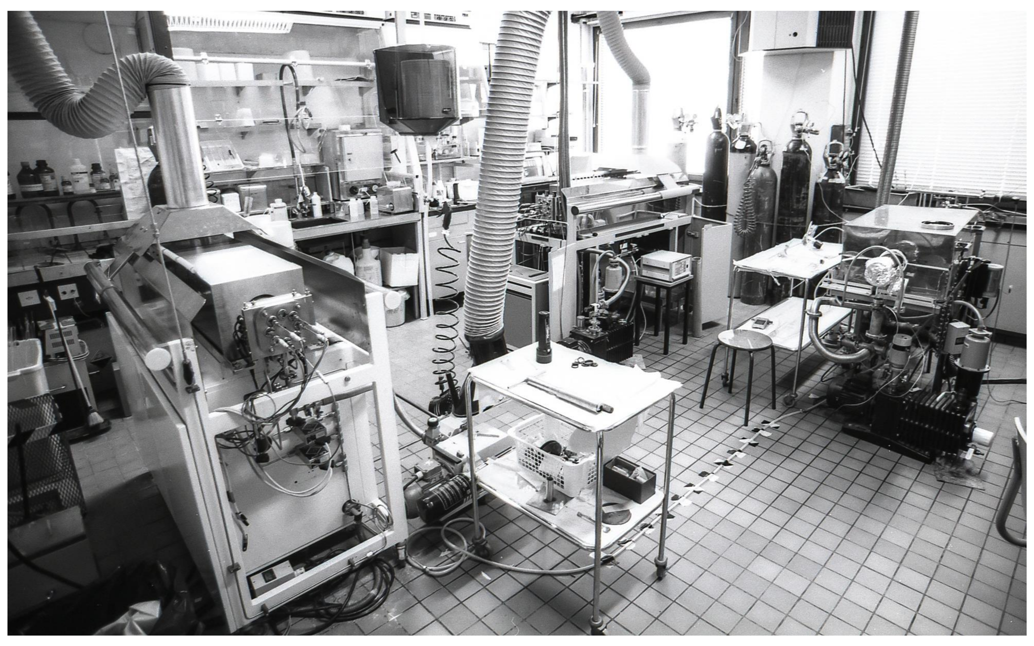


Fig. 1. ALD laboratory at LIAC, JASKA reactor in the right, other two reactors are Microchemistry F-120 reactors. Photo by Matti Putkonen 1998.

Later in 1980's the research efforts were strongly devoted to EL display studies (barrier layers, full color development). The strong background that both M. Leskelä and L. Niinistö had on the chemistry of rare earth elements enabled important ALD process development in the field of rare earth oxide thin films. Key members in the group that time included Milja Asplund (later Mäkelä), Marja Tiitta, Erja Nykänen and Pekka J. Soininen. Because of the projects established, the first commercial ALD reactor, Microchemistry MC-120 was acquired in 1989. One important major EU funded project in the 1990s was focusing on full color EL display development. Also a new F-120 reactor was acquired. In the late 1990s research efforts were slowly turning to areas other than EL display studies. For example, dielectric layers for microelectronics were of an interest. That time Matti Putkonen joined the group and acted as the instructor of numerous thesis while Prof. Niinistö was the supervisor until his retirement in 2007. Today successful ALD/MLD research at the laboratory in the Aalto University is led by Prof. Maarit Karppinen. Today the laboratory has three F-120 reactors and a Picosun reactor.

TKK (+collaboration), Major outcomes:

- ALD process for ZnO (1985)
- ALD processes for NbN_x and TiN_x (1988)
- SnO_2 for gas sensors (1994)
- HfO_2 and TiO_2 from chlorides and H_2O (1994)
- delta-doping of SnO_2 with Sb (1996)
- coating of porous silicon (PS) with SnO_2; aspect ratio 140:1 (1996)
- *in situ* synthesis of ALD precursors (1996)
- ALD processes for RE oxides, also ternary
- YSZ for SOFCs (2002)
- Cp-precursors for Hf-, Zr-, and rare earth oxides (1999-)
- amorphous high-permittivity $YScO_3$ (2006)

ALD precursors tested at TKK

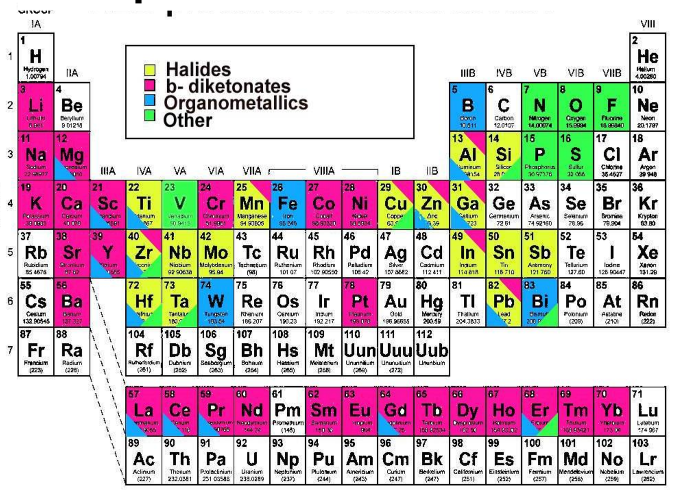


Fig. 2. ALD laboratory at LIAC in 2003, Jenni Harjuoja (left) and Anne Kosola (right) working with F-120 reactors. Photo by Matti Putkonen.

Atomic Layer Deposition Research at University of Helsinki

Thin film research began in Laboratory of Inorganic Chemistry (LIC) at University of Helsinki in 1991 and the group led by professors Markku Leskelä and Mikko Ritala has grown steadily to the present size of about 25 persons. LIC is the world leading developer of new chemistry and processes for ALD.

Beginning Markku Leskelä was nominated as the Professor of inorganic chemistry in 1990. He soon received a project funded by the University, Academy of Finland and TEKES which enabled the purchasing of a X-ray diffractometer and one F-120 ALD reactor from Microchemistry. In the project the main focus was in oxide films and many basic processes on Ti, Zr, Hf oxides from chlorides and water were published. Nucleation of the films was studied by the newly discovered AFM/STM techniques. Later several other oxide processes were examined and materials selection was expanded to nitride films.

1990's During 90s research was made in about 15 projects on electroluminescent thin film displays, corrosion protection and microelectronics. Separate projects to develop new precursor chemistry were also carried out. Mikko Ritala was funded since 1991 by Academy of Finland as a PhD student, post doc and Academy fellow for eleven years. One of the important outcomes from that basic research was the building of *in situ* system where the growth process is monitored by a quartz crystal microbalance and quadrupole mass spectrometer – the first system combining these in the world.

At the end of 90s it became clear that ALD will be extremely important for microelectronics and many processes for dielectric, conducting and barrier films were studied. Already on that time the first processes for ternary oxides ($SrTiO_3$) were developed. Tens of processes were studied for Zr and Hf oxides, making nanolaminates, and doping them.

2000's Studies on noble metal films started in collaboration with ASM Microchemistry. The research has been very successful and processes for all platinum group metals could be developed. Later the use of ozone and oxygen precursor enabled the lowing of the process temperature significantly.
In 2004 the moving of the R&D laboratory of ASM Microchemistry in Department of Chemistry made a big boost in the ALD research in Laboratory of Inorganic Chemistry. This collaboration aims at development of ALD processes for microelectronics. During the years processes have been developed for high-k materials, metals, low-k materials and selective ALD. Another significant funding at that time came from Academy of Finland in the form of Academy Professor position for Markku Leskelä.

Nanotechnology has been an obvious application area for ALD. Nanofibers, nanotubes and nanoporous materials of very different materials have been prepared and characterized.

Since mid 90s different EU projects, Marie Curie activities, MEDEA and COST projects have been actively participated. Many projects have been made in collaboration with foreign companies (e.g. Motorola, IBM, Intel, OmniPV, Philips, Qimonda, Photonis). Collaboration with chemical companies (Air Liquide, Honeywell, Air Products, Praxair, ATMI, Inorgtech/Epichem, SAFC High Tech) was important and many of the new molecules synthesized by them were tested as ALD precursors.

The research has resulted in more than 350 publications, several review papers 23 PhD thesis, more than ten patents, and numerous conference presentations.

Laboratory of Inorganic Chemistry today
The main activities today are devoted to Centre of Excellence in ALD where ALD processes for microelectronics and energy applications are developed. The collaboration with ASM Microchemistry continues and a few EU and company funded projects are ongoing.

The number of ALD reactors in LIC has increased from 1 in 1991 to nine in 2014. One reactor is equipped with QMS and QCM for in situ reaction mechanism studies and one with a plasma source. In addition, LIC has other film deposition techniques available as well as excellent facilities for precursor synthesis and thin film characterization.

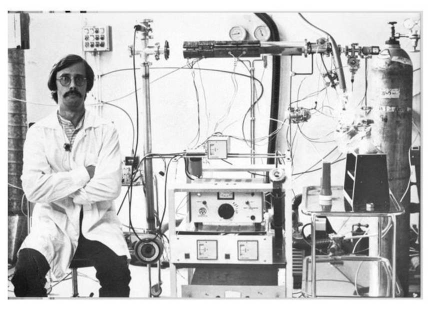
At the moment the personnel in LIC in thin film and nanomaterials group consists of 2 professors, 2 university lecturers, 3 senior scientists, 3 post docs, 13 PhD students, 4 MSc students.

UH, Major outcomes:

- range of new precursors
- reaction mechanism understanding by QCM & QMS
- numerous TiO_2, ZrO_2, HfO_2 processes
- $SrTiO_3$ ALD process
- noble metal and noble metal oxide processes
- nanolaminates
- $Ge_2Sb_2Te_5$ and other chalcogenides with silyl chalcogen precursors
- metal nitride processes
- metal fluoride processes
- TiO_2-based photocatalysts
- PEALD of Ag, Cu and oxides
- selective area ALD

Fig. 3. Timo Asikainen working with F-120 reactor in 1994.

updated 11.6.2014



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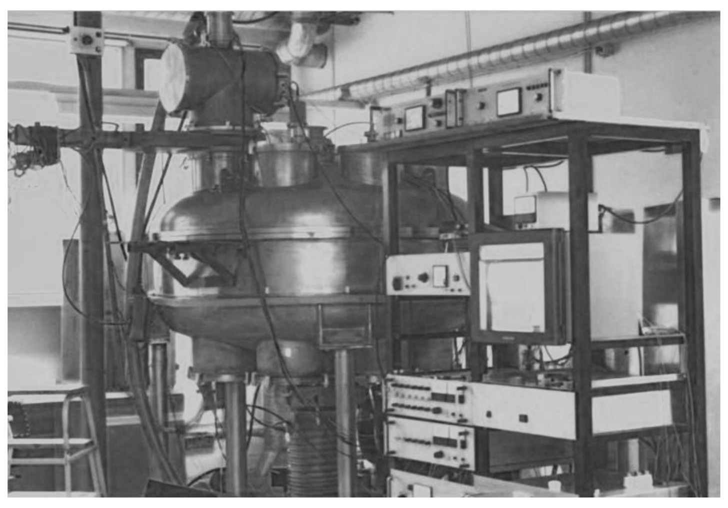
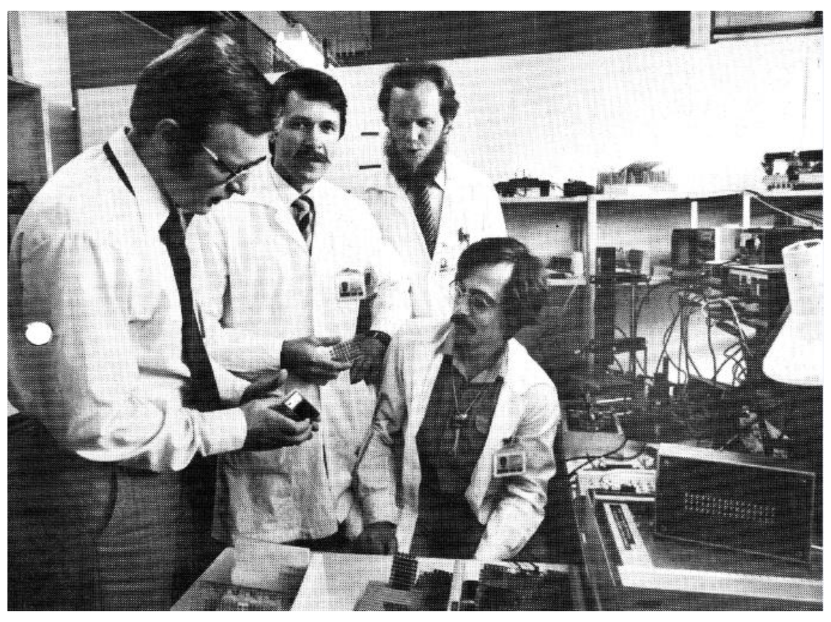
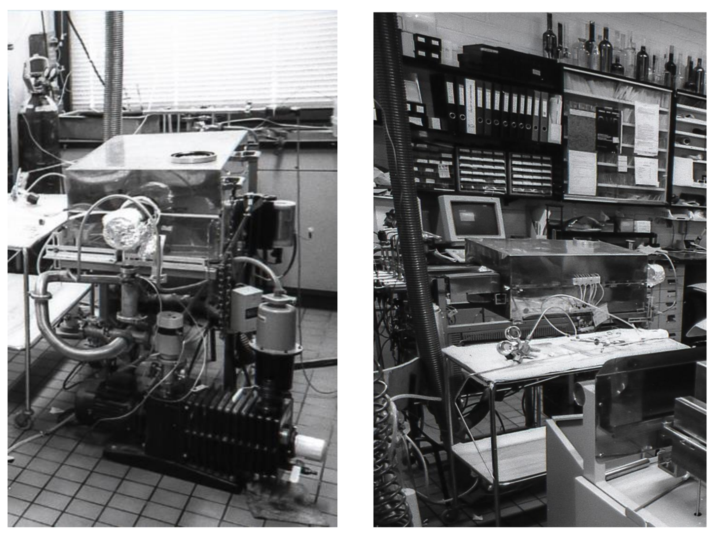
Photos related to the development of ALD in Finland



Big carousel ALE reactor from 1977. Photo provided to the exhibition by Dr. Tuomo Suntola.

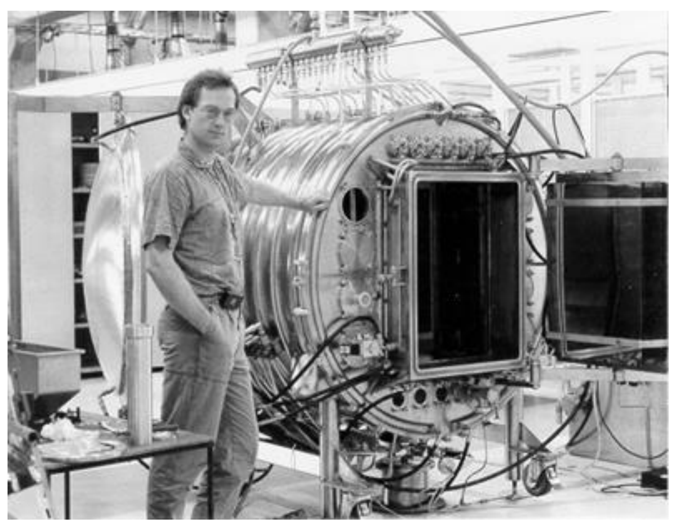


The core ALE-EL team Dr. Tuomo Suntola, Jorma Antson, Arto Pakkala, and Sven Lindfors featured in an article published in Turun Sanomat on 14.5.1980. Copy from Dr. Tuomo Suntola's collections.

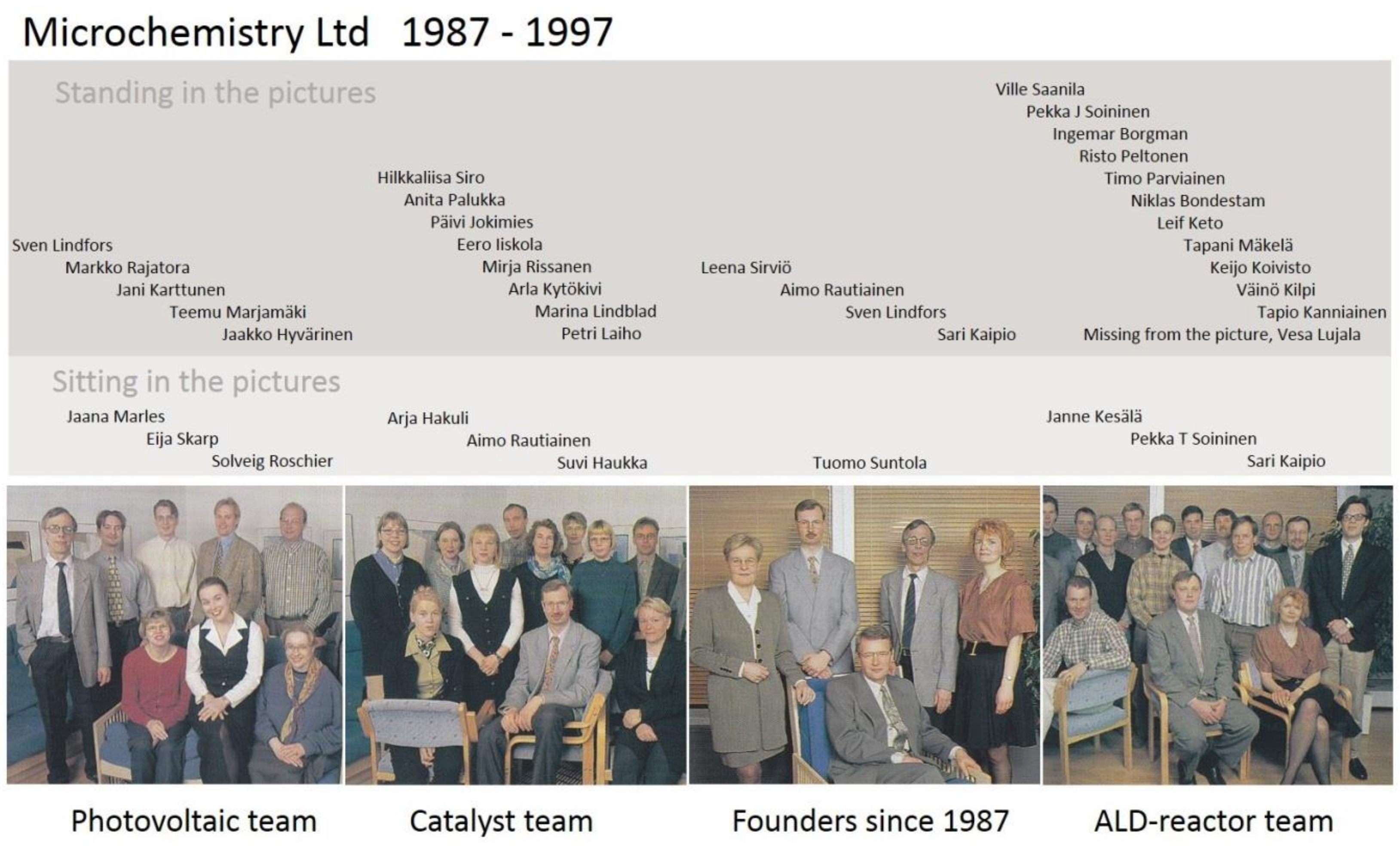


The first ALE reactor at the Helsinki University of Technology known as "Jaska-1", in laboratory of Inorganic Chemistry in 1998. According to Jaakko Hyvärinen, this was Finland's first computer-controlled ALE reactor ("ABC computer" from Sweden). Photo is a copy from the archives of Dr. Matti Putkonen.

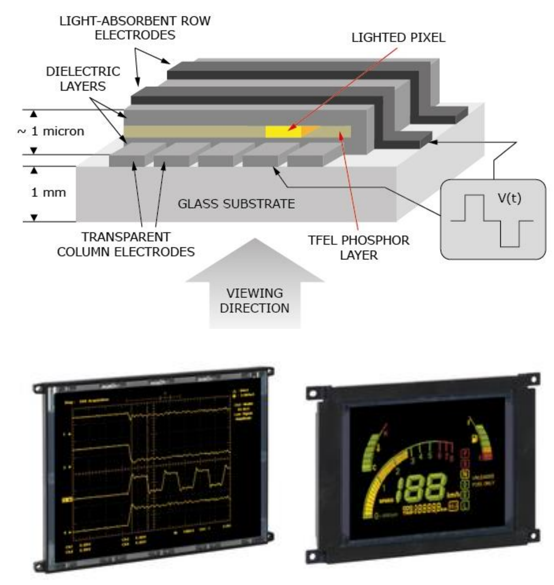
FINLUX flight information board with ALE-EL character modules operating at the Helsinki-Vantaa airport. Photo taken in 1983, when the first boards had been installed. Sitting on the left side is Karri Kuusikko, now Area Sales Director for Lumineq Displays, and on the right Finlux design engineer Jukka Vaajakallio. Photo originally taken for Lohja Oy, now rights owned by Beneq Oy. Provided to the exhibition by Joe Pimenoff.



Jarmo Skarp and the P-1000 pilot batch reactor for flat panel displays. Photo taken 28.8.1985, provided to the exhibition by Dr. Suvi Haukka.



Microchemistry 1987 - 1997. Photo provided by Dr. Tuomo Suntola.

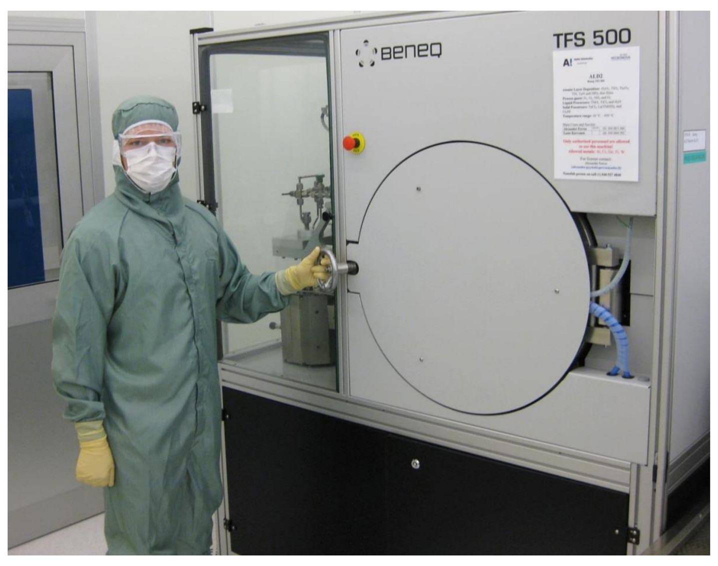


Top: Operation principle of the TFEL structure. Bottom: Two examples of ALD-based TFEL displays. On the left: yellow monochrome display, specified to operate at -60C with no external heating and on the right: tri-colour TFEL display, both by Lumineq Displays, Beneq. (Commercial full-colour displays have not been made by ALD.) Photos: Beneq, provided to the exhibition by Joe Pimenoff.

ALE reactor built for III-V growth at the Semiconductor Laboratory, VTT Technical Research Centre of Finland, basement in Otakaari 7B, Otaniemi, Espoo. III-V research was conducted at VTT by Ilkka Suni, Jouni Ahopelto and Hannu Kattelus. Precursors were gallium and indium chlorides and arsine. The reactor was initially tested by depositing GaN and AlN. Photo is 1987 and was provided to the exhibition by Prof. Jouni Ahopelto.

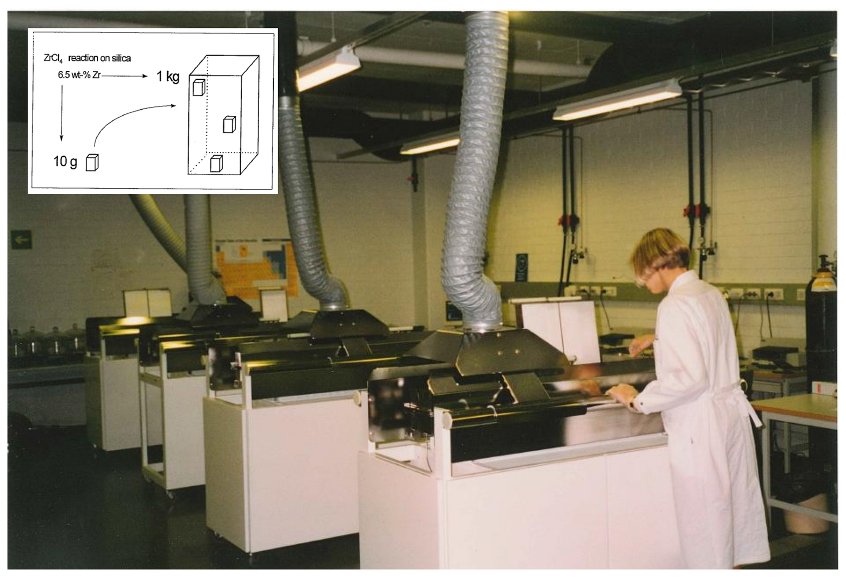
Fluidized bed reactor insert for an F-120 reactor, for creating ALD coatings on particles. The reactor has been used for catalysis ALD research at the University of Joensuu. Photo provided by Prof. Tapani Pakkanen.

MIcronova building, Tietotie 3, Espoo, in a winter evening. VTT and Aalto share the Micronova clean room, which also companies such as Picosun Oy and Beneq Oy make use of. Photographer: Dr. Philippe Monnoyer.



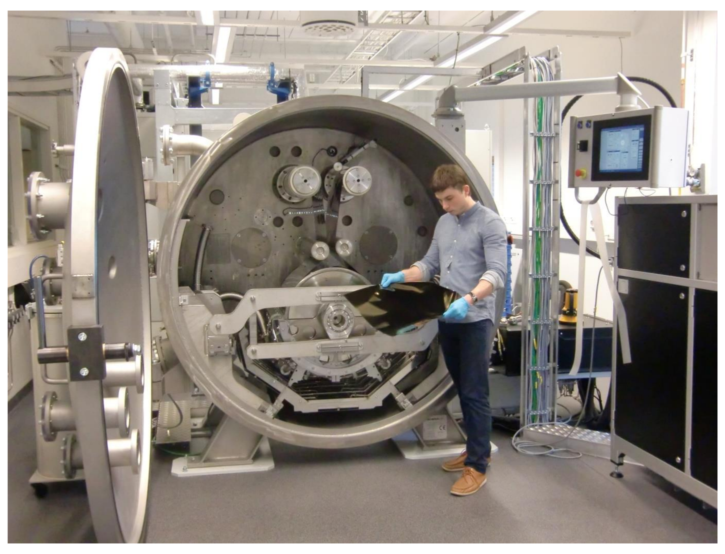
The first Beneq TFS-500 ALD reactor was installed at Micronova, Aalto University, in 2005. Photo provided by Alexander Pyymäki Perros.

VTT bought the first prototype of the Picosun SUNALE™ R-150 reactor in 2004. In the picture, Riikka Puurunen, VTT, operates the Picosun reactor installed in the Micronova clean room, Otaniemi, Espoo, in 2005. Provided to the exhibition by Dr. Satu Ek.



Microchemistry Catalyst laboratory in the 1990's, showing four F-120 reactors, each fitted with a fixed-bed reaction space for about 10 g of material. Jaana Marles is at the reactor. A larger fixed-bed reactor (called "Möhkö") was also built at Microchemistry, demonstrating the scale-up possibilities with ALE catalysts. Photo from Dr. Suvi Haukka, image (insert) from Haukka, Lakomaa, Suntola, Stud. Surf. Sci. Catal. 120A (1999) 715-750.

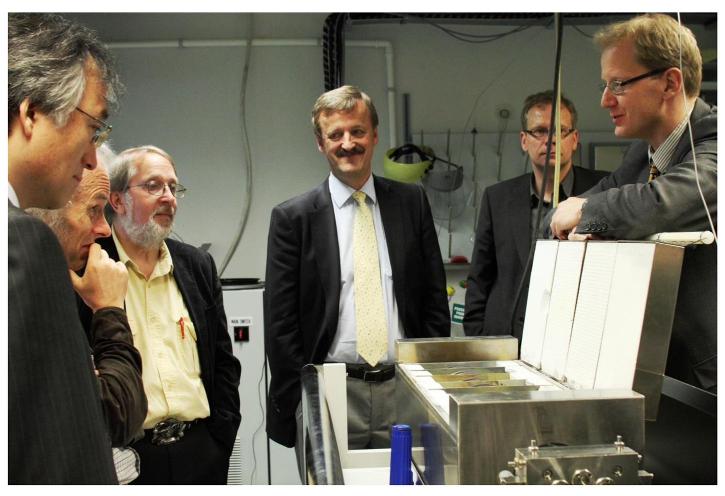
Dr. Tapani Alasaarela introduced the prototype of his cheap build-it-yourself ALD reactor at the ALD miniseminar in Micronova, Espoo, on December 17, 2013. Photographer: Dr. Riikka Puurunen.



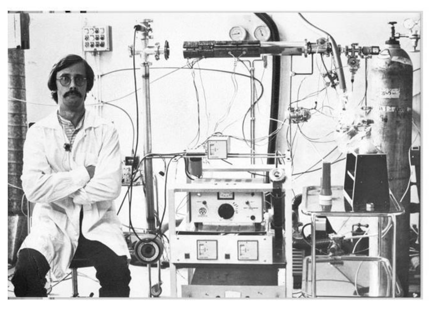
Mr. Philip Maydannik inspecting an aluminium oxide film on a flexible metallised PET substrate made by spatial roll-to-roll ALD with the Beneq WCS500 reactor. Photo taken on 30.5.2013 in the ASTRaL lab in Mikkeli, Finland. Photo provided to the exhibition by Prof. David Cameron, who started the R&R activities in Mikkeli in 2004. Research on ALD in ASTRaL began in 2005 and in spatial ALD in 2008. Photographer: Dr. Kimmo Lahtinen.

Pekka Soininen and Jarmo Skarp showing a Zn(O,S) film on a 1200x1200 mm2 glass substrate, made with the Beneq TFS 1200 in-line ALD reactor in December 2009. Cycle time was 2s, and the film was made from alternated ZnO and ZnS layers. The reactants were diethylzinc, water, and H2S at about 120C. Photo: Beneq, provided to the exhibition by Pekka Soininen & Joe Pimenoff.

PICOSUN™ P-1000 ultra-large scale batch ALD reactor prototype in 2013, with Dr. Marko Pudas. Photo provided to the exhibition by Dr. Minna Toivola.



The Scientific Advisory Board members of the Finnish Centre of Excellence on Atomic Layer Deposition during a laboratory tour at Universtity of Helsinki, Inorganic Chemistry, in June 1, 2012. From left: Prof Hyeongtag Jeon, Prof Marc Heyns, Prof Roger Webb, Prof Markku Leskelä (director of the CoE), Dr. Jaakko Niinistö, and Prof. Mikko Ritala (vice-director of the CoE). In the front, opened, F-120 reactor by ASM Microchemistry. Photographer: Dr. Riikka Puurunen.



40 Years of ALD in Finland – Photos, Stories

Exhibition by the Finnish Centre of Excellence in Atomic Layer Deposition

Academic theses made related to ALD in Finland

Doctoral theses

Author	Year	Title	University	pages
Veli-Pekka Tanninen	1983	X-ray diffraction studies of aluminium powder and electroluminescent zinc sulphide thin films	Helsinki University of Technology, Department of Technical Physics, Laboratory of Physics	
Runar Törnqvist	1983	Electroluminescence in ZnS:Mn thin film structures grown by atomic layer epitaxy	Helsinki University of Technology, Department of Technical Physics, Laboratory of Physics	34
Jukka Lahtinen	1987	Electro-optical studies of semiconductor compounds for electroluminescent and laser devices	Helsinki University of Technology, Department of Technical Physics, Laboratory of Physics	51
Markku Oikkonen	1988	X-ray diffraction and ellipsometric studies of zinc sulfide thin films grown by atomic layer epitaxy	Helsinki University of Technology, Department of Technical Physics, Laboratory of Physics	46
Markku Tammenmaa	1988	The atomic layer epitaxy growth and characterization of zinc sulfide and alkaline earth sulfide thin films for electroluminescent applications	Helsinki University of Technology, Laboratory of Inorganic and Analytical Chemistry	13
Marina Lindblad	1992	Cluster models for chemisorption on non-metallic surfaces	University of Joensuu, Department of Chemistry	25
Suvi Haukka	1993	Characterization of surface species generated in atomic layer epitaxy on silica	University of Helsinki, Department of Chemistry, Analytical Chemistry Division	46
Markku Åberg	1993	An electroluminescent display simulation system and its application for developing grey scale driving methods	VTT Electronics. Acta Polytechnica Scandinavica. Electrical Engineering Series 74.	76
Lars-Peter Lindfors	1994	Hydrogenation of toluene on supported nickel - from catalyst preparation to reaction kinetics	Åbo Akademi, Department of Chemical Engineering	
Mikko Ritala	1994	Atomic layer epitaxy growth of titanium, zirconium and hafnium dioxide thin films	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	48
Markku Ylilammi	1996	Preparation and analysis of thin film electroluminescent devices	VTT Electronics	79
Janne Jokinen	1997	Time-of-flight spectrometry of recoiled atoms in the analysis of thin films	University of Helsinki, Department of Physics, Accelerator Laboratory	38
Arla Kytökivi	1997	Growth of ZrO2 and CrOx on high surface area oxide supports by atomic layer epitaxy	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Inorganic and Analytical Chemistry	54
Marja Tiitta	1998	Studies on precursors and their application in the atomic layer epitaxy growth of thin films for electroluminescent devices	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Inorganic and Analytical Chemistry	44
Arja Hakuli	1999	Preparation and characterization of supported CrOx catalysts for butane dehydrogenation	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Industrial Chemistry	48
Maria Kurhinen	1999	Interactions of Mo(CO)6 and Co2(CO)8 with alumina and silica supports: IR spectroscopic, modelling and temperature programmed studies	University of Joensuu, Department of Chemstry	36
Sari Myllyoja	1999	Chromium hexacarbonyl supported on alumina and silica surfaces by gas phase adsorption; characterisation and activity in hydrodesulphurisation	University of Joensuu, Department of Chemstry	36
Mika Suvanto	1999	Hydrotreating catalysts based on tungsten hexacarbonyl: Controlled preparation, characterisation and activity in thiophene hydrodesulphurisation	University of Joensuu, Department of Chemstry	34
Sari Suvanto	1999	Co2(CO)8 adsorbed on SiO2 and MCM-41: Gas phase preparation and characterisation	University of Joensuu, Department of Chemstry	33
Mikko Utriainen	1999	Exploiting atomic layer epitaxy thin film deposition technique in solid-state chemical sensor applications	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Inorganic and Analytical Chemistry	80
Wei-Min Li	2000	Characterization and modification of SrS based blue thin film electroluminescent phosphors	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	44
Marika Juppo	2001	Atomic Layer Deposition of Metal and Transition Metal Nitride Thin Films and In Situ Mass Spectrometry Studies	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	65
Minna Nieminen	2001	Deposition of binary and ternary oxide thin films of trivalent metals by atomic layer epitaxy	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Inorganic and Analytical Chemistry	57
Jarkko Räty	2001	Re2(CO)10 deposited on g-alumina: temperature programmed studies, modelling and activity in thiophene hydrodesulphurization	University of Joensuu, Department of Chemstry	34
Matti Putkonen	2002	Development of low-temperature deposition processes by atomic layer epitaxy for binary and ternary oxide thin films	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Inorganic and Analytical Chemistry	69
Riikka Puurunen	2002	Preparation by atomic layer deposition and characterisation of catalyst supports surfaced with aluminium nitride	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Industrial Chemistry	78
Antti Rahtu	2002	Atomic layer deposition of high permittivity oxides: film growth and in situ studies	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	69
Timo Sajavaara	2002	Heavy ion recoil spectroscopy of surface layers	University of Helsinki, Department of Physical Sciences, Accelerator Laboratory	64
Jaana Kanervo	2003	Kinetic analysis of temperature-programmed reactions	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Industrial Chemistry	76
Satu Ek	2004	Controlled preparation of aminofunctionalized surfaces on porous silica by atomic layer deposition	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Inorganic and Analytical Chemistry	54
Mohamed Lashdaf	2004	Preparation and characterisation of supported palladium, platinum and ruthenium catalysts for cinnamaldehyde hydrogenation	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Industrial Chemistry	70
Raija Matero	2004	Atomic Layer Deposition of Oxide Films – Growth, Characterisation and Reaction Mechanism Studies	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	66
Titta Aaltonen	2005	Atomic Layer Deposition of Noble Metal Thin Films	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	71
Sanna Airaksinen	2005	Chromium oxide catalysts in the dehydrogenation of alkanes	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Industrial Chemistry	58
Petra Alen	2005	Atomic Layer Deposition of TaN, NbN and MoN films for Cu metallization	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	72
Jaakko Niinistö	2006	Atomic layer deposition of high-k dielectrics from novel cyclopentadienyl-type precursors	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Inorganic and Analytical Chemistry	72
Antti Niskanen	2006	Radical enhanced atomic layer deposition of metals and oxides	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	77
Jani Päiväsaari	2006	Atomic layer deposition of lanthanide oxide thin films	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Inorganic and Analytical Chemistry	69
Jenni Harjuoja	2007	Atomic layer deposition of binary and ternary lead and bismuth thin films	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Inorganic and Analytical Chemistry	59
Kimmo Solehmainen	2007	Fabrication of microphotonic waveguide components on silicon	VTT Technical Research Centre of Finland / Helsinki University of Technology	68
Marko Vehkamäki	2007	Atomic layer deposition of multicomponent oxide materials	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	69
Tarja Zeelie (née Kainulainen)	2007	Rhodium and cobalt catalysts in the heterogeneous hydroformylation of ethene, propene and 1-hexene	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Industrial Chemistry	72
Abuduwayiti Aierken	2008	Passivation of GaAs surfaces and fabrication of self-assembled In(Ga)As/GaAs quantum ring structures	Aalto University, Department of Micro and Nanosciences	46
Kai-Erik Elers	2008	Copper diffusion barrier deposition on integrated circuit devices by atomic layer deposition technique	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	72
Kai Kolari	2008	Fabrication of silicon and glass devices for microfluidic bioanalytical applications	VTT Technical Research Centre of Finland / Helsinki University of Technology	72
Satu Korhonen	2008	Effect of Support Material on the Performance of Chromia Dehydrogenation Catalysts	Helsinki University of Technology, Faculty of Chemistry and Materials Sciences, Department of Biotechnology and Chemical Technology, Industrial Chemistry	72
Ville Miikkulainen	2008	Molybdenum nitride thin films on micro- and nanopatterned substrates: atomic layer deposition and applications	University of Joensuu, Department of Chemistry	44
Tero Pilvi	2008	Atomic Layer Deposition for optical applications: metal fluoride thin films and novel devices	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	103
Jouni Tiilikainen	2008	Novel genetic fitting algorithms and statistical error analysis methods for X-ray reflectivity analysis	Helsinki University of Technology, Faculty of Electronics, Communications and Automation, Department of Micro and Nanosciences	51
Satu Korhonen	2008	Effect of Support Material on the Performance of Chromia Dehydrogenation Catalysts	Helsinki University of Technology, Faculty of Chemistry and Materials Sciences, Department of Biotechnology and Chemical Technology, Industrial Chemistry	72
Leif Backman	2009	Supported Cobalt Catalysts – Preparation, Characterisation and Reaction Studies	Helsinki University of Technology, Faculty of Chemistry and Materials Sciences, Department of Biotechnology and Chemical Technology, Industrial Chemistry	59
Jarkko Ihanus	2010	Atomic layer deposition of electroluminescent ZnS, SrS, and BaS thin films	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	62
Pia Myllymäki	2010	High-κ ternary rare earth oxides by atomic layer deposition	Aalto University, School of Science and Technology, Faculty of Chemistry and Materials Sciences, Department of Chemistry, Laboratory of Inorganic Chemistry	50
Viljami Pore	2010	Atomic layer deposition and photocatalytic properties of titanium dioxide thin films	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	89
Mikko Tapani Saloniemi	2010	The significance of brittle reaction layers in fusing of dental ceramics to titanium	Department of Stomatognathic Physiology and Prosthetic Dentistry; Institute of Dentistry; University of Helsinki, Helsinki, Finland; Department of Electronics, Faculty of Electronics, Communications and Automation, School of Science and Technology, Aalto University, Espoo, Finland	102
Tapani Alasaarela	2011	Atomic layer deposited titanium dioxide in optical waveguiding applications	Aalto University, School of Electrical Engineering, Department of Micro- and Nanosciences, Photonics Group	63
Nikolai Chekurov	2011	Fabrication process development for silicon micro and nanosystems	Aalto University, School of Electrical Engineering, Department of Micro and Nanosciences, Micro and Quantum Systems Group	65
Elina Färm	2011	Selective-area atomic layer deposition	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	71
Terhi Hirvikorpi	2011	Thin Al2O3 Barrier Coatings Grown on Bio-based Packaging Materials by Atomic Layer Deposition	VTT Technical Research Centre of Finland / Aalto University	74
Tommi Kääriäinen	2011	Polymer surface modification by atomic layer deposition	Lappeenranta University of Technology, Faculty of Technology, Mechanical Engineering, Materials Technology	79
Kristina Uusi-Esko	2011	Synthesis and Characterization of Ternary Manganese Oxides	Aalto University, School of Chemical Technology, Department of Chemistry, Laboratory of Inorganic Chemistry	47
Aap Varpula	2011	Electrical properties of granular semiconductors - Modelling and experiments on metal-oxide gas sensors	Aalto University, School of Electrical Engineering, Department of Micro and Nanosciences, Electron Physics Group	78
Kjell Knapas	2012	In situ reaction mechanism studies on atomic layer deposition	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	33
Antti J. Niskanen	2012	Thin film technology for chemical sensors	Aalto University, Department of Micro and Nanosciences, Microfabrication Group	109
Anna Rissanen	2012	Microsystems for biological cell characterization	VTT Technical Research Centre of Finland / Aalto University, School of Electrical Engineering	130
Timothee Blaunquart	2013	Atomic layer deposition of groups 4 and 5 transition metal oxide thin films: focus on heteroleptic precursors	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	67
Jani Hämäläinen	2013	Atomic layer deposition of noble metal oxide and noble metal thin films	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	131
Lasse Karvonen	2013	Nanostructures for photonic applications	Aalto University, School of Electrical Engineering, Department of Micro- and Nanosciences, Nanotechnology Group	64
Mizohata Kenichiro	2013	Progress in Elastic Recoil Detection Analysis	University of Helsinki, Department of Physics, Accelerator Laboratory	53
Juuso Korhonen	2013	Studies on Wettability - From Fundamental Concepts and Nanofibrous Materials to Applications	Aalto University, School of Science, Department of Applied Physics, Molecular Materials & Soft Matter and Wetting	70
Marja-Leena Kääriäinen	2013	Atomic layer deposited titanium and zinc oxides; structure and doping effects on their photoactivity, photocatalytic activity and bioactivity	Lappeenranta University of Technology, LUT School of Technology, LUT Energy, Energy Technology	74
Jari Malm	2013	Surface functionalization by atomic layer deposited binary oxide thin films	Aalto University, Department of Chemistry, Laboratory of Inorganic Chemistry	28
Joan J. Montiel i Ponsoda	2013	Analysis of photodarkening effects in ytterbium-doped laser fibers	Aalto University, School of Electrical Engineering, Department of Micro- and Nanosciences, Nanotechnology Group (earlier: Photonics Group)	81
Päivi Sievilä	2013	Microfabrication technologies for single-crystal silicon sensors	Aalto University, School of Electrical Engineering, Department of Micro and Nanosciences, Micro and Quantum Systems Group	53
Tommi Tynell	2013	Atomic layer deposition of thermoelectric ZnO thin films	Aalto University, Department of Chemistry, Laboratory of Inorganic Chemistry	40
Henri Jussila	2014	Integration of GaAsP based III-V compound semicinductors to silicon technology	Aalto University, Department of Micro and Nanosciences	68
Juha Nikkola	2014	Polymer hybrid thin-film composites with tailored permeability and antifouling performance	VTT Technical Research Centre of Finland / Tampere University of Technology	84
Yoann Tomczak	2014	In situ characterization of ALD processes and study of reaction mechanisms for high-k metal oxide formation	University of Helsinki, Faculty of Science, Department of Chemistry, Laboratory of Inorganic Chemistry	52

Licentiate's theses

Author	Year	Title, English	University	pages
Olli Jylhä	1986	Cadmium Telluride Based Thin Film Structures Grown by Atomic Layer Epitaxy	Tampere University of Technology, Department of Electrical Engineering	32
Pipsa Makkonen	1987	Pintareaktioiden teoria: pii-pintojen rakenne ja epitaksiaaliset kasvatusmenetelmät	Joensuun yliopisto, Matemaattis-luonnontieteellinenl tiedekunta, Kemian laitos	
Jouni Ahopelto	1991	Atomic layer epitaxy of III-V compound semiconductors in a hydride vapor phase system	Helsinki University of Technology, Department of Electrical Engineering, Electron Physics Laboratory	54
Outi Tolonen	1992	Study of thin film structures by EXAFS method	Helsinki University of Technology, Department of Chemical Engineering, Inorganic Chemistry	122
Heini Mölsä	1994	Preparation of oxide thin films for the electronic applications of superconductors	Helsinki University of Technology, Department of Chemical Engineering, Inorganic Chemistry	46
Heli Viirola	1994	Deposition of tin dioxide thin films by atomic layer epitaxy.	Teknillinen korkeakoulu, Prosessi- ja materiaalitekniikan osasto	
Timo Asikainen	1995	Deposition of indium oxide and indium tin oxide thin films by Atomic Layer Epitaxy method	Helsingin yliopisto, Kemian laitos, Epäorgaanisen kemian laboratorio	52
Timo Ranta-aho	1996	Preparation and Study of Thin Film Materials and Structures for Blue Electroluminescent Devices	Helsinki University of Technology, Faculty of Information Technology, Department of technical Physics	20
Mikko Utriainen	1997	Feasibility of Combining Atomic Layer Epitaxy Thin Films with Porous Silicon Substrate in Gas Sensor Applications.	Helsinki University of Technology, Department of Chemistry, Laboratory of Inorganic Chemistry	
Hannele Heikkinen	1999	Optoelektroniikassa käytettävien sulfidiohutkalvojen valmistus ja karakterisointi	Helsinki University of Technology, Department of Chemistry, Laboratory of Inorganic Chemistry	55
Riikka Puurunen	2000	Preparation of aluminium nitride on porous silica and alumina	Helsinki University of Technology, Department of Chemical Technology, Laboratory of Industrial Chemistry	27
Petra Alén	2003	Atomic Layer Deposition of Conductive Tantalum Nitride Thin Films	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	
Päiväsaari, Jani	2003	Thin Films of Rare Eath Oxides by Atomic Layer Deposition.	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	56
Kosola, Anne	2005	Atomic layer deposition and post-deposition annealing of perovskite oxide thin films.	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	75
Hoffrén, Hannu	2009	Applications of photonic crystal fibers with coated or liquid crystal filled inclusions.	Teknillinen korkeakoulu, Mikro- ja nanotekniikan laitos, Optiikka ja molekyylimateriaalit	69
Eero Santala	2009	Inorganic nanostructures prepared by electrospinning and atomic layer deposition	University of Helsinki, Department of Chemistry, Laboratory of Inorganic Chemistry	62
Sari Sirviö	2014	Characterization of Atomic Layer Deposited Thin Films: Conformality in High Aspect Ratio Pores and the Electrical Properties of Capacitors	Aalto University, School of Electrical Engineering, Doctoral Programme in Electrical Engineering	77
Suvi Särkijärvi	2014	Atomic layer deposition growth of epitaxial zinc oxide	Aalto University, Department of Micro and Nanosciences	79

Master's theses

Author	Year	Title, English	University	Pages
Arto Pakkala	1976	Electroluminescence in zinc sulfide	Helsinki University of Technology	
Markku Ylilammi	1979	Growth of thin oxide and aluminium oxide films through alternating surface reactions	Teknillinen korkeakoulu, Sähköteknillinen osasto	67
Jarmo Skarp	1980	Doping of ALE ZnS for producing different colours in electroluminescent thin films	Teknillinen korkeakoulu, Sähköteknillinen osasto	57
Jaakko Hyvärinen	1981	Doping and electrooptical characteristics of ZnS:Mn thin film deposited by alternating surface reactions [in Finnish]	Teknillinen korkeakoulu	
Marja Hamilo	1983	Tutkimuksia epäpuhtauksista Atomic Layer Epitaxy -menetelmällä valmistetuissa elektroluminenssi ohutkalvorakenteissa.	Teknillinen korkeakoulu, Kemian osasto	101
Aimo Rautiainen	1987	Atomikerroksittain tapahtuva ohutkalvojen kasvatusmenetelmä	Joensuun yliopisto	
Heini Mölsä	1991	Tutkimuksia suprajohtavan YBa2Cu3O(7-x)-ohutkalvon valmistamiseksi ALE-menetelmällä	Teknillinen korkeakoulu, Prosessi- ja materiaalitekniikan osasto, Epäorgaaninen kemia	96
Markku Sopanen	1991	ALE-menetelmällä valmistettujen kadmiumtelluridiohutkalvojen fotoluminesenssi	Teknillinen korkeakoulu, Tietotekniikan osasto, Fysiikka	45
Jutta Laine-Ylijoki	1992	Haihtuvien yhdisteiden tutkiminen ALE-prosessia varten	Teknillinen korkeakoulu, Prosessi- ja materiaalitekniikan osasto, Epäorgaaninen kemia	
Vesa Lujala	1992	Scale-up studies on the preparation of heterogeneous catalysts by Atomic Layer Epitaxy	Teknillinen korkeakoulu, Prosessi- ja materiaalitekniikan osasto, Epäorgaaninen kemia	86
Janne Rautanen	1993	Lyijysulfidiohutkalvojen valmistaminen ALE-menetelmällä ja karakterisointi IR-sovellutuksia varten	Teknillinen korkeakoulu, Prosessi- ja materiaalitekniikan osasto, Epäorgaaninen kemia	142
Kai-Erik Elers	1994	Ti-, Zr- ja Hf-alkoksidit	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	53
Arja Hakuli	1994	FTIR gas analysis in n-butane dehydrogenation studies	Teknillinen korkeakoulu, Prosessi- ja materiaalitekniikan osasto	
Jouni Meriläinen	1995	Raman spectroscopy in Cr catalyst characterization	Helsingin yliopisto, Kemia, analyyttinen	90
Marika Juppo	1996	Kupariohutkalvojen kasvatus kemiallisesti kaasufaasista	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	92
Diana Riihelä	1996	Chemical vapor deposition of AlN, GaN and InN thin films	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	105
Hannele Sala	1996	Ceriumin kemiallisen tilan määrittäminen SrS:Ce-ohutkalvoissa röntgenfotoelektronispektrometrialla	Teknillinen korkeakoulu, Prosessi- ja materiaalitekniikan osasto, Epäorgaaninen kemia	95
Juha Sarlund	1996	The surface treatments of CdTe thin films	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	71
Hilkkaliisa Siro	1996	Oxidative carbonylation of methanol on heterogeneous copper catalysts	Teknillinen korkeakoulu, Kemian tekniikan osasto, Kem-40 (Chemical Engineering)	85
Kirsi Vasama	1996	Aluminum, gallium and indium β-diketonates in their use in growth of thin films	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	50
Timo Hatanpää	1997	The volatile complexes of alkaline earth and rare earth metals	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	112
Jani Karttunen	1997	Amorfisesta piistä valmistetun aurinkopaneelin absorption lisääminen	Teknillinen korkeakoulu, Materiaali- ja kalliotekniikan osasto, Metalli- ja materiaalioppi	66
Putkonen, Matti	1997	Deposition of magnesium oxide thin films by atomic layer epitaxy	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	91
Mervi Jussila	1998	The binding of Yttrium and Other Rare Earth Beta-Diketonate Complexes on Silica and Their Application to Thin Film Growth	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	101
Riikka Puurunen	1998	Processing of aluminium nitride on porous silica by atomic layer epitaxy	Teknillinen korkeakoulu, Kemian tekniikan osasto, Teknillinen kemia	129
Ville Saanila	1998	Preparation of volatile precursors via gas-solid reactions	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	96
Petra Alen	1999	Titaani(III)- ja Ta(III)-kompleksit ja niiden haihtuvuus	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	104
Eeva Kallio	1999	Electrical characterization of thin ALCVD grown high K oxides	Teknillinen korkeakoulu, Sähkö- ja tietoliikennetekniikan osasto, Elektronifysiikka	71
Pia Kalsi	1999	Deposition of tantalum nitride thin films by CVD methods	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	85
Veli-Pekka Koivunen	1999	Developing the product structure and manufacture of a thin film reactor	Teknillinen korkeakoulu, Konetekniikan osasto, Koneensuunnitteluoppi	88
Jarmo Koskenala	1999	Tantalum Hafnium Oxide Capacitor	Teknillinen korkeakoulu, Materiaali- ja kalliotekniikan osasto	96
Henrik Leskinen	1999	Developing the productional structure of a small thin film reactor	Teknillinen korkeakoulu, Konetekniikan osasto, Koneensuunnitteluoppi	76
Antti Niskanen	1999	Jännitykset, niiden synty ja poistaminen kaasufaasimenetelmillä kasvatetuissa ohutkalvoissa	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	110
Anita Palukka	1999	Oksidikatalyyttimateriaalien happamuusominaisuuksien määrittäminen spektroskooppisin ja termoanalyyttisin menetelmin	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, Analyyttinen kemia	116
Kirsi-Marja Teppo	1999	Mangaanin levitys ZnS-ohutkalvolle ALE-prosessissa	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	121
Juhana Kostamo	2000	CuS:lla päällystetyn huokoisen piin valmistus sensorisovelluksia varten	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	110
Jaakko Niinistö	2001	Uusien Y- ja La-lähdeaineiden soveltuus ALE-kasvatuksiin	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	101
Huotari, Hannu	2002	Materials in metal electrode applications: properties of atomic layer deposited titanium nitride thin films	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	128
Laitinen, Otto	2002	Deposition of aluminium, hafnium and zirconium oxide thin films for gate oxide applications	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	102
Lang, Teemu	2002	Atomikerroskasvatuksen optimointi optisten interferenssisuodattimien valmistuksessa.	Teknillinen korkeakoulu, Sähkö- ja tietoliikennetekniikan osasto, Optoelektroniikka	71
Kaasinen, Sanna	2003	Growth of epitaxial oxide thin films on silicon by atomic layer deposition	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	83
Markus Lautala	2003	Chemical treatments of hydrogen terminated silicon surfaces for the deposition of gate oxides for transistors	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	86
Viljami Pore	2003	Self-cleaning photocatalytic titanium dioxide surfaces	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	112
Färm, Elina	2004	Itsejärjestäytyvien monokerrosten valmistus selektiivistä ALD:tä varten	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	100
Heikkilä, Mikko	2004	Sensitizing of Photoconducting Type PbS and PbSe Infrared Detectors	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	101
Hämäläinen, Jani	2004	Passiviset ohutkalvointerferenssisuodattimet DWDM-aallonpituusjaotinteknologiassa	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	100
Knapas, Kjell	2004	Alkoholien reaktiot metallioksidien pinnoilla	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	60
Mattila, Elina	2004	Deposition of organic layers on silicon dioxide surfaces using gas-solid phase reaktions.	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	87
Salmio, Hanna	2004	Atomic Layer Deposition as a Coating Method for Plastic and Metal Parts .	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	84
Harju, Maija	2005	Uusien mangaanilähdeaineiden soveltuvuus oksidiohutkalvojen atomikerroskasvatukseen	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	83
Malm, Jari	2005	Vacuum line chemistry in selected atomic layer deposition processes.	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	74
Martikainen, Juhani	2005	Ohutkalvopaineanturin kehittäminen	Teknillinen korkeakoulu, Sähkö- ja tietoliikennetekniikan osasto, Elektronifysiikka	70
Ralli, Kirsi-Leena	2005	High-k dielectric thin films on germanium.	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	102
Heli Vuori	2005	Preparation of noble metal catalysts by atomic layer deposition	Teknillinen korkeakoulu, Kemian tekniikan osasto, Teknillinen kemia	73
Teemu Alaranta	2007	Optimization of the dielectric properties of Ta2O5 thin films prepared by the ALD method	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	88
Bosund, Markus	2007	Passivation of GaAs Surface Using Atomic Layer Deposition.	Teknillinen korkeakoulu, Sähkö- ja tietoliikennetekniikan osasto, Optoelektroniikka	71
Ollikainen, Antti	2007	Atomic layer deposition and characterization of BiFeO3 thin films.	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	84
Sahramo, Elina	2007	(ZnO-ohutkalvojen atomikerroskasvatus funktionaalisille pinnoille)	Teknillinen korkeakoulu, Kemian tekniikan osasto, Epäorgaaninen kemia	76
Salmi, Leo	2007	Er2O3 in thin films and optical fibers	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	58
Xu, Tingmo	2007	Investigation of conductive ZnO/Al2O3 nanolaminates grown by atomic layer deposition.	Teknillinen korkeakoulu, Sähkö- ja tietoliikennetekniikan osasto, Optiikka ja molekyylimateriaalit	49
Härkönen, Emma	2008	Preparation of through porous structures of silicon and aluminum oxides	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	93
Kivelä, Tiina	2008	Potential controlled surface phenomena in microchip analysis	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	93
Li, Shuo	2008	Optimization of precursor pulsing in atomic layer deposition.	Teknillinen korkeakoulu, Mikro- ja nanotekniikan laitos, Optiikka ja molekyylimateriaalit	54
Ranki, Hannu	2008	Kuumennuksen vaikutus korkean permittiivisyyden ohutkalvoihin: Tutkimuksia korkean lämpötilan röntgendiffraktometrillä sekä atomivoimamikroskoopilla	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	88
Hashmi, Syed Ghufran	2009	Sub-wavelength coatings on metals by ALD.	Teknillinen korkeakoulu, Mikro- ja nanotekniikan laitos, Optiikka ja molekyylimateriaalit	60
Malinen, Ville	2009	Coating of stainless steel and aluminum by atomic layer deposition	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	57
Nykänen, Harri	2009	Design and fabrication of multi-pixel micro-LED array	Teknillinen korkeakoulu, Mikro- ja nanotekniikan laitos, Optoelektroniikka	66
Poutiainen, Juho	2009	Designing the vacuum chamber of ALD reactor.	Teknillinen korkeakoulu, Koneenrakennustekniikan laitos, Koneensuunnitteluoppi	76
Kariniemi, Maarit	2010	Atomikerroskasvatusprosessien kehittäminen titaanioksidi- ja strontiumtitanaattiohutkalvojen valmistamiseen	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	113
Klonner, Maria	2010	Stamp Fabrication for ultraviolet Nanoimprint Lithography.	Aalto-yliopisto, Elektroniikan, tietoliikenteen ja automaation tiedekunta, Optoelektroniikka	64
Mäntymäki, Miia	2010	Litiumin kaksoismetallialkoksidien valmistus ja käyttö ohutkalvojen lähdeaineena	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	160
Riuttanen, Lauri	2010	Thermal Annealing of AlN Thin Films Fabricated by Plasma-Enhanced Atomic Layer Deposition for GaN Epitaxy.	Aalto-yliopisto, Elektroniikan, tietoliikenteen ja automaation tiedekunta, Optoelektroniikka	55
Sarnet, Tiina	2010	Faasimuutosmateriaalien atomikerroskasvatus	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	125
Shah, Syed Ali Raza	2010	Buried channel technology for chemical microsystems.	Diplomityö, Elektroniikan, tietoliikenteen ja automaation tiedekunta, Optiikka ja molekyylimateriaalit	68
Sundberg, Pia	2010	Deposition of inorganic-organic hybrid thin films by ALD and MLD.	Aalto-yliopisto, Kemian ja materiaalitieteiden tiedekunta, Epäorgaaninen kemia	64
Viitanen, Timo	2010	Growth of thermoelectric Calcium Cobalt Oxide thin films.	Aalto-yliopisto, Kemian ja materiaalitieteiden tiedekunta, Epäorgaaninen kemia	79
Haggrén, Tuomas	2011	Growth of gallium arsenide nanowires on silicon	Aalto-yliopisto, Mikro- ja nanotekniikan laitos, Optoelektroniikka	71
Kauppi, Emilia	2011	Alucone and alumina thin films grown on bio-based packaging materials by atomic layer deposition and molecular layer deposition techniques.	Aalto-yliopisto, Puunjalostustekniikan laitos, Epäorgaaninen kemia	70
Mäkinen, Maija	2011	Protective MnCo2O4 coatings on ferritic stainless steel solid oxide fuel cell interconnects by atomic layerdeposition.	Aalto-yliopisto, Kemian laitos, Epäorgaaninen kemia	89
Seppälä, Sanni	2011	Gadoliniumoksidia sisältävät ohutkalvot	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	90
Su, Jie	2011	Optimization of TiO2 Thin Film Growth at Different Temperatures by Atomic Layer Deposition.	Aalto-yliopisto, Mikro- ja nanotekniikan laitos, Optoelektroniikka	52
Aittakari, Jari	2012	Water vapor transmission ratio measurement for the atomic layer deposited moisture barrier thin films on flexible substrates.	Aalto-yliopisto, Puunjalostustekniikan laitos, Puunjalostuksen kemia	79
Ali, Saima	2012	Characterization of atomic layer deposition films by x-ray scattering and atomic force microscopy.	Aalto-yliopisto, Mikro- ja nanotekniikan laitos, Optoelektroniikka	51
Holopainen, Jani	2012	Luun kasvun tukirakennemateriaalien valmistus sähkökehräyksellä ja atomikerroskasvatuksella	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	119
Midili, Virginio	2012	Realization of a capacitance-voltage measurement system for semiconductor characterization	Mikro- ja nanotekniikan laitos, Elektronifysiikka	71
Nisula, Mikko	2012	Effects of atomic layer deposited aluminum oxide coating on lithium iron phosphate.	Aalto-yliopisto, Kemian laitos, Epäorgaaninen kemia	93
Ristimäki, Ville	2012	Atomic layer deposition of dense mixed ionic-electronic conducting membrane on porous tubular substrate for oxygen combustion.	Aalto-yliopisto, Koneenrakennustekniikan laitos, Lämpötekniikka ja koneoppi	75
Ruoho, Mikko	2012	Nanostructured thermoelectric materials	Aalto-yliopisto, Mikro- ja nanotekniikan laitos, Optiikka ja molekyylimateriaalit	41
Kuitunen, Juha	2013	Kohdistettu ionisuihku mikro- ja nanorakenteiden valmistustyökaluna	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	54
Lapinsuo, Sakari	2013	Thin film coatings by atomic layer deposition for improvement of the flexural strength of glass.	Aalto-yliopisto, Mikro- ja nanotekniikan laitos	63
Saarno, Oskar	2013	Deposition and characterization of TiO2:Ti-ODA hybrid nanolaminate thin films	Aalto-yliopisto, Kemian laitos, Epäorgaaninen kemia	76
Venäläinen, Annika	2013	Nanocluster Depositions on Surfaces	University of Helsinki, Department of Physics	53
Kaipio, Mikko	2014	Ex situ in vacuo and in situ studies on mechanisms of ALD processes	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	120
Mikkola, Jyrki	2014	Scaling up MnCo2O4 Atomic Layer Deposition Process for Fuel Cell Applications.	Aalto-yliopisto, Kemian tekniikan korkeakoulu, Kemian tekniikan koulutusohjelma	83
Popov, Georgi	2014	Perovskite ternary halides in solar cell applications	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	120
Rontu, Ville	2014	Atomic layer deposition of niobium nitride thin films	Aalto University, School of Chemical Technology, Degree Programme of Materials Science and Engineering	70
Virtanen, Lotta	2014	Hopean passivointi	Helsingin yliopisto, Matemaattis-luonnontieteellinen tiedekunta, epäorgaaninen kemia	129
Weckman, Timo	2014	A density functional study of the TMA/H2O ALD-process.	Aalto-yliopisto, Kemian tekniikan korkeakoulu	83

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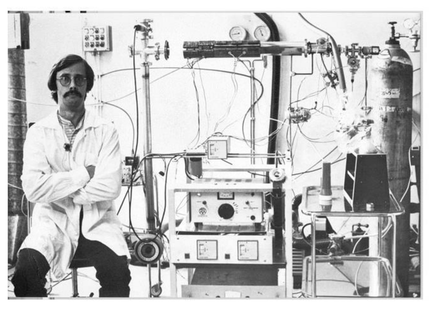

40 Years of ALD in Finland – Photos, Stories

Exhibition by the Finnish Centre of Excellence in Atomic Layer Deposition

Organizations currently active on ALD in Finland

Aalto University
www.aalto.fi
- Departrment of Chemistry, Laboratory of Inorganic Chemistry, Prof. Maarit Karppinen
  - Laboratory has been active in ALD since early 1980s
  - Currently the focus in ALD research is on novel hybrid inorganic-organic materials, superlattices and nanolaminates plus complex oxide materials
  - Potential applications range from thermoelectrics, superconductors and Li-ion batteries to various protective/barrier coatings
  - Research carried out with four ALD reactors (1 Picosun R-100 plus 3 ASM Microchemistry F-120)
- Department of Micro- and Nanosciences Prof. Harri Lipsanen:
  - Founded 2010, active in ALD since 2005
  - Focus in ALD for applications in nanoelectronics, solar cells and sensors
  - Aalto houses three ALD reactors in Micronova (2 Beneq TFS-500, 1 Picosun R-200) and characterization equipment
  - Department of Micro- and Nanosciences has 6 ALD researchers
  - Located in Micronova, Tietotie 3, 02150 Espoo
- Department of Biotechnology and Chemical Technology, prof. Juha Lehtonen and M.Sc. Emma Sairanen,
  - ALD for catalyst preparation for carbon nanotube growth, for fuel cell applications and for other chemical conversions.
- Chemistry Department, prof. Kari Laasonen and M. Sc. Timo Weckman, quantum mechanical modelling of ALD process

aldpulse.com
www.aldpulse.com
- ALD Pulse, Home of the ALD Community,
- Created in Finland and launched in April 2013, is a forum made to collect and provide easy access to useful, high quality information from around the world to the ever growing community of researchers, professors, scientists and thin film enthusiasts.
- Being the first and only website entirely dedicated to ALD, it has quickly become popular within the ALD Community, as it collects, shares and publishes without any bias, information about Applications, Vendors, Laboratories, Links to publications, Images, Events and many other interesting ALD topics.
- One of the unique features of the site, are the exclusive interviews which are part of a YouTube channel rich with ALD content which truly enhances the visitors' experience.
- ALD Pulse is highly active in, and empowered by mainstream Social Media.

ASM Microchemistry Oy
www.asm.com
- Founded 1987 as Mikrokemia Oy by Dr. Tuomo Suntola, the developer of atomic layer epitaxy (ALE).
- From the beginning, ALD research and process development have been the core activities along with studying ALD reactor concepts for industrial applications. The first projects were related to developing solar cell technologies using ALE, accompanied with additional projects, for example to study the feasibility of ALE in making heterogeneous catalysts.
- In 1999 ASM international N.V. acquired Microchemistry Oy a pioneer of ALD in the semiconductor industry, and invested further in research and development of equipment and processes for enabling the reliable and efficient use of ALD technology by advanced semiconductor chip manufacturers.
- Today ASM Microchemistry Oy concentrates solely on new ALD chemistry development projects for producing innovative and high-quality thin film processes and materials for semiconductor industry.
- Research and development laboratories in Finland are located at the Kumpula campus of University of Helsinki enabling a close long-term co-operation with the University.
- ASM Microchemistry Oy team consists presently of about 15 scientists and engineers with long-term experience in ALD.

Beneq Oy
www.beneq.com
- Founded 2005, active in ALD since 2005
- Beneq is a leading player in thin film coating equipment. Our product and service range covers thin film production and research equipment, comprehensive contract coating services and thin film electroluminescent displays, both standard and transparent. Beneq also boasts the world's largest ALD-dedicated production facility, and with it its immense coating capacity.
- Beneq's mission is the same as the company slogan: Turning Innovations into Success. With this statement, Beneq wants to highlight its role as an integral part of its customers business, their value-chain and development activities. It is crucial for us to accomplish benefit and success for our customers through our own innovations, and thus help them forward in their research and business.:
- Beneq develops and sells ALD coating equipment for research and industrial production. Our machine offering covers a vast array of processes, applications and ALD disciplines, including single substrate, batch, large scale (both substrate size and substrate amount), continuous ALD, Roll-to-Roll ALD, Plasma-Enhanced ALD and much more.
- Our personnel represents a committed and efficient team of seasoned professionals. We know ALD, all 140 of us.
- Located in: Finland, Germany, U.S.A., Russia and China, plus approximately 30 agents and 20 distributors worldwide.

Finnish SpecialGlass Oy
www.finnishspecialglass.fi
- Founded 1974, active in ALD from the beginning.
- Finnish SpecialGlass is a family-owned company continuing in the centuries-old tradition of glassblowing craftsmanship to the most exacting standards. Our expertise includes processing of ceramic parts and optical and special glass products, laboratory glassblowing, and product development carried out jointly with the client. We manufacture all special products according to the client's own design and specification, from individual prototypes through to large series. Top quality tools, raw materials and skilled craftsmanship guarantee work of high precision.
- Our clients consist of companies operating in various branches of industry, ranging from the largest concerns to small machine workshops and specialist manufacturers. Those relying on our expertise include institutes of higher education, universities, high-technology companies and the car, paper and outsourcing industries.
- Located in Paasikuja 3, 02330 Espoo, Finland

Lappeenranta University of Technology
www.lut.fi
- Founded 1969, active in ALD since 2004
- ALD work of LUT is carried out by ASTRaL research group, which belongs to LUT Savo Sustainable technologies unit. The mission of ASTRaL is to develop the leading edge materials technologies that are essential for finding innovative solutions to the growing materials problems of the world. The collaboration with industry enables this knowledge to be applied to the development of innovative high value products and lower cost production solutions while at the same time reducing their environmental impact.
- LUT has many years experience in ALD and other thin film deposition techniques. It has been in the forefront of development especially in spatial and roll-to-roll ALD processing. LUT has considerable experience in low-temperature deposition on polymers and in detailed characterisation of coating structure and properties. The target applications for the studies include e.g. spatial ALD process development, diffusion barriers for OLEDs, PVs and packaging, semiconducting and conducting layers, and photocatalytic layers.
- ALD equipment: Beneq TFS 500 batch ALD reactor with plasma activation capability, Beneq TFS 200 batch ALD reactor with plasma activation capability, Beneq TFS 200R continuous ALD reactor which acts as a test-bed for the true roll-to-roll ALD system, Beneq WCS 500 roll-to-roll ALD system
- Personnel: Currently 6 researchers and 3 research assistants
- Located in LUT Savo Sustainable Technologies, Sammonkatu 12, 50130 Mikkeli, Finland

Nanobakers Oy
http://www.nanobakers.com/
- Nanobakers Oy provides consulting services specializing in atomic layer deposition and optical thin films.
- Nanobakers also offers custom vacuum tool design and building services, and develops low-cost ALD research systems.

Oconco Oy
- Consulting services based on 30+ years of personal experience in ALD
- Olli Jylhä olli.jylha@oconco.fi

Picosun Oy
www.picosun.com
- Founded 1997, in ALD business since 2004
- Provides the leading quality ALD thin film coating solutions for global industries and R&D alike. Mission: to enable the industrial leap into the future by innovative, groundbreaking ALD thin film coating solutions
- Unique in the field:
  - Pioneering ALD experience for four decades – Dr. Tuomo Suntola, the inventor and original patentee of the ALD method in 1974 is one of Picosun Board Members. Exclusive concentration on ALD since the beginning – at the moment, 15th ALD tool generation on the market
- Fast-growing company (personnel almost doubled in only three years), has been profitable for several years in a row. Core team consists of long-term experts and Ph.D's in ALD. Employs ca. 40 people at the moment and several tens of subcontractors
- Headquarters in Espoo, Finland, production facilities in Kirkkonummi, Finland, US subsidiary Picosun USA in Detroit, Michigan, Asian subsidiary Picosun Asia in Singapore – recently also founded a new subsidiary in China

University of Eastern Finland (Joensuu)
www.uef.fi

University of Helsinki
www.helsinki.fi
- Founded 1640, active in ALD since 1990
- Laboratory of Inorganic Chemistry, http://www.helsinki.fi/kemia/epaorgaaninen/
  - Laboratory of Inorganic Chemistry (LIC) led by Markku Leskelä and Mikko Ritala is the world leading developer of new chemistry and processes for ALD. The ALD research is focused on materials needed in microelectronics, optics and nanotechnology. LIC is coordinating the Finnish Centre of Excellence in ALD (2012-2017) (http://www.aldcoe.fi/).
  - Nine ALD reactors: seven ASM Microchemistry F-120 from which one is equipped with QMS and QCM for in situ reaction mechanism studies, one Picosun R150 and one Beneq TFS-200 equipped with plasma source. LIC has facilities for precursor synthesis and characterision (XRD, NMR, MS, TGA, FTIR) and for characterization of thin films: XRD and XRR, both with a high temperature option; FESEM-EDX; FIB/SEM dual beam microscope with EDX, nanomanipulator, heater stage, and environmental (low vacuum) mode; SPM (AFM/STM with many variations); UV-Vis and FTIR; electrical measurements (I-V, C-V, surface resistance, polarization).
  - Personnel: 2 professors, 2 university lecturers, 3 senior scientists, 3 post docs, 13 PhD students, 4 MSc students
  - Address: A.I. Virtasenaukio 1, 00560 Helsinki
- University of Helsinki, Department of Physics, Division of Materials Physics, Ion beam analysis laboratory, http://www.physics.helsinki.fi/tutkimus/mat/english/research/ionbeam,
- Active in ALD sample characterizations since 1995.
- Focus on physical processes taking place in solid matter during and after irradiation of energetic ions.
- Equipment for ion beam based characterization and modification of ALD grown structures.
- Personnel (2 professors, 2 university lecturers, 5 post docs, 4 postdoctoral and undergraduate students, technical personnel)
- Address: Gustaf Hällströmin katu 2, 00560 Helsinki

University of Jyväskylä
www.jyu.fi
- Active in ALD with an own reactor since 2013: Joint activity of NanoScience Center (NSC) and Accelerator Laboratory. Contact person: Dr. Timo Sajavaara (timo.sajavaara@jyu.fi, www.jyu.fi/accelerator/abasedmat)
- Mission: To serve research groups of physics, chemistry and biology within JYU. Cooperation with local, national and international scientific and industrial partners. Examples of ongoing activities: Wettability studies of polymers coated by ALD, ion track structures, superconducting thin films, impurity studies of low-temperature ALD processes.
- Speciality: Characterization of ALD films using ion beam analysis techniques.
- Equipment: Beneq TFS 200 with plasma option and loadlock. Wafers and 3D samples possible. 3 liquid, 2 solid and 1 thermal / 2 plasma reactive gas sources. Situated in the NSC cleanroom. Accelerator-based elemental analyses (TOF-ERDA, RBS and PIXE) readily available, access to XRD, XRR, Raman, Ellipsometry, SEM, TEM, AFM, stylus profilometry etc.
- Research group: 2 seniors, 2 post docs (one in ALD development), 4 PhD students, several MSc students
- Street address: Survontie 9, Fi-40500 Jyväskylä, FINLAND

Volatec Ltd
www.volatec.fi
- Mission: Most appropriate precursors for each application of ALD technology
- Founded in 1993
- Provides the high quality ALD source materials together with different services for global research groups and industry
- Core team consists of long-term experts and Ph.D's in ALD
- Headquarter and production facilities in Porvoo, Finland - Volatec Ltd, Koneistajantie 2B, 06450 Porvoo

VTT Technical Research Centre of Finland
www.vtt.fi
- Founded in 1942. A not for profit research organisation.
- VTT produces research services that enhance the international competitiveness of companies. Our focus lies on applied research, integration of ALD processes, and use of ALD films in new applications and devices.
- ALD activities started initially in 1984-1985. Activities discontinued in the 1990s were re-initiated in 2004. Since 2012, VTT is part of the Finnish Centre of Excellence on Atomic Layer Deposition, coordinated by the University of Helsinki (www.aldcoe.fi).
- Equipment: VTT has two own ALD reactors: Picosun™ R-150 single-wafer reactor (Micronova clean room) and Picosun™ R-200 semi-batch reactor. The Micronova clean room facility (www.micronova.fi), which is a collaboration of VTT and Aalto University, is fully equipped with process and characterization tools and belongs to the National Research Infrastructure.
- Personnel: 2900 as of 31.12.2013.
- Headquarters: Tekniikantie 4A, Otaniemi, Espoo. Micronova: Tietotie 3, Espoo.

About the exhibition

40 year ago, the atomic layer deposition (ALD) technique was independently invented in Finland, and Tuomo Suntola and Jorma Antson filed their famous patent application on Atomic Layer Epitaxy, FIN 52359, on November 29, 1974.

Celebrating the round years, the Finnish Centre of Excellence on Atomic Layer Deposition, led by professor Markku Leskelä of the University of Helsinki, is organizing an exhibition: "40 years of ALD in Finland: photos, stories". The exhibition is organized concurrently with the international Baltic ALD conference, which takes place on May 12-13, Helsinki, Finland (www.aldcoe.fi/bald2014). After Baltic ALD, the exhibition material can be viewed for a few weeks at the Chemistry Department of the University of Helsinki (A.I. Virtasenaukio 1, Helsinki) as well as the VTT Technical Research Centre of Finland, Micronova building (Tietotie 3, Espoo).

The main organizers of the exhibition have been by Dr. Riikka Puurunen (VTT) and Dr. Jaakko Niinistö (University of Helsinki). Riikka and Jaakko would like to thank a large number of people, who have kindly helped by providing information or material for the exhibition or assisting otherwise with the organization:

Prof. Jouni Ahopelto, VTT
Dr. Tapani Alasaarela, Nanobakers Oy
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Mr. Timo Asikainen, ASM Microchemistry Oy
Prof. David Cameron, Miktech Oy
Dr. Satu Ek, Picosun Oy
Dr. Kestutis Grigoras, VTT
Dr. Suvi Haukka, ASM Microchemistry Oy
Ms. Emma Härkönen, University of Helsinki
Mr. Jaakko Hyvärinen, Endeas Oy
Mr. Kari Härkönen, Beneq Products Oy
Mr. Olli Jylhä, Oconco Oy
Dr. Jaana Kanervo, Aalto University
Prof. Maarit Karppinen, Aalto University
Prof. Hannu Kattelus, VTT Memsfab Ltd.
Mr. Väinö Kilpi, Picosun Oy
Mr. Juhana Kostamo, Picosun Oy
Prof. Outi Krause, Aalto University
Dr. Marja-Leena Kääriäinen, University of Colorado
Dr. Tommi Kääriäinen, University of Colorado
Prof. Kari Laasonen, Aalto University
Dr. Kimmo Lahtinen, Lappeenranta University of Technology
Ms. Riitta Laitinen, VTT
Prof. Juha Lehtonen, Aalto University
Mr. Tuomo Latopelto, Finnish SpecialGlass Oy
Prof. Markku Leskelä, University of Helsinki
Prof. Harri Lipsanen, Aalto University
Ms. Jaana Marles, VTT
Mr. Jarmo Maula, Beneq Oy
Dr. Philippe Monnoyer, VTT
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Prof. Tapani Pakkanen, University of Eastern Finland
Mr. Joe Pimenoff, Beneq Oy
Dr. Matti Putkonen, VTT
Dr. Antti Rahtu, Vaisala
Prof. Robin Ras, Aalto University
Prof. Mikko Ritala, University of Helsinki
Prof. Jyrki Räisänen, University of Helsinki
Prof. Tapio Salmi, Åbo Akademi
Mr. Jarmo Skarp
Mr. Esa Tanskanen, VTT
Dr. Timo Sajavaara, University of Jyväskylä
Mr. Pekka Soininen, Beneq Oy
Ms. Tuija Soininen, VTT
Dr. Jonas Sundqvist, Fraunhofer.IPMS-CNT, Germany
Dr. Tuomo Suntola, Picosun Oy
Dr. Marja Tiitta, Volatec Ltd.
Ms. Annina Titoff, aldpulse.com
Prof. Ilkka Tittonen, Aalto University
Dr. Minna Toivola, Picosun Oy
Dr. Marko Tuominen, ASM Microchemistry Oy
Dr. Markku Ylilammi, VTT

Tuomo Suntola's ALE in Short

By Riikka Puurunen, VTT Technical Research Centre of Finland, Espoo, Finland

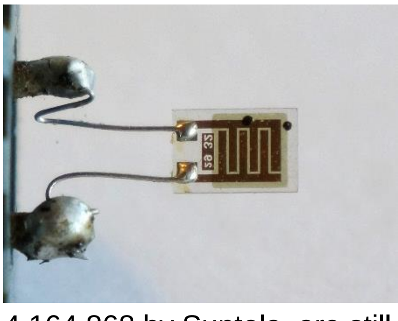
Prologue

Tuomo Suntola was born in 1943 in Tampere. He went to school in Turku and completed his Matriculation examinations (nation-wide examinations, which qualify to university studies) in 1962.

In 1963, Suntola started his studies on electrical engineering at Helsinki University of Technology (HUT - Teknillinen korkeakoulu, TKK). In 1967, he completed his Master's thesis ("diplomityö" in Finnish) on Schottky diodes for radar detectors (metal-semiconductor diodes for microwave frequencies).

In the late 1960s, picosecond "Ovonic" switches using amorphous chalcogenide compound thin films were a hot research topic. Being an assistant teacher at HUT, Suntola proposed to Prof Thor Stubb to investigate the picosecond switches more closely. Prof. Stubb gave him green light for the study. After a few years thorough experimental and theoretical work Suntola was able to find out the mechanism behind the switching phenomenon. Prof Stubb saw that the solution was worth a doctoral thesis. (In short, the mechanism which was found is that the current concentrates into a thin filament, where the temperature rises above 700°C, which makes the material highly conductive. For once a real postulate was defended in the thesis: one cannot expect wide technical use of the amorphous switches. In a quick schedule, Tuomo Suntola finished a licentiate's thesis and a doctoral thesis on the topic. In December 1971, Tuomo Suntola, at the age of 28, became the youngest Doctor of Science in Technology from the Electrical Engineering department by that time.

In the early 1970's, semiconductor research was still young in Finland. VTT Semiconductor Laboratory had been established in 1964 as an integral part of Professor Stubb's Electron Physics laboratory in the HUT. In the autumn of 1971, the Semiconductor Laboratory received its first industrial order, a task to develop a miniature solid-state humidity sensor for Vaisala Oy, a Finnish company specializing in meteorological instrumentation. Prof Stubb assigned the challenge to Suntola a few months before Suntola had finished his thesis. To get off to a quick start, his colleague Tapio Wiik was assigned to make a pre-study on the state of the art in humidity sensing.



According to the pre-study, the most promising solutions demonstrated were capacitive devices made of porous aluminum oxide thin films or polymer foils. Suntola started to work on polymers; instead of using foils, he dissolved the polymer material and created a thin film on a solid substrate by vaporizing the solvent. The thin polymer film approach turned out to be successful and he completed the development with Jorma Antson by the end of 1973. Vaisala's instruments using the Humicap sensor, based on the original patent 1972-11-12/1979-08-21, US 4,164,868 by Suntola, are still (2014) the world market leaders in humidity measurements. Picture: A prototype of the Humicap humidity sensor from 1973. The sensor was fully functional after 41 years when photographed in 2014.

Since the late 1960s, beside his main work on technology Suntola became interested in the basis of physics, including antique metaphysics, eastern philosophy and the philosophy of science in general. Suntola was also an active lecturer for INSKO, Insinöörijärjestöjen Koulutuskeskus, giving lectures for example on display technologies. All these, and especially the very successful humidity sensor, turned out to have important impact on Suntola's later career.

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1974 - Initiation of the EL development and the invention of ALE

Late in 1973, Suntola was contacted by the top management of Instrumentarium Oy. At that time, Instrumentarium, originally a trading house importing medical instrumentation, was looking for possibilities for making their own products. They had established a small technology company Datex Oy that had introduced a novel bed scale for weighing dialysis patients and a CO_2/CO meter for anesthesia control. When the key technical team of Datex left the company in late 1973, Suntola was invited to establish a research group directly under the management of Instrumentarium – with an open request to "suggest and find out something". Such an unprejudiced suggestion was inspired by the success of the humidity sensor, already known to some industrialists.

In the beginning of 1974, Suntola and the small humidity sensor team moved from VTT to Instrumentarium (Ahertajantie 3, Tapiola, Espoo). First, Suntola made a market need/technology mapping study. The outcome from the mapping was that, in the interest area of Instrumentarium, sensor technologies are diversified into small unities, which makes it difficult to build a technology platform on such a basis. At the other end of an instrument the human interface, typically a display, is in common for most instruments. There was an urgent need for flat panel displays which fit e.g. in the beds of the patients and in small hospital rooms.

Suntola's proposal to Instrumentarium's management was to start working for two goals, for ion-selective sensors and for flat panel display. That was quite abstract for the management, the CEO's comment after Suntola's presentation was: "I am still confused, but at a higher level". Anyway, Suntola got the approval to go on. Jorma Antson started the study on sensors and Suntola directed himself to solve the flat panel objectives.

A literature study on the state of the art in flat panel displays showed, that electroluminescence had been tried in many ways with limited success. Suntola concluded that successful manufacturing of thin film EL devices cannot be solved with conventional thin film technologies like sputtering or vacuum evaporation. Based on his prior experience with amorphous thin films, Suntola reasoned that to get controlled properties, one has to have well controlled crystal structure in the thin films. To obtain a controlled material structure, "deposition", or material transfer, may not be sufficient – in principal, what is needed is to create conditions for controlled buildup of the material layer.

The actual invention of ALE matured in early June 1974, Suntola describes it as follows:

"We had still an empty laboratory with just tables and chairs and a Periodic Table of the Elements hanging on the wall. Looking at the Periodic Table, and thinking of the overall symmetry in nature, to me came the idea of "serving" the complementary elements of a compound sequentially, one at a time, onto a surface. Monoatomic layers may be obtained if the complementary elements make a stronger bond with each other than they do with their own kind of atoms".

Suntola decided to proceed in this direction. Once the basic idea of supplying elements in an alternated, cyclic manner to the surface had been conceived, active planning started. From the kinetic theory of gases, Suntola calculated the pressures and temperatures needed for the growth of zinc sulphide, the light emitting material in a thin film electroluminescent display. He designed the equipment for testing the growth hypothesis applying standard vacuum components; a 12" T-piece formed the chamber − installations were built on the flanges on the top and the bottom of the T-piece. The design was completed in June 1974 and the drawings were sent to the in-house mechanical workshop.

The equipment operated at a low pressure of about 10^{-6} Torr. The sources for elemental Zn and S, reasonably isolated from the vacuum environment, were heated to obtain the desired vapor pressures. The equipment can be classified as a hybrid ALE-PVD system: due to its low vapor pressure, the Mn dopant was added by evaporation from an open source. (The option of evaporation and sputtering was long retained in the high vacuum reactors for "partial ALE" of oxides). The first reactor had a window in the top flange, which facilitated following the development of the thickness during the process by observing interference colors on the glass substrate.

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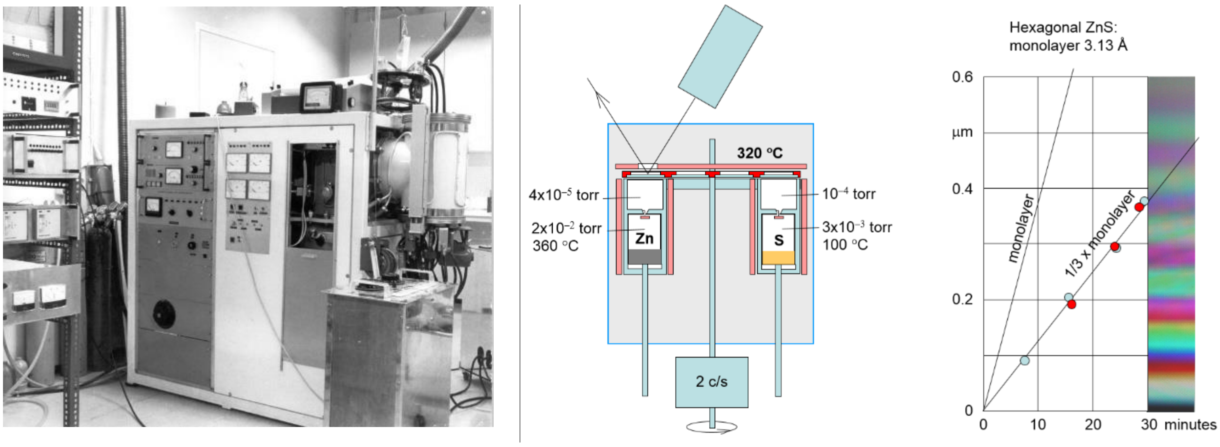

Pictures: The ALE-reactor was built in the T-piece on right of the pumping unit. It consisted of a rotating carousel holding the substrates, and the sources for the reactants. Windows in the top flange allowed observation of the interference color of the film building up in the course of the process. By comparing the color to an interference color map the increase of the thickness could be monitored during the process.

The first growth experiments were made in August-September 1974. Cycle time in the rotating carousel reactor was 2 cycles/sec. The very first experiment was successful; a film was observed to grow – not, however at the expected rate, one monolayer in a cycle, but only a third of a monolayer in a cycle. It took quite some time before it was understood – how the surface rearrangement accounted for the less-than-monolayer growth.

The films were analyzed by x-ray diffraction, and first of course, by look and feel. The first impression was that the films were mechanically harder than films made by conventional methods (evaporation or sputtering). The x-ray studies showed that the ALE-ZnS had hexagonal crystalline structure, in contrast to evaporated or sputtered ZnS films that had cubic crystalline structure. It was also noticed that the films were hydrophobic: a water droplet rolled on them. (Interestingly, the films made later from ZnCl2/H2S were, in contrast, hydrophilic: a water droplet did not roll on them.)

Early ZnS(Mn) samples showed faint electroluminescent light with direct DC excitation. The level of Mn doping was 0.1-1%. The indium tin oxide transparent electrodes were made in the carousel reactor by "semi-ALE" by sputtering the metal components.

Tuomo Suntola decided to call the new growth technology "Atomic Layer Epitaxy". Epitaxy is Greek, and means "on-arrangement". It was meant to emphasize the "surface control" feature of the process – to make a distinction with traditional "source controlled" deposition methods like evaporation and sputtering.

The ALE invention was protected by international ALE patent. The priority date of the patent comes from the Finnish application, filed on November 29, 1974. The patent was applied and granted in more than twenty countries, including the United States, Japan, and the Soviet Union. In the study of the prior art, the closest reference found was a German patent from mid-50s, describing vacuum evaporation of compound materials from the components of the compound in carousel system, much like ALE in the first reactor. The essential feature of ALE, the saturation of the surface reactions was not, however, claimed in the patent.

Hearings related to the patent were organized in several countries, including The United States, Japan, and The Soviet Union. The patent hearing in Moscow in 1977 or 1978 Suntola describes as "memorable", "ikimuistoinen". Suntola went there alone. Two local attorneys, well acquainted to the application, assisted him to defend to work. The hearing took about two hours. The discussion was mainly focused on the generality and broadness of the claims. No critical prior art, neither from the

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USSR nor from other countries, was presented by the examiners. The patent was granted as applied.

In hindsight, in 2014, Tuomo Suntola concludes that the 1974 patent was successfully formulated: it covered the basic ALE concept well. In fact, no infringements were reported during the whole lifetime of the patent. Suntola thinks that the main reason for that was that ALE was regarded as too slow and complex for any practical use. The first patent focused on ALE from elemental reactants, however, the option of using compound reactants (H_2S) was mentioned, which slightly limited the formulation of the second important patent applied in early 1979.

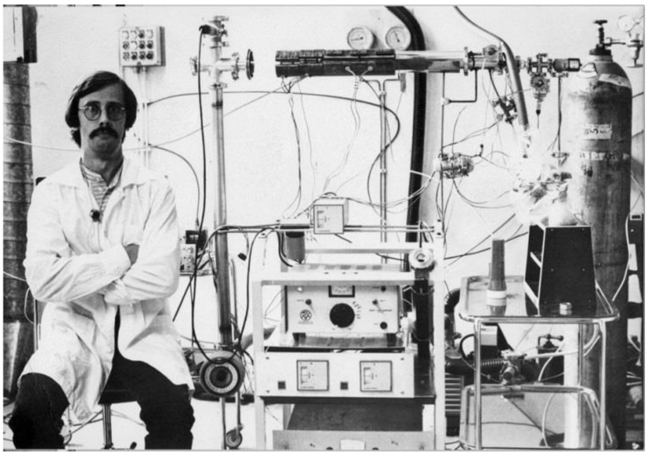
Steps towards practical processing of EL displays

The performance of the early EL demos made with high-vacuum reactors were far from sufficient. Also, it became obvious, that a high-vacuum system would not be production-worthy; the source had to be in line-of-sight to the substrate, and up-scaling would be limited. Also, the use of chemically aggressive materials possibly needed in "full-ALE" oxides in a high vacuum reactor was dubious. The ALE-process itself was encouraging, but it was understood that there was still a long way to go. It was realized that the basic work needed had limited motivation in the small research team in a medical instrumentation trading company. In 1978, the ALE-EL project with the personnel and the facility was sold to the building material company Lohja Oy. Lohja had recently started the production of TV-sets and was looking for opportunities for further growth in electronics.

The transfer to Lohja allowed a substantial increase in the resources for the development. One of the new tasks triggered was a study of the possible replacement of the high-vacuum equipment with more compact flow-type reactors allowing the use of volatile reactants and a batch type arrangement of the substrates effectively utilizing the self-control feature of ALE.

Suntola recalls that the decision to switch from elemental reactants to compound reactants was not easy. "We did not have the necessary understanding of the chemistry, also, we did not have experience in working with aggressive chemicals such as chlorides and H_2S." First, a simple glass tube reactor was constructed for the experiments. The very first experiment for a chloride process was tried by combining $ZnCl_2$ and elemental S, then by adding H_2 flow over the heated sulphur source. The experiments were not successful.

An abrupt change occurred when a H_2S gas bottle was acquired and coupled to the reactor thus enabling the real $ZnCl_2$ + H_2S process. In the memorable first $ZnCl_2$ + H_2S run the walls of the reactor tube were evenly covered with a strictly uniform film – with a "starting profile" essentially steeper than in the ZnS processes based on elemental reactants – thus telling about the high reactivity of the reactants.



The famous picture of Sven Lindfors sitting next to the glass tube ALD reactor was taken after the first successful $ZnCl_2/H_2S$ process – what is missing in the picture was Arto Pakkala and his first reaction to the success: "Siinä se on!" ("That's it!") He was right: the ALE-EL development was immediately switched to chloride processes, and to the design of a new generation of inert gas reactor.

The chloride processes for the ZnS and the dielectrics allowed the buildup of the whole EL-structure, dielectric – ZnS(Mn) – dielectric, in one process. Proper choice of chlorides allowed the running of the process at a constant temperature. The temperature limits, however, were tight – the upper limit was set by the soda glass used as the substrate and the lower limit by the vapor pressure of the manganese chloride. Sodium diffusion from the substrate was stopped by an ALE Al_2O_3 diffusion barrier.

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In the high vacuum reactors the separation of the ALE sequences was based on the motion of the substrates between the fixed sources. In an inert gas reactor there was a need for high-speed valves capable of valving aggressive gases for thousands of times at the source temperatures. The chemically inert high speed valving problem was solved by "inert gas valving" based on controllable blocking and transporting of the inert gas flows between the sources and the reaction chamber. The new reactors allowed uninterrupted processing of the whole EL-structure comprising the stack of the first dielectric layer – zinc sulphide(Mn) layer – the second dielectric layer. The production capability of the inert gas reactor was based on batch processing and the inherent self-control feature of ALE. Processing of a full EL structure was typically an overnight process without human oversight in the facility. A batch of 20-30 pieces of 100x200 mm^2 substrates were stacked back-to-back in a reaction chamber of a few liters only.

The inert gas reactor and the basic processes for the ALE-EL displays were protected by the 2nd international ALE patent, with the priority date Feb 28, 1979 of the Finnish application. Also, the ALE dielectric layers are documented in the patent.

The dielectric strength of the ALE dielectrics, above 4 MV/cm, (nowadays close to 10 MV/cm) was observed to be far better than ever reported for conventionally produced thin films. In fact, it turned out that the role of ALE was even more important for the properties of the dielectrics than for the light emitting ZnS(Mn) layer in the EL display.

ALE-EL displays showed outstanding electrical and optical characteristics. The high dielectric strength of the dielectrics, with an intrinsic pinhole-free feature, allowed effective excitation of the light emitting ZnS(Mn) layer. Due to the hexagonal crystalline structure of the ALE-ZnS, the color of the light emitted was beautiful yellow, it was less orange than the EL devices based on sputtered ZnS with cubic crystalline structure. The fully transparent structure allowed a black background layer resulting in an outstanding contrast also in bright ambient light. Picture: Arto Pakkala operates reactors used for early ALE-EL prototypes.

Later, the single oxide dielectrics were replaced by multilayer oxides such as an aluminum-titanium oxide nanolaminate (ATO). In the ATO, the evolution of crystallinity in the Al_2O_3 layer is interrupted by amorphous TiO_2 films. In the flow-reactor, the doping of the ZnS layer with manganese was carried out by supplying manganese chloride with every 50th zinc chloride pulse.

1980 - The first public disclosure of TF-EL display

In the 1970s, the ALE-EL development was carried out strictly confidentially – until the first disclosure at the Society for Information Display (SID) conference in San Diego, California, in April 29 to May 1, 1980. There were about 1000 people present. The presentation of the revolutionary EL display was given by an unknown person (Suntola), coming from an unknown country (Finland). In addition to the overhead slides, Suntola had an EL demonstration box with him. The demo had such a high contrast that the EL light was well visible to the audience even in the bright projector light. The presentation was a great success – Suntola had not realized before, how much their EL technology was ahead of all the competitors.

Picture: SID representative and Tuomo Suntola and his key colleagues, Jorma Antson, Sven Lindfors and Arto Pakkala (from the left to the right) received the 1980 SID Outstanding Paper Award for the EL work. The Award was handed to the group in the next year SID conference in 1981.

After the SID presentation, Lohja got about 3000 to 4000 requests for products. Two persons were hired

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to go through the requests. "What a tragedy, wasted marketing", says Suntola – "we had neither the production line constructed nor the product developed". The demand for flat panel displays, however, was confirmed.

Commercialization of ALE-EL

In 1983, the pilot production of TF-EL displays started in Lohja, Kunnarla plant.

The first real proof-of-concept of the ALE-EL displays was the large information boards installed in the departure hall at Helsinki-Vantaa airport in 1983.

Final testing of the alternatives for dielectric layers in the thin film structure – Al_2O_3, ATO (Aluminum-Tantalum Oxide nano-laminate) or Ta_2O_5 – was carried out in a prototype board installed in an underground cave under the airport. Test modules with ATO dielectrics showed the best performance and reliability.

In fact, the reliability of the panels turned out to be outstanding: in the final installations in the departure halls, the large information boards were running continuously for 15 years, day and night, without a single character module replaced.

Picture: Ralf Graeffe inspects the display modules in the test assembly in airport underground cave in 1983.

Year 1983 was financially favorable for Lohja. Also, there was much EL enthusiasm in the air. The same year, the Finnish government offered remarkable tax reductions for production investments completed by summer 1984. Lohja utilized the opportunity and a large EL production facility with high level cleanrooms was built in Espoo, Olarinluoma. The schedule was extremely challenging, but the building was finished in time. Picture: Olarinluoma plant in 2014; EL-production continues, operated by Beneq Oy since 2012.

The schedule for the construction of the production machines and the final development of the main product, a data display module with integrated high voltage drivers, was at least as challenging as the new building. The production of the data modules started gradually in the Olarinluoma facility in 1985 and distribution channels were opened in the USA and in some other major industrialized countries. Lohja's electronics activities were reorganized from a research unit to a business unit. The Display Electronics division got an experienced business oriented manager, Antti Piippo, who was invited to Lohja from Aspo Oy.

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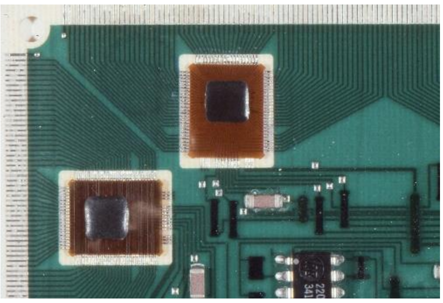
Suntola continued as the Chief Scientist with a mandate to look for new application areas for the ALE technology. In the picture: Tuomo Suntola (right) and Ulf Ström, the managing director of Finlux Inc. responsible for marketing the ALE-EL panels in the USA.

The technologically unique achievement had created wide publicity to the EL project in mid 1980s in Finland – which, together with the big investments into the development and production had piled up enormous expectations and pressures on its commercial success. Following Sharp in Japan and Planar in the US, Lohja became the third EL manufacturer in the world. Lohja's product was superior, but the startup of the production in Olarinluoma was delayed and the growth of the EL business was slower than expected.

In 1990, Lohja's EL business was sold to its American competitor Planar Inc. The Finnish EL-activity, Planar International, became a subsidiary of the US Planar. ALE showed its superiority in the manufacturing of the EL panels; in a few years all EL production of Planar was concentrated to the Olarinluoma plant. Gradually, other flat panel display technologies, first of all liquid crystal technology supplemented with thin film transistor active matrix, took over from the EL technology which suffered from the high driving voltage, about 200 VAC, and the lack of full color capability. EL displays remained a specialized device for demanding environments such as extreme temperatures or exposure to high mechanical shocks. In 2012, Planar's EL plant in Olarinluoma was acquired by Beneq Oy, who continues the production of the ALE-EL panels as the only commercial manufacturer in the world (2014).



The major developments made on the driving circuitries and the associated manufacturing technology for the EL panels under Ahti Aintila had created unique knowhow and experience, such as advanced fine-line printed circuit board technology and Tape Automated Bonding (TAB) of integrated circuits. When EL-manufacturing was sold to Planar Inc. in 1990, the microelectronics division of Lohja was separated as an independent company Elcoteq Oy. Antti Piippo and his closest colleagues bought Elcoteq in 1991. Elcoteq was an important contract manufacturer e.g. to Nokia during the fast growth of the mobile phone markets. The structural changes in the electronics industry led to the bankruptcy of Elcoteq in 2011. Picture: A corner of the EL panel circuit board with high voltage IC drivers.

After leaving Lohja microelectronics division in late 1984, Ahti Aintila worked for several companies developing further microelectronics production technologies. In 1991, Aintila established Picopack Oy for producing smart cards and other products utilizing the TAB technology.

1987 - 1998 Microchemistry Ltd

In the mid-1980's the national oil company Neste Oy had established a corporate venture unit, Neste Advanced Power Systems (NAPS), to study and develop business possibilities in the emerging field of renewable energy solutions. Based on the success of ALE in the EL technology in Lohja, the head of the venture team, Tapio Alvesalo, saw the technological potential of applying ALE for the manufacturing of thin film solar panels, and suggested research activity on this topic in Neste. In 1987, Tuomo Suntola was offered the chance to set up Microchemistry Ltd, as a subsidiary of Neste, for the development of ALE-based solar panels. Suntola saw also the possibility of applying ALE to heterogeneous catalyst, which complemented the objectives decided for the new research unit.

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After a pre-study, the ALD solar panel research was directed towards cells based on CdTe thin films. Lohja's EL production reactors were considered too big and slow for effective solar cell research. As the first task in Microchemistry, Tuomo Suntola and Sven Lindfors designed a quick, compact research reactor for the PV development. The outcome was the F-120 reactor which also became the first commercial ALD reactor. The F-120 was optimized for fast cycling and easy operation and maintenance.

Picture: The F-120 reactor. The heavy bars on each side of the reactor were meant to let the researcher lean and ponder while looking at the progress of the process.

The work on CdTe thin film cells brought with it a lot of expertise and insight in photovoltaics, i.e. not only in CdTe cells but more broadly in PV technologies and renewable energy systems in general. In 1990, NAPS acquired a factory in France producing solar panels based on amorphous silicon (a-Si) thin films. Microchemistry got the challenge of working on the development of the production process and the improvement of the low conversion efficiency (3 to 4 %) of the a-Si panels. Some improvement in the initial efficiencies of the panels was obtained. It turned out, however, that the increase in the efficiencies was lost in half a year's use of the panel. A useful improvement, however, was the reaction injection molding techniques used for encapsulating the a-Si modules. As the market segment of a-Si panels remained very limited NAPS withdrew from the French factory and the a-Si technology in 1997.

The top achievement in the ALE-CdTe-cell development in Microchemistry was a test cell with 14% conversion efficiency, which in the early 1990s was one among the highest efficiencies reported for thin film cells. The advantage of ALE on the CdTe-cell, however, was marginal over the less expensive, robust technologies and therefore a decision was made not to enter into production scale with ALE-CdTe cells.

One of the succesful side paths in the PV research in Microchemistry was the development of new testing techniques for the PV panels. Jaakko Hyvärinen constructed a fast and handy solar simulator that measured IV-characteristics of the panel in response to a single flash of a xenon lamp. The method and the equipment turned out to be a success; after the PV-activity in Microchemistry and NAPS, Jaakko Hyvärinen established Endeas Oy http://www.endeas.fi/ to produce and sell the "Quicksun" simulators.

The work on ALE-catalysts brought highly desired chemistry expertise into Microchemistry. Aimo Rautiainen and Eeva-Liisa Lakomaa were the first chemists in the team. Several ALD catalysts were successfully demonstrated, and importantly, the work had major impact on the understanding of the surface chemistry in the ALE process. Several doctoral dissertations were made: Marina Lindblad (1992), Suvi Haukka (1993), Arla Kytökivi (1997). Co-operation with the experienced catalyst researchers in Neste Central Research laboratories was initially hampered by the barrier between traditional methods and the novel ALE approach which turned thinking from moles to molecules. Once the initial suspicions about ALE were overcome, the co-operation became very fruitful and ALE also served as an effective tool for studying the catalytic surfaces at atomic level.

Microchemistry focuses on ALE

When Microchemistry Ltd was established in 1987, the ownership was divided between Neste and Lohja as 80% / 20%. Lohja's ALE-patents were licensed to Microchemistry for PV and catalyst applications only. When Lohja's EL-activity was sold to Planar in 1990, Neste bought Lohja's share of Microchemistry, and soon the ALE license was extended to cover the manufacturing and sales of ALE-reactors and the ALE technology for any use not covered by Lohja's EL patents.

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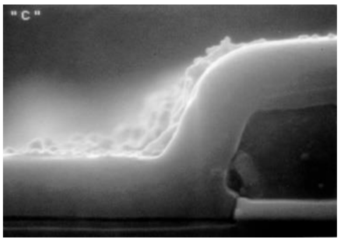
Suntola had seen the large potential of ALE from the very beginning of the invention, especially in semiconductor manufacturing. The knowhow in the ALE process and the reactors, not disclosed in the patents, had been kept strictly confidential throughout the development and commercialization of the EL-technology. The new situation offered Microchemistry the possibility to start working on ALE and the reactors for new applications.

As a first step, the F-120 was offered for research purposes. Next step was the design of the F-450 reactor which enabled processing of silicon wafers, and glass substrates up to 450x450 mm^2 size. Samples of 8" silicon wafers with aluminum oxide layer were processed in an F-450 prototype, and computer-generated pictures of the commercial version were produced to a product brochure and a full size exhibition booth wall. The new product was covered with a new patent portfolio, now assigned to Microchemistry.

Since the SID conference in the 1980, and from numerous other conferences and meetings, Suntola had contacts with key people in the display and semiconductor fields. Thanks to the confidence acquired with ALE-EL Suntola was offered the possibility for a visible introduction of ALE in the MRS 1994 Annual Meeting in Boston. The talk "ALE for Semiconductor Applications" was complemented by Microchemistry's ALE-booth in the exhibition held parallel to the conference. In the picture (left) Heli Vaara, assistant to Tuomo Suntola (right).



The Boston MRS presentation demonstrated the excellent uniformity and conformal coating characteristics of ALE technology. Based on the measurements of the refractive index, the high density of the Al_2O_3 films was also introduced. Actual 8" wafers were shown at the exhibition booth. Picture: Demonstration of the conformality of the ALE-Al_2O_3 passivation layer at the edge of a contact pad in an integrated circuit.

Gradually, major semiconductor manufacturers and equipment manufacturers became interested in the technology. The uniformity, conformal coating and the highly repeatable material characteristics obtained with ALE were more and more acknowledged and appreciated. There were still doubts of the production efficiency of the process. The main doubts, however, were cast over the unknown equipment manufacturer from an unknown country. When a representative of a major semiconductor manufacturer expressed their interest to the ALE reactor, he stated that, in general, they do not do business with companies with the annual turnover less than 100 million dollars.

Introduction of ALE technology and the Microchemistry reactors was continued in several exhibitions in the US, Europe and Japan. In addition, samples were delivered and Suntola gave presentations in many universities and research institutes. Gradually, ALE became a subject in materials research conferences and specific ALE conferences were organized. The increased activity generated new demonstrations of the special material properties obtained by ALE.

Obviously, ALE reactors and business with semiconductor manufacturers could not be considered as a natural business area in an oil company. This was also realized by the real actors in the field; Microchemistry was approached by several major semiconductor equipment manufacturers with proposals of co-operation or acquisition.

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In 1997, as a part of a larger rearrangement in Neste Oy, Microchemistry was devoted to ALE technology and the reactor business only, and the PV- and catalyst research activities were taken under Neste's Central Research. Suntola was offered a Research Fellow's position in Neste, and a business oriented managing director, Matti Ervasti, was invited to take care of Microchemistry and the further ALE business development.

At that time there was already a clear indication of the commercial potential of ALE. Microchemistry had sales representatives in the US, Japan and South Korea and several reactors had been delivered to customers. The most impressive contract consisted of several large ALE reactors capable of handling big batches of 500x500 mm^2 substrates (picture).

The original name of the process Atomic Layer Epitaxy met continuous resistance among semiconductor people who had devoted the term epitaxy for the meaning of single crystal growth only. The practical choice was to accept the reality and change the process name Atomic Layer Epitaxy, ALE into Atomic Layer Deposition, ALD.

In 1999, Microchemistry with all its personnel, ALE expertise and the patent portfolio was sold to the Dutch company ASM. The ALE activity continued as ASM Microchemistry in Finland. After a few years, ASM moved the ALE reactor manufacturing to their plant in Phoenix, Arizona. ASM's activity in Finland was focused on ALE research, at present in a close connection to the Inorganic Chemistry Laboratory of the University of Helsinki. As a consequence, highly experienced reactor designers were freed. Sven Lindfors, the closest coworker to Suntola in ALE reactor design since 1975 set up Picosun Oy with Kustaa Poutiainen in 2004. In 2005, Beneq Oy was founded and Pekka Soininen started the ALE reactor activity in the company.

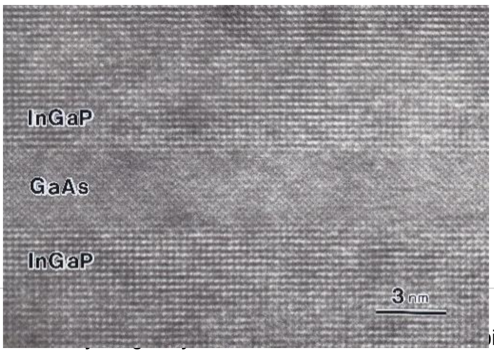
Spreading of ALE by year 1990

ALE in Japan

After the successful SID conference, Suntola was invited to give a presentation on ALE at the International Conference on Vapor Growth and Epitaxy, 5 in San Diego, California in 1981. The presentation was not a big success; there was very little scientific information available on the ALE grown material. Furthermore, the use of the term "epitaxy" for non-single-crystal thin films offended the "silicon guys" in the audience.

An outcome from the 1981 San Diego conference, however, was that the seeds were laid for ALE research in Japan. Professor Jun-ichi Nishizawa from Japan was among the participants, and he realized the significance of ALE. Nishizawa started to investigate the possibility of ALE growth of GaAs in his laboratory in Japan.

Nishizawa introduced his MLE-GaAs in the 16^{th}, (1984 International) Conference on Solid State Devices and Materials, in Kobe, Japan, 1984. Suntola was invited to give a general talk on ALE in the conference. The conference was grand, Suntola gave the talk to a room overfull of people. Also, Japanese TV was present and Suntola was interviewed about ALE. Professor Nishizawa initially renamed ALE as "Molecular Layer Epitaxy" (MLE), thereby making a distinction with Suntola's original work. Ten years later he admitted that it was one and the same technique.



Suntola recalls that the Japanese III-V activity in mid 1980s was the most visible research effort on ALE. One of the most impressive demonstrations, introduced by Nippon Electric Company (NEC), was the InGaP-GaAs-InGaP superlattice, where the GaAs layer is obtained by applying 11 ALE cycles (the center layer in the picture).

...ition", has been published in the Chemical Vapor Deposition journal in a slightly modified version, http://onlinelibrary.wiley.com/doi/10.1002/cvde.201402012/abstract (Open Access)

Most of the Japanese works on the III-V were carried out in ultrahigh vacuum MBE equipment modified to operate in the ALE mode. In some experiments laser irradiation was used to enhance the process.

ALE in the USA

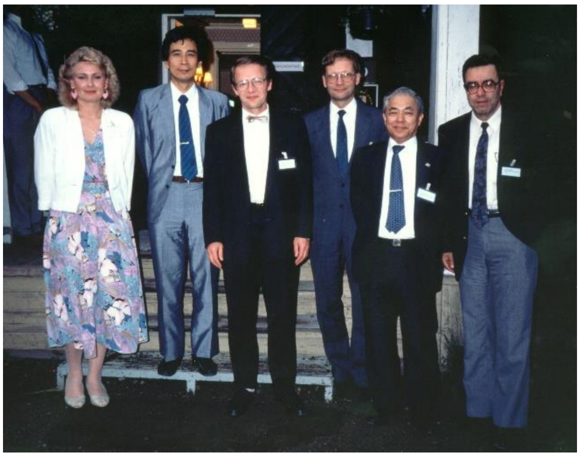
In 1980s professor Bedair and his group in North Carolina State University demonstrated ALE of GaAs in a CVD system, where sequencing of the reactants was obtained with a fast rotating disk. Prof. DenBaars and his group in the University of Southern California, Los Angeles, demonstrated ALD of GaAs in an atmospheric MOCVD system.

First ALE conferences in Finland

In 1984, the first specific ALE conference, the "First Symposium on Atomic Layer Epitaxy", VTT Symposium 54, was organized in Espoo, Finland, by VTT Semiconductor Laboratory upon the initiation by prof. Lauri Niinistö. In the late 1970s, Suntola had contacted Lauri Niinistö, the professor of Inorganic and Analytical chemistry at Helsinki University of Technology (HUT) asking for scientific consultation in ALE chemistry and luminescent materials. The challenge was well received by Prof. Niinistö, and a fruitful, long-lasting co-operation was started.

The preface of the proceedings of the First Symposium on Atomic Layer Epitaxy states:

"This proceedings contains the invited papers presented at the First Symposium on Atomic Layer Epitaxy, held on December 13-14, 1984 in Espoo. Most of the papers have been devoted to this novel growth method. Related growth techniques, methods of layer characterization and potential applications account for the rest of the content. The contributions represent the growing number of research groups active in this area in Finland...".



The first International Symposium on Atomic Layer Epitaxy was held in Espoo, June 11-13, 1990. The conference chair was Prof Lauri Niinistö.

In the picture, participants at the conference dinner in Herttoniemi, Helsinki, restaurant Vanha Mylly: Dr. Aoyagi, RIKEN, Tokyo Institute of Technology; Erja Nykänen, HUT; Tuomo Suntola, Microchemistry Ltd.; Prof. Niinistö, HUT; Prof. Nishizawa, Semiconductor Laboratory, Sendai; Prof. Bedair, North Carolina University.

ALE in Otaniemi, Helsinki and Joensuu

The strict confidentiality regarding the ALE reactors and the process knowhow resulted in severe limitations to the co-operation with universities. The first reactor supplied to the HUT in the early 1980s, to professor Niinistö's laboratory, was placed in a locked room accessible only to authorized operators and researchers. However, this early co-operation served as the start for extensive ALE activity in HUT and a seed to the presently vital ALE activities headed by professors Markku Leskelä and Mikko Ritala, as the Finnish Centre of Excellence in Atomic Layer Deposition, in the Laboratory of Inorganic Chemistry in the University of Helsinki.

ALE activities were initiated also in the Semiconductor Laboratory of VTT Technical Research Centre of Finland in mid-1980s. Initial focus was on III-V materials.

Theoretical work on ALE was free of the limitations created by the confidentiality of the technology. Fruitful co-operation with Professor Tapani Pakkanen in the University of Joensuu (Finland) resulted in a quantum chemical analysis of the ZnS growth, finally completed by Marina Lindblad in her doctoral thesis in 1992 in Microchemistry.

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Early ALE in Tampere

In 1973, before Suntola had started the work leading to ALE and EL, he was invited to give lectures on semiconductor physics in Tampere University of Technology (TUT). Suntola was nominated a docent in TUT, and he continued to give the lectures every second Friday for a couple of years. In 1975, after the early results in the EL project, Suntola encouraged the small TUT group to work on ALE of the III-V semiconductors. The challenge was taken by M. Ahonen, who at that time was a graduate student. In 1978, Markus Pessa was nominated the professor of physics in TUT. He took care of the lectures on semiconductor physics and directed a vital research activity on the III-Vs, using the conventional MBE technology. Pessa's work led to effective GaAs laser technology and successful industrial activities in optoelectronics.

Pioneering work in the Soviet Union

When Suntola, for his 1989 review paper, mapped known laboratories working on ALE, there was no information of the Soviet group that had been studying "Molecular Layering". In 1990, Dr. Victor Drozd contacted Suntola and invited him to visit the University of Leningrad (at present St. Petersburg) to meet Prof. Aleskovskii. Aleskovskii presented his works dealing with oxide layer buildup based on saturated surface reactions just as in the ALE process. The discussion confirmed that the essence of ALE had been independently discovered by the Aleskovskii group. Suntola was impressed by the deep understanding of the surface chemistry related to the molecular layer buildup. Samples or applications using Molecular Layering were not shown, neither was the equipment used for the experimentation. Since the visit, representatives of Aleskovskii's group participated in ALE conferences organized in Finland.

Little known episodes of ALE research

There are many "episodes" in the ALE-EL development that have remained more or less unknown.

ALE became limitedly known after Suntola's first public conference presentations. Among some experts in conventional thin film technologies ALE technology was seen as weird and most probably useless. A German professor, a well-known thin film expert, sent a letter to Suntola, where he proved, based on his experiments, that the ALE mechanism is "impossible". At this time, ALE-EL displays were already in pilot production, so, Suntola did not respond. Later, when Suntola met the professor at a conference, the doubts were settled.

In the early 1980s, the ALE-EL technology was licensed to France. First contacts were made at the SID conference in 1980. The big French state-owned company CGE became interested and contacted Lohja in order to discuss on a license for the technology. The discussions led to an agreement and an ALE-EL license was granted in 1983 (now to Sintra Alcatel due to local arrangements between French state owned companies). An ALE reactor capable of handling 500x500 mm^2 substrates was delivered to France as a part of the license arrangement.

Suntola tells that Lohja had a high level meeting in New York with a big American semiconductor manufacturer in 1982. Lohja offered the company an ALE license for semiconductor applications, possibly to be used for thin gate electrodes and for passivation layers. The advantages of ALE were told to be excellent thickness uniformity, controllable dielectric constant, extremely high dielectric strength and the conformal coating characteristic, already proven with the EL devices. It turned out that the advantages of ALE were not understood; the then existing technologies were seen to fulfill the needs. In early 1980s this was certainly true. The time was not yet right for ALE.

Also, it is interesting to know that in addition to the EL patents, Suntola had a patent on the Liquid Crystal Displays (LCD's). The patent "Method for generating electronically controllable color elements and color display based on the method," which was applied for in 1985 and granted in 1990 was meant to guarantee a technology for full color display for the case that full colors are not achieved with the EL. Suntola's concern about the color limitation with the EL became true, but the sequential color concept in Suntola's patent has not been taken into use yet.

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Epilogue

When handing Microchemistry Ltd. over to his successor Matti Ervasti in the beginning of 1998, Suntola left behind his active role in ALE, now renamed as ALD. Seeds had been sown, and the technology had matured ready for success.

As a research fellow in Neste, now in Fortum Oyj, after the fusion of the state oil company Neste and the electric utility company IVO, Suntola worked for long-term research and global energy issues with main emphasis on renewable energies. He was a member of World Energy Council and served as the attorney of Fortum Foundation until part-time retirement in 2000 and full retirement in the beginning of 2004. In the same year, Suntola joined Picosun Oy as the scientific adviser and a board member.

The year 2004 brought a delightful surprise to Suntola in the form of the European SEMI Award 2004 *"Honoring the Pioneer in Atomic Layer Deposition Techniques ... that paved the way for the development of nanoscale semiconductor devices"*.

The Award was handed to Suntola at the Munich Electronics Show 2004 by Stanley Myers, the President & CEO of SEMI (USA).

The founders of Picosun Oy, Sven Lindfors and Kustaa Poutiainen accompanied Suntola to Munich to participate in the Award Ceremony.

When receiving the award, Suntola expressed his acknowledgements to his many coworkers with whom he shares the honor brought by the Award. Also, he saw the Award as a recognition of the importance of the often invisible long-span efforts behind the progress of science and technology.

Back to the basis

In the biography on his www-page (http://www.sci.fi/~suntola/) Suntola states: "*Considerations of the philosophy of science and the foundations of physics have been a source of inspiration throughout my career, both in technological developments and the search of a holistic view of physical reality"*.

Since the mid-1990s, with early roots extending to the late 1960s, Suntola has worked on a holistic picture of reality published in 2011 in the book "The Dynamic Universe – toward a unified picture of physical reality". For linking his philosophical and theoretical considerations to the historical development, he worked on the book "The Short History of Science – or the long path to the union of metaphysics and empiricism", published in 2012. (http://www.physicsfoundations.org/).

As of 2014, Suntola continues as a board member of Picosun Oy, the chairman of the Physics Foundations Society and a board member and a frequent lecturer in the Finnish Society for Natural Philosophy.

Suntola reminds us that the success of ALE-ALD development became possible through the contribution of many highly competent and motivated people, and he wishes to express his gratitude to all of them—equally to those mentioned by name in this history, and to the big majority not mentioned. *"It looks like our long run led rather to a start than to a completion – I am deeply impressed by the huge advance that has followed in the ALD technology and the scope of its applications."*

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Acknowledgements

This history has been written in close collaboration with Dr. Tuomo Suntola. Tuomo: thank you for sharing with me—a youngster, born about at the same time as the first ALE patent was filed—these and other details of the development of ALE and EL. It has been a great honor and privilege to work with you and to write this up. Writing this history was triggered by the parallel-running worldwide Virtual Project on the History of ALD (VPHA). Warmest thanks, Tuomo, for your support to the VPHA, too. Thank you also for the discussions on your philosophy and your visions on science and technology. I have enjoyed and learned a lot from them.

This history has been written for the "40 years of ALD in Finland" –exhibition, to be organized during the International Baltic ALD conference (Helsinki, May 12-13, 2014). Thanks to Prof. David Cameron for polishing the English language. Funding by the Academy of Finland's Centre of Excellence in Atomic Layer Deposition (ALDCoE) is gratefully acknowledged.

About the author

Dr. Riikka Puurunen (born in 1974 in Pori, Finland) completed the matriculation examinations in Länsi-Porin Lukio in 1993. After an academic exchange year (1993-1994) in Beloit College, Wisconsin, USA, Riikka started studying Chemical Engineering at Helsinki University of Technology (HUT) in Espoo, Finland. In 1998, she obtained the degree of Master of Science and in 2002 Doctor of Science in Technology from HUT, majoring in Industrial Chemistry with professor Outi Krause. Riikka Puurunen has worked with the ALD technique through all her scientific career, first related to catalyst synthesis (1998-2002, first Microchemistry, then Neste Oil and Gas and Helsinki University of Technology, Finland), then for transistor gate oxide development (2003-2004, IMEC, Belgium), and currently for the development of microelectromechanical systems, MEMS (2004 on, VTT Technical Research Centre of Finland). Riikka knows Dr. Tuomo Suntola since 1998, and in 2010 they co-authored, with research professor Hannu Kattelus, a chapter on the ALD for MEMS. In 2007, Riikka received the Young Researcher of the Year award of Tekniikan Edistämissäätiö. Riikka's interest in the history of ALD dates back to her postdoctoral times at IMEC, when she wrote an overview of ALD including notions on the history, published in Applied Physics Reviews in 2005. Currently, the ALD history work continues as an open collaborative worldwide initiative called the Virtual Project on the History of ALD.

Picture: Riikka Puurunen, who has just arrived to start writing the story of ALE. Photo taken on March 6, 2014, by Tuomo Suntola.

Small correction made to the story, 25.11.2014, by Riikka Puurunen:
In the ALE-1 photo, second-left participant info updated

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Atomic layer deposition - Source materials enabling thin film depositions

author Marja Tiitta, Volatec Ltd[1]

Helsinki University of Technology, Laboratory of Inorganic Chemistry, head Professor Lauri Niinistö, had a pioneering group in the development of atomic layer deposition and its applications in the 1980s. The main focus of the research was on the applications, and depending on these, different source materials were chosen for thin film depositions. Nowadays, the group of professors Markku Leskelä and Mikko Ritala in the University of Helsinki take the lead in the academic development of novel atomic layer deposition precursors in Finland.

This short story describes some memories from 1987-1990 from the research work made on atomic layer deposition (ALD) from the view point of source materials in Helsinki University of Technology in the Laboratory of Inorganic Chemistry. The ALD researchers were Erja Nykänen and Pekka Soininen besides me. I luckily took over the position of Milja Mäkelä (former Asplund) who decided to live in Lahti. There were also several diploma workers in the group. Laboratory engineer Lasse Hiltunen was an essential person in very many equipment and facilities related questions. Lot of knowhow and a large network were created at this time which later on enabled the growth of ALD based industry in Finland and in other countries as well. With this story I would like to express my gratitude to professors Lauri Niinistö and Markku Leskelä for their important support in the beginning of my professional carrier.

ALD researchers of Helsinki University of Technology, Laboratory of Inorganic Chemistry in 1990, Erja Nykänen, Marja Lahonen (former Leppänen, diploma work), Pekka Soininen and Marja Tiitta.

[1]Volatec Ltd was established in 1993 to prepare source materials for atomic layer deposition needs from laboratory to industrial scale. Nowadays, the laboratory and production facilities are located in Porvoo, Finland, and customers are from all over the world.

Metal chlorides - standard chemicals for atomic layer deposition growth

Metal chlorides such as aluminum, zinc and titanium chloride were useful and commonly used precursors when aluminum, zinc or titanium based thin films were grown. They were, and are, cost effective precursors when corrosion does not need to be considered. From the young researcher's point of view in 1987, the only big issue with chloride based precursors was to fill the source material vessel fast enough to avoid too much vapor in the air and too much exposure to the lungs while loading the ALD reactor. Nowadays, we are able to fill ALD vessels with metal chlorides in an oxygen and moisture free environment. This avoids all the impurities from air in precursors. At this time, these items were not recognized to be important. We had, however, a research question - how to deposit thin films without chlorine impurity? This has led to synthesis and trialing of chlorine-free precursors also for aluminum, zinc and titanium. The other target in the study of alternatives for metal chlorides was to decrease the thin film growth temperature. This was not possible with aluminum and zinc chloride without loss of thin film quality.

Metal alkoxides - common industrial chemicals

Metal alkoxides were good candidates for developing chlorine free thin film depositions. Their preparations were also straight-forward, and different recipes were found in the literature. So, let's start with aluminium alkoxides. I prepared a set of the aluminium based alkoxides and titanium ethoxide, and we tested them in the film depositions. The preparation of metal alkoxides required at least a little skill in synthesis. In the preparation of aluminium ethoxide, the reaction was cooled with the ice, and a small addition of a catalyst to the mixture caused a highly exothermic reaction with shaking of the whole glass equipment which was built under a hood. I was lucky not to have my head in the hood when the reaction was initiated. At this time, I did not recognize the meaning of yields – these have greatly increased when working with moisture free conditions.

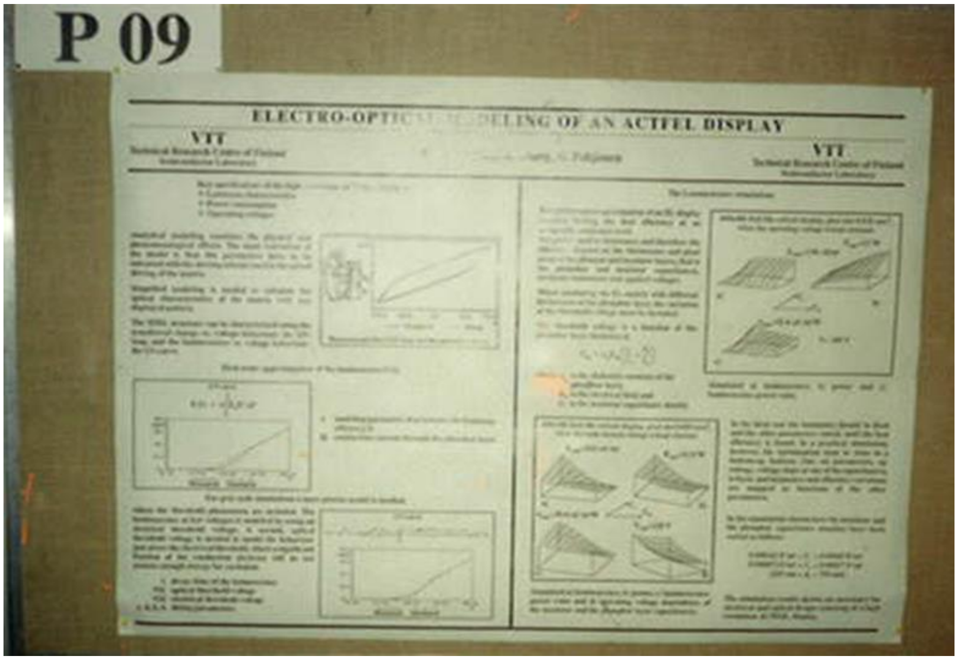
We made excellent thin films from the metal alkoxides but the problem with metal alkoxides was the narrow range of deposition conditions because of their tendency to decompose.

Metal beta-diketonates - volatile compounds known from gas chromatography applications

Metal beta diketonates, especially metal 2,2,6,6-tetramethyl-3,5-heptanedionates (thd) were used in the growth of films by atomic layer deposition for electroluminescent applications. These studies were started when Markku Tammenmaa was researcher in the ALD group of Helsinki University of Technology in the 1980s. Metal beta diketonates in general, and thd compounds in particular, were easy to prepare by precipitation reactions. All the lanthanides have volatile and relative highly thermally stable thd compounds. With the help of these compounds, the different colors (white, green, blue and red) for atomic layer deposition based electroluminescent device were prepared. The first pilot demonstration was in 1989 but unfortunately it was decided not to send it to that year's Amsterdam exhibition. The problem was not in the colors but in the break-through voltage of the devices. Instead of the demonstration, Erja, Pekka and I were sent to this event like overtime compensation.

Pekka and Marja are studying Philips' presentation of liquid crystal based flat displays. Erja has taken the photo.

Erja and Einstein. We visited together the waxwork museum in Amsterdam.



VTT presented their results in Amsterdam exhibition. Jaakko Hyvärinen from Lohja-Finlux is visiting the poster of VTT.

At this time, there was a break-through in science as new yttrium copper barium oxide based superconductors were invented. This also encouraged our group to make attempts to grow these structures by ALD. We got wonderful oxide layers from thd based materials but unfortunately this superconductive spinel structure has remained a challenge for the next generation of researchers.

When the synthesis work of metal beta diketonates had begun, I was the largest user of absolute ethanol in Helsinki University of Technology, Chemistry department at that time. Ethanol was the most common used solvent in the synthesis of metal complexes. I have brought several flasks (12 liters each) downstairs to the Inorganic Chemistry laboratory, and my colleagues have warned me not to do it just before 1st of May.

Metallocenes - high reactivity

Though metal beta diketonates were useful precursors, there was a wish to develop depositions with higher growth rate than obtained using them and also at low temperature. My first metallocene preparation was yttrium cyclopentadienyl. Since budgets were restricted, the first task was to discover if source materials for its synthesis were already available in the university. I got sodium string from the Organic department, and cyclopentadienyl was in storage. The recipe is easy: first distillation to get monomer, and then reaction with sodium to get sodium cyclopentadienyl. This sodium cyclopentadienyl was reacted with yttrium salt to get final product. Everything was fine, there was a nice product in the vessel but my problem was born in the cleaning of the reaction glass vessels. This was my first fire that I have caused in the laboratory, when unreacted sodium reacted with water. Fortunately, I used to make the first trials with small amounts and I did not cause too much damages.

Metal carbamates

We also developed lead sulphide films. There was a suspicion that there could be oxygen impurities in the sulphide films because of the beta diketonate precursor used in the depositions of lead sulphide film. We started to look for oxygen-free precursors for lead. Carbamates were recognized to be good possibilities. At this time, safety was important but not so important as it should have been. However, we did not choose lead alkyls but lead carbamates for precursor due to safety issues.

Metal alkyls

Metal alkyls were starting to be considered for use in atomic layer depositions in 90s. They have high reactivity and volatility but their disadvantage is in the precautions needed for their handling. No leakages are allowed in the equipment when metal alkyls are handled. Based on rumors, the use of metal alkyls has caused most of the dangerous situations compared with any other ALD source materials in the depositions of thin films.

Sulphide, oxide and nitride thin films and their precursors

The source material for sulphur of sulfide films was dihydrogen sulphide. We know that its smell is not very attractive. We developed a trap for dihydrogen sulphide for ALD equipment that was based on copper solutions. The laboratory personnel upstairs were happy about this innovation.

The most used source material for oxygen was water. There was a concern that water caused problems at the semiconductor-dielectric interface when deposited in the multilayer structures. The alternatives that we studied were alcohols. At this time, and especially when superconductive layers were developed, ozone was considered as an alternative, more reactive oxygen source to improve thin film properties. An ozoniser was bought and the ALD reactor system was equipped with the ozoniser by Heini Mölsä.

Ammonia was the source material for nitrogen when nitride films were deposited. Ammonia has also a strong smell and the fume hoods needed to be working properly when we prepared nitride films.

Characterization of source materials

Thermogravimetry was most common used method when different source materials for ALD were studied. This easily showed if the synthesis product has high enough volatility, and how much decomposition occurs under standardized conditions. Mass spectrometry was later used in conjunction with thermogravimetry and also made it possible to identify the vapor composition of precursors. This was the topic of the diploma work of Eija Koriseva.

We had X-ray diffraction (XRD) in our laboratory, and it was very much used for thin film characterizations but to some extent also for source material analysis. Mainly the crystalline impurities were identified, and with help of XRD, we developed the washing and drying procedures for ALD-precursors.

Infrared spectrometry was in the department of Physical Chemistry, and Gitte Härkönen taught me to also use IR for characterization of precursors and thin films.

Acknowledgment Prof David Cameron, Scientific Advisor in Miktech Oy, is thanked for the improvement of English.

Remembering Milja Mäkelä

By Marja-Leena Kääriäinen, University of Colorado

Dr. Milja Mäkelä (nee Asplund) was born 1954 in Äänekoski, Finland. She received her doctorate in chemistry at the University of Gothenburg in 1984. She was one of the forerunners in the field of ALD in Finland. In the spring of 2001 she became a founder member and a technology director for an ALD company called Nanoscale Oy. Nanoscale was started by a few Finnish companies; Neopoli Oy, Elmont Oy, Innoware Oy, and Microdata Oy. Nanoscale served institutions and other companies by providing ALD thin films and solutions for multiple, various applications. Right from the beginning Milja became the heart and the brain of Nanoscale. She always seemed to be ahead of the time with her ideas when others followed good five years after her. Hence also Nanoscale started at a time when the need for nanoscale thin films, outside semiconductor industry was just emerging. Nevertheless, Dr. Milja Mäkelä's endless enthusiasm and innovative touch throughout her work made "atomic layer deposition" a familiar set of words for numerous people. In 2008 Nanoscale Oy merged to Deep Sea Engineering Oy where Dr. Mäkelä served as a technology director. Dr. Milja Mäkelä was also a docent at the University of Helsinki, and served as a development manager in Mictech Oy. In addition she founded another ALD company Surfon Oy. In 2010 she moved to Beneq Oy where she worked as a senior scientist till 2013. Dr. Milja Mäkelä passed away in January 2013 due to a prolonged illness.

In the photo, Dr. Milja Mäkelä in 2007 in Nanoscale Oy, Lahti. Photo courtesy of Milja's sister Merja Kuuslampi.

Story on ZyALD

By Jonas Sundqvist, Österbottning, ex-Qimonda ALD Process engineer and precursor developer

In 2005 at 90 nm DRAM node Samsung introduced ALD deposited hafnium-based high-k dielectrics for the DRAM capacitor module and the rest of the industry soon followed. In the beginning alkyl amide precursors such as TEMAHf dominated the market. However, as the aspect ratio of the memory cell increased to gain more cell capacitance scaling down node by node it became clear that there was a need for a replacement of the rather thermally instable hafnium alkyle amides. At the same time the industry decided to switch to zirconium based dielectrics to gain a relatively higher k-value at a lower thermal budget. Unfortunately, the corresponding zirconium alkyle amide had an even lower thermal stability than their hafnium counterpart. Therefore, the DRAM companies pushed the ALD precursor suppliers to come up with more thermally stable, yet highly reactive volatile alternatives to the alkyl amides.

In 2007 the development had led to synthesis of novel monocyclopentadienyl zirconium precursors from world class research and development by Air Liquide Electronics and The Department of Chemistry at Helsinki University showing excellent ALD conformality at higher deposition temperatures than ever before. The first public presentation was at BALD2007 in Warsaw by Jaakko Niinistö and those results were later published in scientific journals [1, 2]. Based on these results, Qimonda, one of the DRAM companies, took a fast decision in 2008 to scale up one of the precursors to a 300 mm ALD process for ZrO_2-based dielectric. This work was done at Fraunhofer CNT in Dresden on a Jusung Eureka Single wafer ALD paltform. The first ever large sample (500 g) can be seen on the picture below. Later Qimonda and most DRAM companies went into full production using ZyALD™, which was the trade name given by Air Liquide Electronics for $ZrCp(NMe_2)_3$. Since then ZyALD™ has been used for a large part of the world wide DRAM production and possibly still today and must be regarded as one of the most successful ALD precursors for semiconductor applications.

Air Liquide Electronics reported on March 21st, 2013 that now also the Chinese Patents Office has granted a patent related to the application of ZyALD™ in semiconductor processing. This followed the recent granting of patents in several other states, including South Korea, Singapore, Taiwan and several European countries, with a favorable outcome also expected in various other jurisdictions. The usage of ZyALD™ and other similar molecules for high-k deposition is now covered by 11 granted patents worldwide and 13 additional pending applications.

[1] J. Niinistö, K. Kukli, M. Kariniemi, M. Ritala, M. Leskelä, N. Blasco, A. Pinchart, C. Lachaud, N. Laaroussi, Z. Wang, C. Dussarrat. Novel mixed alkylamido-cyclopentadienyl precursors for ALD of ZrO2 thin films, J. Mater. Chem. (2008) 18, 5243.
[2] K. Kukli, J. Niinisto, A. Tamm, M. Ritala, M. Leskela. Behavior of zirconium oxide films processed from novel monocyclopentadienyl precursors by atomic layer deposition, J. Vac. Sci. Technol. B 27, (2009) 226

The first ever large sample (500 g) of ZyALD^™ in 2008. Photo provided by Dr. Jonas Sundqvist who is in the photo himself.