

Argon recycling offers per wafer cost reduction opportunity

Dr Rob Grant FRSC founded GR2L with the objective of commercialising chemical looping combustion technology into the gas purification market. Here, he gives us an insight into the untapped potential of argon recycling.

In the production of silicon wafers to be fabricated into solar cells and micro-electronic devices high purity argon (better than 99.999%) is used to control the impurity levels present during the manufacturing process to an acceptable level. The trend in the solar industry has been to reduce the argon purge flows to a minimum, to keep the cost per wafer down; however this comes at the expense of wafer purity which results in lower performance solar cells. High purity wafers for use in micro-electronic applications typically

utilise two to three times as much argon as for the solar PV application.

The emerging trend in the solar PV market is to move to higher purity high performance and/or n-type doped wafers, to maximise the efficiency of the resulting solar cells. This will require argon purge gas flows to increase again and since the supply of high purity argon is essentially fixed, the laws of 'supply and demand' dictate that the argon price will increase. For manufacturers faced with this 'double



Dr Rob Grant



Figure 2. The Argon0 Point of Use Recycle System



Figure 1. A CZ furnace hall. Photograph courtesy of Norsun, Norway

whammy’ of increasing argon usage and cost, the appeal of argon recycle, which practically reduces the argon cost per wafer by a factor of ten, is becoming ever more attractive.

ArgonØ recycle strategies

A typical production facility will have many tens of furnaces located in a furnace hall, Figure 1, and in order to recycle the argon, it first needs to be recovered from the exhaust lines of the vacuum pumps associated with the individual furnaces, then purified and returned to the argon furnace purge supply line. There are two philosophies to the recycle of exhaust gases from process tools: Point of Use recycle where the exhaust gas is diverted to a local purification system, close coupled to a limited number of tools, say <10, and Central processing where all the facility exhaust lines, say >50, are combined together and fed to a single, larger, purification system.

The positives and negatives associated with such approaches are summarised in Table 1 opposite, but it is interesting to note

that in the microelectronics industry, faced with the similar problem of Point of Use or Centralised scrubbing, almost all fabrication facilities have adopted the Point of Use approach, largely for the flexibility that it offers.

There is a ‘common sense’ rule in gas recycle and that is to avoid adding any

extra gas/contamination after the recovery point, which then needs to be subsequently removed before returning the gas into the process again. Unfortunately the Centralised system approach breaks this rule by mixing the exhaust gas from furnace pump-down, which contains air, with the exhaust gas during process runs, which just contains the process contaminants.

Table 1. PoU versus Centralise argon recycle – positives and negatives

	PoU recycle	Centralised recycle
Footprint	Small – can be co-located with the process equipment	Very large – needs to be located in a separate building next to the process facility
Installation	Phased install with the process equipment, no doubling up of Ar supply or exhaust	Single one-off facility wide installation, must double up on Ar supply and exhaust lines
Retrofit?	Yes – Easy to retrofit	No – very difficult to retrofit
Need to remove Air?	No – exhaust gas from pump-down vented until air free, then diverted to recycle	Yes – exhaust gas from pump-down is mixed with the other air free exhaust gases
Use with Wet vacuum pumps?	Yes – can be used with both wet and dry vacuum pumps	No – can only be used with dry pumps, oil vapours poison the catalyst
Utilities	Electrical power, compressed air, cooling water	Electrical power, Compressed air, cooling water, high purity O ₂ or H ₂ , Cryogenics

About the author

Dr Rob Grant is the founder and CEO of Gas Recovery and Recycle Limited. He has a masters degree and PhD from Cambridge University, UK, and is a fellow of the Royal Society of Chemistry. In 2008 Rob set up Gas Recovery and Recycle Limited to exploit ideas he had for a new gas purification system, based on chemical looping combustion with collaborators at Cambridge University. Rob was previously Head of Technology at Edwards Vacuum, where he was responsible for the company's systems engineering and PV business areas pioneering the introduction of many new vacuum and gas processing technologies. Rob can be contacted at rob.grant@GR2L.co.uk

Thus Centralised systems, by their very nature, are forced to employ additional purification technologies to remove O₂ and N₂ as well as the CO, H₂ and hydrocarbons from the process proper; this is not the case for Point of Use systems

Purification technologies

There are a number of purification technologies available in the market today, but the majority of these have been developed for the microelectronics industry, to purify process gases which are contaminated, at worst, at the ppm level and purify them to the ppb level. For the Solar PV application, a different approach is needed as the impurity levels in the exhaust argon can be as high as 10,000ppm. The majority of potential Ar recycle system providers have adopted a similar chemical strategy but with grossly different implementations.

Typically this involves a combustion step to convert the CO and H₂ to CO₂ and moisture respectively, followed by an adsorption or separation step to remove the CO₂ and moisture, leaving high purity argon. For the Point of Use system supplied by GR2L, with the ArgonØ, this is exactly the approach taken; it uses a unique chemical looping combustive purification reactor that utilises solid state oxygen carriers to ensure oxygen free recycled gas. A chemical looping combustion reactor typically comprises a packed bed made of a metal/metal oxide couple, that is repeatedly cycled from the oxidised state (metal oxide MO), to the reduced state (Metal M) and back again.

The process is strongly exothermic and is stoichiometric, rather than catalytic in nature. The overall process is chemically the same as gas phase oxidation, although during the process gas clean-up step there is no introduction of any gas phase oxygen. In fact the reactors will very effectively, and

simultaneously, remove any O₂ from the gas stream that may be present. The gaseous oxygen required for regeneration comes from ambient air and so the CLC approach does not require any additional gas supply.

The Centralised systems are inevitably more complex, as they also need to remove the O₂ and N₂ introduced into the recovered gas stream during the pump-down from atmospheric pressure of the vacuum furnaces. They also need to accommodate the wide variations in total flow as different furnaces end and start their respective production cycles at different times.

The concentrations of O₂ and N₂ can vary from as much as 10,000 ppm down to effectively zero; with wide variations in both feed gas flow and impurity concentration this presents a problem. In general purification systems work best with stable flows and concentrations. One manufacturer from Asia, attempts to avoid the problem by dynamically measuring the feed gas flow and impurity concentration to adjust an O₂ feed into a precious metal catalysed combustion reactor to just below stoichiometry – thus most CO and H₂ is converted to CO₂ and moisture, but not all.

However, the recovered gas does remain oxygen free. This mixture is then passed into a series of Pressure Swing Adsorption, PSA, beds to remove the residual CO & H₂ and the N₂, CO₂ and H₂O. The system is reported to operate at a recycle rate of approx. 80%, with a gas purity of N4.0. Additional purification steps can be added to meet the gas purity requirements from the customer.

Unfortunately, the catalyst in the precious metal catalytic combustion reactors is susceptible to poisoning by compounds found in vacuum pump oil (for example sulphur compounds) and so these systems are only suitable for dry vacuum pumped installations. While the current trend is towards dry vacuum pumps to evacuate the vacuum furnaces, at least 50% of the installed base utilise oil lubricated vacuum pumps and so rule out Ar recycle systems employing precious metal catalytic combustors.

The ArgonØ PoU Recycle System

The ArgonØ, figure 2. from Gas Recovery and Recycle Limited, GR2L, in the UK, is designed to recover, purify and recycle exhaust purge gas from both CZ and DS vacuum furnaces and the system uniquely copes with both oil sealed and dry vacuum pumps. It utilises chemical looping combustive

purification technology, jointly developed with Cambridge University, that enables system recycle rates of at least 95%, with recycled argon gas purities exceeding industry expectations.

The system is Point of Use and can connect to multiple vacuum furnaces, subject to a maximum exhaust gas recycle flow limit of approximately 15 Nm³/hr for each ArgonØ, enabling straightforward retrofit to current vacuum furnaces, along with a phased installation, in line with the installation of the vacuum furnaces, in new establishments. In one location in Taiwan an ArgonØ system connected to eight DS vacuum furnaces typically recycles approximately 11 tonnes of argon per month with an ArgonØ recycle rate of better than 95%.

None of the ~60 high performance ingots produced per month show any variation outside of normal measurement parameters. Sam Huang, Vice General Manager, said 'I have to say that the ArgonØ is a more flexible and compact system, compared with a centralised system concept' adding 'the installation process was straightforward and resulted in minimal disruption to our production schedules'.

In summary, the GR2L ArgonØ gives wafer manufacturers the opportunity of reducing their Ar consumption by up to a factor of 10 without impacting quality. The ArgonØ can be retrofitted to existing facilities with minimum interruption to production schedules, as well as be incorporated into new facilities, phased with the vacuum furnaces. It will also deliver a CO₂ footprint reduction of ca 3-5 Tonnes per vacuum furnace per year. ■

www.gr2l.co.uk

About the company

GR2L was founded in March 2008 by Dr Rob Grant FRSC with the objective of commercialising chemical looping combustion technology into the gas purification market. This was jointly developed with Cambridge University. GR2L is a cleantech company specialising in the recovery, purification and recycling of purge gases used in the photovoltaic, microelectronics and the material processing industry sectors. GR2L's primary focus is on the photovoltaic industry, where argon is used in large quantities during the crystallisation of silicon into ingots.

GR2L entered into partnership with the UK Gas Technologies group in 2010 to bring GR2L's flagship product ArgonØ –™ to market. The UK Gas Technologies Group has a track record in high purity gas installations in the semiconductor and medical markets and is synergistic with GR2L.

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