Converting Carbon Dioxide to Chemicals

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RSC Environment, Sustainability and Energy Forum

Executive Summary

Carbon dioxide can be converted into chemicals, fuels and polymers and there are technologies available today to achieve this. In context, the entire output of the chemicals industry (excluding fuels) is equivalent on a carbon basis to around 1-2% of the total annual anthropogenic carbon emissions (6Gt). Therefore, chemicals based on CO_2 could contribute to reducing carbon emissions but not at a significant level; the synthesis of fuels (for example via Fischer-Tropsch chemistry) could substantially increase carbon emission reduction. CO_2 as a building block in long-lived polymers can be viewed as a mechanism to sequester carbon in the long-term. There are a number of potential technologies that require further research and development; for example the photochemical conversion of CO_2.

In the medium-term it is expected that fossil fuel power plants fitted with carbon capture and storage technologies will provide a source of relatively pure CO_2. In order to take advantage of this it is suggested that CO_2-to-chemicals technologies should be applicable at relatively low CO_2 pressures to avoid energy intensive pressurisation steps. It is important that CO_2 conversion technologies are developed alongside work on carbon capture and storage (CCS) technologies so that they can be considered in the design of pilot and full scale CCS plants.

The full life cycle analysis of technologies for converting CO_2 into chemicals must be considered. This will enable an unbiased assessment of the technology options as well as an opportunity to compare against other carbon abatement technologies. It is also critical that an economic argument can be made to support technologies to convert CO_2 to chemicals.

Government and funding bodies should consider establishing a UK research centre with a focus on CO_2 chemistry. It is unclear whether this might be included in the remit of the recently announced energy and environmental research institute. In addition, to reduce the time for technological breakthroughs funding mechanisms that allow researchers from outside of the EU to collaborate with UK research groups should be introduced. There is a need to attract, train, enthuse and retain high calibre talented young people in disciplines relevant to this field.

Recommendations

Funding bodies

- There is a critical need for funding mechanisms that go beyond the laboratory scale and to the pilot plant demonstration scale.
- A dedicated funding programme into converting CO_2 into chemicals and developing a fundamental understanding of CO_2 chemistry is required, but it must compliment existing energy programmes.
- Funding mechanisms that encourage multi-disciplinary consortia are required; this should include mechanisms for international collaborative projects and partners.
- A new research centre to bring together researchers in this field would facilitate collaboration on the level required.

Skills and education

- There is a perceived science and engineering skills shortage looming that needs to be addressed now.
- To be successful in this field talented young scientists and engineers need to be attracted, trained, enthused and retained.
- Highlighting the positive environmental impacts of research in converting CO_2 into chemicals could attract researchers into the field.

Government bodies

- Government needs to accept the risk associated with bringing promising research through to commercial demonstration as there are few other UK mechanisms to achieve this.
- Clarification is required on the exact nature of the energy and environmental research institute and whether its scope will encompass research into converting CO_2 into chemicals.

Learned societies

- The RSC will disseminate the report of this workshop to key policy makers, funding bodies and interested others.
- Learned societies must collaborate to identify and focus upon the key research areas for converting CO_2 into chemicals and inform Government and funding bodies as well as the wider scientific and engineering communities. This could be achieved by a focused multi-organisation workshop.
- Learned societies, with their extensive networks can help to identify and disseminate current world research into converting CO_2 into chemicals.
- Learned societies can also assist in identifying people with requisite expertise that will compliment R&D consortia in this area.

Introduction

The workshop was organised by the Royal Society of Chemistry Environment, Sustainability and Energy Forum. It aimed to explore the chemical science behind the conversion of CO_2 into chemicals; to determine where the UK is in terms of academic expertise in this field and to indicate research priorities for the UK in the future. This workshop was attended by scientists and engineers from different backgrounds in both academia and industry. This report summarises the presentations and the discussion during the breakout sessions. Copies of the presentations are available from the RSC website[1].

The Presentations

Chairman's introduction
Professor Chris Rayner
University of Leeds

During his brief introduction Professor Chris Rayner put the scale of the CO_2 problem into context as he showed the International Panel on Climate Change (IPCC) hockey stick diagram and demonstrated the relationship between increasing world primary energy use and rising CO_2 levels. He then went onto put world annual CO_2 production (1×10^{10} tonnes per year) into the context of the chemical industry that produces around 50 – 100 times less carbon based chemicals annually. He stated that the challenges are to reduce CO_2 emissions, reduce the use of petrochemical feedstocks and increase the utilisation of CO_2. He noted that in recent years the number of CO_2 publications has been rising "almost as fast as the levels of CO_2 itself". He ended by stating that "chemists are uniquely positioned to make fundamentally important contributions to climate change and reduction of carbon dioxide levels, but this represents an enormous challenge".

Carbon capture and storage
Professor Colin Snape
University of Nottingham

Professor Colin Snape began by stating a few facts about the current UK position in world energy and CO_2 emissions (UK emits 2% of world CO_2 emissions). Currently the UK (and elsewhere) faces an energy gap as nuclear and coal power plants of 25GW capacity are due to close by 2023 and that it is likely that a mixture of nuclear, coal, gas, renewables and energy efficiency will fill this gap. In context, China currently annually installs electricity generation capacity equal to the total UK demand.

It is anticipated that cheap and available coal, the most CO_2 intensive of the fossil fuels, will continue to be a major source of power well into this century and it is vital that clean coal technologies are developed to mitigate CO_2 emissions. This was recognised in the recently published DTI 'The Energy Challenge' report which called for the development of a commercial demonstration of a coal power plant fitted with carbon capture and storage (CCS) technology. To be cost competitive with CCGT coal gasification

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[1] http://www.rsc.org/ScienceAndTechnology/Events/ConvertingCO2toChemicals.asp

(IGCC) technology coupled with CCS technology requires that the EU Emissions Trading Scheme offers a price for carbon at €40-50 per tonne. Coal (and natural gas) gasification coupled with CCS also offers a route to clean hydrogen production; this is an option for decarbonising vehicles. A number of coal technologies linked with CCS technologies are currently planned for demonstration. There remain a number of scientific and engineering challenges to address before clean coal technologies can be commercialised including, improving plant efficiency, materials to capture CO_2, reducing the energy intensity of CO_2 compression and CO_2/H_2 separation membranes.

Captured CO_2 can feasibly be stored in depleting/depleted oil and gas fields, deep saline aquifers, unminable coal seams or as chemical products. Global capacity for storage (perhaps >10,000 Gt) is significantly greater than annual (natural and anthropogenic) CO_2 production (25Gt per year) and therefore CCS could make a substantial impact of world CO_2 levels over 100's of years. However, it is critical that CO_2 does not leak and therefore monitoring of storage sites is crucial.

Carbon dioxide as a building-block for molecular organic compounds
Michele Aresta
University of Bari

Professor Michele Aresta introduced the talk by showing us his paper on CO_2 chemistry that was published in Chemical Communications in 1975. Since 1975 he has not been idle and has published numerous papers and books on the subject. Professor Aresta stated that CO_2 essentially has two classes of reaction, firstly the entire molecule can be incorporated into a product in a non-energy intensive process and secondly it can be converted to a C1 molecule in an energy intensive reaction.

Professor Aresta described an array of processes by which CO_2 can be converted into industrially relevant chemicals, including:

- CO_2 can be bonded to metals such as nickel through an η^2-C, O bond, or to other metals in several other modes of bonding, including η^1-C and η^1-O. In these systems CO_2 acts as a nucleophile through the oxygen and is reactive towards a number of electrophiles (for example protons, alkyls, silyls, metals and also hydrides).
- CO_2 can be converted to methanol through a multi-step enzymatic process.
- CO_2 can be directly coupled with methane to produce acetic acid, however, this is a developmental process and yields are currently low.
- Metal complexes have been used to couple CO_2 with terminal olefins (the target reaction is $RCH{=}CH_2 + CO_2 \rightarrow RCH{=}CHCOOH$) but it is important to avoid the formation of an O-M and H-M bond as if this happens the product is not released from the complex.
- There has been intensive study into the preparation of lactones, carboxylic acids and esters from dienes using metal ligand complexes. There is a challenge in producing linear products, but cyclic products are readily prepared (for example six- or five-membered lactones). The ligands at the metal centre are critical in determining selectivity.

- The clean synthesis of carbamates (intermediates for polymer synthesis) are the subject of an EU integrated project where it is hoped to eliminate the need to use phosgene and reduce energy requirements. Several convergent strategies are being examined including the reaction of CO_2 with amines and alkylating agents, including organic carbonates.
- The synthesis of cyclic carbonates can be achieved by reacting CO_2 with an epoxide or an alkene plus oxygen. Professor Aresta's group has examined the mechanism of cyclic carbonates synthesis and developed a heterogenous catalyst for the system. Asymmetric cyclic carbonates can also be synthesised. The group has also developed a room temperature reaction to synthesise linear carbonates, this is carried out under the auspices of the EU project TOPCOMBI.
- CO_2 can be used as a mild oxidant in the dehydrogenation of alkanes; this potentially offers a route to hydrogen production.
- CO_2 can also be used as a solvent and also as a refrigerant.

In summary Professor Aresta said that if all the technologies he described were implemented at industrial scale then carbon emission reduction could be as high as 350Mt per year (ca. 6% of annual anthropogenic carbon emissions) alongside the associated reduction in the amount of fossil fuel extracted. He also stated that it is very important that all such technologies are examined on the basis of their whole life cycle to highlight significant social, economic or environmental impacts occurring as a consequence of the process.

Converting CO_2 into Fuels and Chemicals: The Formic Acid Economy
Professor Peter Hall
Strathclyde University

Professor Peter Hall described a consortium that arose out of an EPSRC sandpit event that aims to examine the feasibility of a formic acid economy. The consortium is examining projects on formic acid synthesis (from CO_2), clean hydrogen synthesis, conversion of formic acid into chemicals and polymers as well as using formic acid in fuel cells. He started by describing both the advantages and disadvantages of hydrogen as an energy vector and in particular stated that in 12 years research into hydrogen storage he has seen no success.

There are several options for chemical storage of hydrogen such as methane, methanol and formic acid. The technology to reform methane into hydrogen is well established, but because methane is a gas there are still storage and handling problems (although a methane infrastructure exists). Methanol as a liquid fuel is easier to handle than hydrogen and can be used in direct methanol fuel cells (DMFC), however, it suffers from slow oxidation kinetics and has a problem with methanol diffusion to the cathode.

To date there has been limited interest in formic acid and applications have generally been on small scale. Professor Hall has been examining the use formic acid in a DMFC. In initial experiments there was significant power drop-off associated with poor fuel diffusion. Increasing the formic acid concentration increased the power, however, mass transfer remains a problem. A potential solution may be the use of an oscillating baffle reactor.

Professor Hall indicated that a direct formic acid fuel cell could potentially be used to convert glycerol (a by-product of biodiesel synthesis) to formic acid and hydrogen.

CO_2 as a feedstock in polymer synthesis
Dr Charlotte Williams
Imperial College London

Dr Williams began by highlighting the size of the plastics industry, particularly with respect to packaging and building materials. Polymers made from petrochemicals are often durable and this is resulting in a build up in landfill sites around the world. Dr Williams proposed a biorefinery concept where polymers are made from renewable raw materials and are designed to biodegrade at end of life.

Carbon dioxide was used in polymer synthesis as far back as the 1960's where Japanese researchers showed that epoxides could be reacted with CO_2 to form polycarbonates. More recently the mechanism has been determined and it is known that a Lewis acid metal complexed with bulky ligands is required and that the CO_2 is inserted into regioselectively onto the least hindered carbon of the metal alkoxide.

Dr Williams was interested into whether CO_2 could be reacted with propylene imine compounds to selectively form polyurethanes at mild temperatures and pressures. Her research showed that oligomers of up to 14 units could be formed under the right conditions of temperature, pressure and using an aluminium-salen as initiator. Dr Williams also found that 2,2-dimethylaziridine will react at room temperature with CO_2 in the presence of chromium-salen initiator to form a good yield of the poly(amine-urethane) with a regioselective addition at the CH_2 next to the NR`.

Dr Williams also discovered that *N*-substituted aziridines react with CO_2 in the presence of the catalyst to form the corresponding oxazolidone in good yields.

Save the world? Maybe!
Professor Malcolm Green
Oxford University

Professor Green opening his presentation by over viewing oxyreforming processes and describing the work of his group on interesting molybdenum carbide catalysts for dry reforming ($CH_4 + CO_2 \Rightarrow 2CO + H_2O$). Professor Green then went onto explain that still nobody understand the mechanism of the Fischer-Tropsch reaction and that if it were possible to increase the selectivity towards carbon chains of specific lengths then this would be a major breakthrough.

Professor Green then went on to say that the hydrogen economy is <u>NOT</u> the answer to life after fossil fuels or global warming! He restated Professor Hall's comments about the difficulty in storing hydrogen, especially in the gaseous phase. Professor Green suggested that to save the world you need energy to make hydrogen, CO_2 and water to make CO and CO and hydrogen to make liquid fuels and that this cycle is sustainable. In this system the net effect contribution of CO_2 to global warming would be zero.

Energy to make H_2 from the electrolysis of water could be derived from renewable power (such as solar or wind) or from nuclear fission or fusion. CO_2 would be derived mainly from biomass. Professor Green went on to describe the concept of international energy centres where huge concentrations of power generation (e.g. giant wind farms or units of several hundred nuclear power stations all in suitable remote locations) supply the power for electrolysis of water. The Fischer-Tropsch plants would also be located at these sites.

Diesel and gasoline would be created in vast quantities and shipped around the world by sea (driven by some of the produced fuel). On delivery the ships would fill up with dried biomass for the return journey where it would be converted to more fuel (biomass + air $\Rightarrow CO_2 + CO + H_2$).

Professor Green finished with some key research targets:

1) To optimise the production of H_2 from water
2) Selective Fischer-Tropsch conversion of syn-gas to hydrocarbons
3) More research into photovoltaics
4) Efficient biomass production

Photochemical transformation of CO_2
Dr Mercedes Maroto-Valer
University of Nottingham

Dr Maroto-Valer began by stating that carbon management will require us to improve our efficiency, sequester CO_2 and reduce our carbon intensity. The worldwide demand for chemicals only accounts for around 1-2% of global carbon emissions, whereas fuels account for a higher proportion. Photochemical transformation of CO_2 to fuels is therefore potentially an attractive route to utilising CO_2.

Photocatalysis is the acceleration of a photoreaction in the presence of a phtocatalyst. In essence light is used to causes an electron (e^-) to move into a conduction band which leaves behind an electron hole (h^+). The electron, if it has a sufficient lifetime, can be used in reduction reactions. A number of semi-conductors have been used, but in her presentation Dr Maroto-Valer chose to focus on TiO_2. Unfortunately unmodified TiO_2 is not activated by sunlight, which would be an ideal system, however, it can be modified so that sunlight can be used promote photochemical reduction of CO_2 with water. In this reaction key products (isolated in yields of <1%) include methane, formaldehyde and acetic acid and can involve processes that require up to $8e^-$ transfer (which is very difficult to achieve). An FTIR study of the system has shown that H_2O acts as an electron donor. The operating conditions are very important in the system, a high temperature is required to overcome the band gaps to improve conversion, high CO_2 pressure is required to maximise the concentration of CO_2 in water and the photocatalyst is best dispersed inside a zeolite. The reactor design for a 2-phase or 3-phase system is also critical, and both fixed and fluidised bed designs have been used. Ideally the reactor would use sunlight and most of the prior knowledge here has been based on systems for waste water treatment.

A doped TiO_2 system has been successfully produced that uses UV/visible light for photoreduction reactions. In this case it was proven that without light radiation no reaction was taking place.

In summary Dr Maroto-Valer stated that whilst photocatalytic reduction of CO_2 is a promising strategy, there is much (multi-disciplinary) research still needed to achieve breakthroughs.

Topics

A summary of the key points arising from discussion is described below:

Context

The workshop participants expect the UK to meet its 2012 Kyoto target for carbon emission reduction (12.5% below 1990 levels). However, there was less certainty whether the UK would achieve the 2050 domestic target to reduce carbon emissions by 50% - carbon capture and storage (CCS) technologies, along with other technologies and energy efficiency measures will be vital achieve this. A sector by sector analysis should be carried out to examine exactly what can and cannot be done towards achieving CO_2 targets. Improving the efficiency of existing processes (for example increasing the efficiency of coal burning power stations) should be carried out alongside the development of new technologies.

Globally 6 giga tonnes of anthropogenic carbon are produced annually and the UK accounts for about 2% of these emissions. To put the UK contribution in context, it is approximately the same as that released by the entire EU refining industry. Technologies such as CCS will need to be adopted by countries that have the highest carbon emissions, such as America, China and India. However, countries such as the UK should demonstrate such technologies at a commercial scale. To encourage the development and commercialisation of technologies that reduce carbon emissions in the UK a long-term commitment to measures, such as the EU emissions trading scheme and the UK renewables obligation is critical.

It is expected that a commercial demonstration of CCS technology will be running by 2020-25, if not before. Therefore, there is a window of opportunity to incorporate CO_2 conversion technology into such plants if it can be developed in time.

It was noted that CCS technology fitted to large combustion plants co-firing biomass actually represents a mechanism for reducing the amount of CO_2 in the atmosphere as the CO_2 captured during biomass growth would be locked away in geological storage sites.

UK strengths

Participants indicated that the UK has strengths in the following areas:

Catalysis – The UK has world class researchers in the field of catalysis. Professor Michele Aresta demonstrated in his lecture that catalysis is an enabling technology and that there is huge scope for the development of catalysts for converting CO_2 into chemicals.

Materials – the UK has a strong materials research capability and new materials and polymers could be developed using CO_2 as a feedstock.

Materials and polymers, as previously discussed, offer an opportunity to sequester CO_2 for the long-term.

High added value chemicals – the commodity and fine chemical industry and research base in the UK has expertise that will be able utilise CO_2 in high value chemical synthesis. There is a natural supply chain in place in the UK to supply chemicals from these industries to industries such as the pharmaceutical industry where further value be derived.

Biomass – biomass incorporates CO_2 during its growth and therefore converting biomass in chemicals is an indirect mechanism to utilise CO_2. The UK is building up research capability in this area particularly in biomass gasification, fermentation and catalytic transformation of biomass.

Supercritical CO_2 as a solvent – the UK has considerable experience in the use of CO_2 as a solvent for chemical reactions and for extraction at both the research and commercial scale.

Large scale processes – the UK has expertise in large scale chemical processes of direct relevance to CO_2 handling and conversion such as hydrogen production, synthesis gas production and Fischer-Tropsch upgrading.

Political will - the UK will be an early mover with large scale CCS demonstration projects. This offers an opportunity to examine the feasibility of CO_2 technologies.

Academic – industry collaboration - this was considered to be strong in the UK and is important in bringing ideas through from basic research to demonstration.

UK Focus

There was considerable discussion relating to what the UK should focus on in relation to CO_2. It was broadly agreed that no single technology will arise as a silver bullet and that it is important to focus across a number technology platforms.

Technologies to convert CO_2 to chemicals applied to a large scale CCS plants ideally would utilise the low pressure (<20 bar) CO_2 stream that is produced; this would avoid the need to compress CO_2 which is energy intensive (and thus expensive).

There was uncertainty over the scale that CO_2 conversion technology could be applied. Specific technologies such as syn-gas and Fischer-Tropsch are known and can be applied at large scale, however, many of the newer technologies discussed (for example photochemistry, which potentially represents a truly sustainable energy supply if the energy is derived from sunlight) are currently at the laboratory scale stage.

It is possible to chemically convert CO_2 into chemicals, polymers and fuels. In discussion, three schools of thought emerged on how to focus efforts. The first premise was that effort should focus upon **large-scale production of fuels from CO_2** as the scale of the industry will maximise the impact of the technology. The second premise is that a **broad range of chemicals at varying scales** could be synthesised from CO_2. The final premise was that CO_2 conversion should concentrate upon **polymers** where the CO_2 would be locked away for significant timescales.

Since a number of technologies are at the research stage there is an opportunity to incorporate the principles of green chemistry and to seek areas where energy use can be minimised. Where energy is required, ideally it would be supplied by renewable resources.

It is important to consider the full life cycle analysis (cradle to grave or cradle to cradle) of technologies for converting CO_2 into chemicals. The energy required to compress, transport and store CO_2 and the fossil fuel displaced by chemical products from CO_2 needs to be taken into account in such calculations. This will enable an unbiased assessment of the technology options as well as an opportunity to compare against other carbon abatement technologies (CATs). It is also critical that an economic argument can be made to support technologies to convert CO_2 to chemicals.

Collaboration

Participants emphasised that a multi-disciplinary approach is required in order to significantly progress the research into converting CO_2 into chemicals. In particular collaboration between engineers, biologists, physicists and materials scientists is required. To provide a complete and robust case for technology options economists, life cycle analysis experts and social scientists will also need to be incorporated into research teams. As will be discussed later, it is important that funding mechanisms are available that allow such consortia to be constructed.

Industry must be engaged from the start of research projects to provide focus and steer towards viable research goals and realistic technology options. Early industry engagement also produces partnerships that can be exploited as research progresses towards the development and demonstration phases. Industry will also be able to provide critical economic and market data that will steer projects towards realistic targets.

To be successful consortia will need to attract a balance of the right skills, expertise, facilities and commercial partners. Elements of these may only be available outside the UK and therefore European funding platforms and international collaboration mechanisms need to available in order to maximise the probability of success. Applicants can apply through Engineering and Physical Sciences Research Council (EPSRC) responsive mode to allow them to work with international academics - be that through Overseas Travel Grants, bringing across researchers to the UK - Visiting Researchers, or by including said international academics on responsive mode grants as project partners[2].

Government and Funding

Participants strongly emphasised the need for UK mechanisms that go beyond the laboratory scale and to the pilot plant scale in order to truly demonstrate the market feasibility of processes. It was considered that the UK Government needs to take the risk in order to make this happen as venture capitalists and industry are risk averse at this crucial phase.

Is there a need for a dedicated funding programme into converting CO_2 into chemicals? The UK research councils are substantially increasing the funding available for energy research over coming years (which has lead to the formation of the UK Energy Research Centre and the SUPERGEN project) and the DTI technology programme mechanism also funds energy projects. Any dedicated funding mechanism would need to link such initiatives together. Whether it will be possible to tap into this money for R&D into converting CO_2 into chemicals is uncertain. It was rightly highlighted that the research council responsive mode is a flexible programme that could provide a mechanism to bring together research teams in this area. Responsive mode covers both laboratory and pilot-scale projects. It was also noted that in two recent EPSRC sand-pit events (the IDEAS Factory sandpit[3] and the Green and Sustainable Chemical Technologies Sandpit[4]) that two consortia (including the one that Professor Peter Hall described) that are looking into converting CO_2 into chemicals have been developed and funded.

Future funding mechanisms and research initiatives in this area must be long-term and provide a clear mechanism for taking basic research through the development and demonstration phases.

Research Centres

There is a need for a new large UK research centre/central laboratory to carry out research into converting CO_2 into chemicals, possibly alongside other energy research. The research centre could either be hosted at one site or could be a virtual research centre to exploit researchers across a range of UK universities. It was also suggested that a European research centre could also be established using EU FPVII funding.

Participants were interested in the £1 billion energy and environmental research institute that was announced in the 2006 budget by Gordon Brown. However, there was uncertainty as to what exactly this research institute will do and whether it offers an opportunity for research into converting CO_2 into chemicals. Limited information about this initiative can be found on the DTI website[5].

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[2] http://www.epsrc.ac.uk/InternationalActivity/HowtoGetFunding/default.htm
[3] http://gow.epsrc.ac.uk/ViewGrant.aspx?GrantRef=EP/D023653/1
[4] http://gow.epsrc.ac.uk/ViewGrant.aspx?GrantRef=EP/E010318/1
[5] http://www.dti.gov.uk/science/science-funding/eti/page34027.html

Education

Participants were worried about skills shortages and whether this area would suffer from the same problems in attracting skilled and enthusiastic people in the same way that the nuclear and oil industry have suffered in recent times. It was noted that a UK contractor has recently employed a record number of retired chemical engineers due to this problem. Converting CO_2 into chemicals should be marketed as green and sustainable science in order to attract talented people.

A wider point was raised regarding the need attract, train, enthuse and retain high calibre talented young people generally, and in disciplines relevant to this field.

Workshop participants

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Dr Donald Barr	Degussa Stanlow Limited
Dr Will Barton	Oxford Catalysts Ltd
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