Graphene oxide based nanocomposite membrane for efficient CO₂ separation

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Abstract: Graphene oxide (GO) is a one-atom-thick two dimensional material which has unique mechanical, electrical and structural properties. Recently, the attempt to apply GO in gas separation membranes has attracted increasing attention due to the molecular sieving effect of the controlled GO defects and its potential to form high efficient gas flow channels. However, most current works focus on the flat sheet GO membrane prepared via filtration or blending, and performance of the GO nanocomposite hollow fibre membranes still remain poorly understood.

In this study, a series of hollow fibre gas separation membranes were prepared with GO and PEBAX[®] as the selective layer via a dip coating method on commercial PVDF microfiltration membrane surface. Prior to the GO/PEBAX coating, the support membrane was initially coated with a layer of PTMSP in order to minimize the penetration of the GO/PEBAX into the membrane pores. The gas separation results under room temperature revealed the GO content had significant effect on the membrane performance. With the increase of GO content in the PEBAX layer, the CO₂ permeance of the composite membrane firstly decreased then increased. GO nanosheets are impermeable to gases, thus the presence of GO layer inside the PEBAX would increase the membrane resistance. On the other hand, the confined laminar structure also hindered the crystallization of PEBAX, which led to higher free volume near the polymer-GO interface, which provided high efficient gas transport channels and resulted in higher permeance. The optimized GO/PEBAX nanocomposite membrane could improve the membrane permeance by 50 % while remain the selectivity unchanged when compared with pure PEBAX benchmark.

Keywords: gas separation, graphene oxide, membrane.

1 Introduction

Organic-inorganic mixed matrix membranes have been prepared for gas separation purposes and they have shown promising gas mixture separation performance. The basic concept of the mixed matrix membrane for gas separation is based on either selective adsorption or molecular-sieving mechanisms. Recently, the application of the graphene based materials is considered promising for gas separation membrane preparation. The special one carbon atom thick structure could potentially provide separation function with minimized transport resistance and maximized permeation flux. In addition, the graphene material has good mechanical strength and conductivity, which could also provide unique functions to the membrane [1].

However, the membranes prepared with pure graphene sheets have been proved to be impractical for gas separation as even the smallest gas molecules like hydrogen and helium can't permeate through the sheet. Recently, porous graphene sheets with nitrogen-functionalized subnanometer pores has been investigated for gas separation, and it exhibited superb gas separation performance. However, due to the strong aggregation tendency, the use of such materials for membrane preparation is still challenging. In addition, the free standing graphene membrane with no substrates is too brittle thus difficult to use in practical applications. The growth of graphene layer on an inorganic membrane surface, on the other hand, is difficult and costly. Therefore, more advanced approaches are required to prepare the graphene based organic membranes.

The most commonly applied gas separation polymeric membranes are glassy polymers, for example polyimide (PI), polyamide (PA), polycarbonate (PC), polysulfone (PSf), polyphenylene oxide (PPO), cellulose acetate (CA) and their derivatives [2, 3]. The main separation mechanism is based on the size sieving by the rigid chain structures. However, the CO_2 permeation rates were not satisfactory to

deal with large volume industrial scale applications such as flue gas separation and natural gas purification. On the other hand, the membranes prepared with rubbery polymer have higher CO_2 permeability due to the flexible chain structure. However, their low selectivity is detrimental. In order to solve this problem, the application of co-polymers containing both glassy and rubbery chains has been investigated. Pebax[®] (tradename of Atofina company for polyether (PE)-block-polyamide (PA) copolymer (PEBA)) is a commercialized block copolymer containing –N-H-, H-N-C=O, and O-C=O groups, and it has been proven to have satisfactory CO_2 separation performance [4].

One major challenge to prepare the graphene functionalized membrane is to control its stacking structure, which has strong connections to the membrane gas separation performance. We thus applied the dip-coating technique in this work, trying to form a thin coating layer with parallel staked graphene sheets with the assist of gravity.

In this study, the Pebax/graphene oxide (GO) composite gas separation membranes were prepared with PVDF microfiltration membrane substrates. The membranes were pre-coated with PTMSP gutter layer to minimize the penetration of the Pebax layer into the membrane pores. Several different graphene oxide concentrations and coating cycles were tested and compared to optimize the performance of the composite membrane.

2 Results and discussion

2.1 Characterization of the composite membrane

The SEM images of the PVDF-PTMSP-Pebax/GO nanocomposite membranes are shown below in Figure 1. The coating layer thickness was around 1 micron and the parallel stacked GO sheets were clearly visible in the cross-section images, and the nanosheets were evenly distributed within the Pebax layer. Such an observation is preferable as the formed nanoscale flow channels between the neighbouring GO sheets would provide higher concentration gradient for CO_2 transport and thus potentially improve the permeability. In terms of the surface morphology, the addition of GO would increase the surface roughness. The membrane surface without GO sheets was smooth and even (results not shown), and with the addition of GO sheets, the formation of hills and valleys was clearly observed on the membrane outer surface.

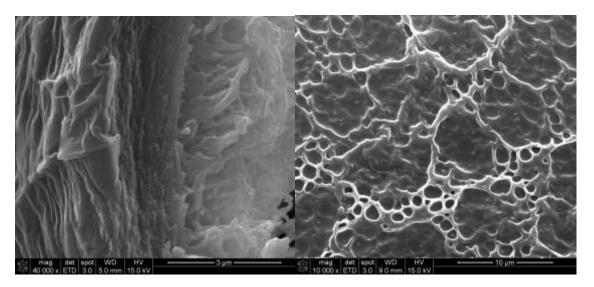


Figure 1: SEM image of the composite membrane (left: cross-section, right: outer surface)

2.2 Gas permeation test with the composite membrane

The content of GO in the Pebax layer had significant effect on the membrane performance. As shown in Figure 2, the addition of GO into Pebax layer would increase the permeability of CO_2 gas, and the highest value was achieved when 0.1 wt % GO was applied. On the other hand, the CO_2/N_2 selectivity

of the membrane was relatively unchanged with the addition of GO sheets (CO_2/N_2 selectivity: 40±5). As presented above, the addition of GO would form a series of parallel flow channels for the membrane, and when CO_2 passed through these channels the confined space would provide higher concentration gradient, which eventually increased the gas permeation rate. On the other hand, the presence of GO within the Pebax layer hindered the crystallization of the polymer, leaving void flow channels near the polymer/GO surface. Such channels provided high efficient gas transport through the membrane thus facilitated the gas transport. However, the addition of the GO sheets had negative effect on the CO_2 separation mechanism through the Pebax layer thus the selectivity was relatively unchanged. When the GO content was higher than 0.1 wt %, the impermeable layers provided extra mass transfer resistance for CO_2 to pass through the membrane, thus the permeation rate was significantly reduced. The proposed mixed matrix membrane gas permeance is presented in Figure 3.

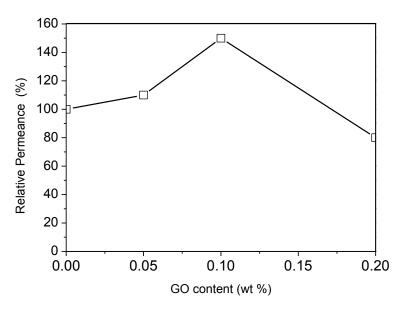


Figure 2. Change of CO₂ permeance and selectivity with different GO content (2 coating cycles)

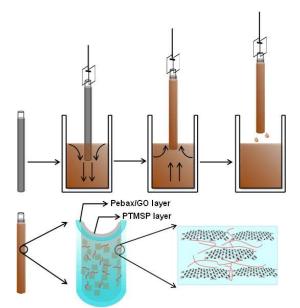


Figure 3. Schematic diagram of the dip coating and gas transport through the GO/Pebax layer

3 Conclusions and recommendations

This work prepared a series of the Pebax/GO based nanocomposite membranes and tested them for CO_2/N_2 separation. The results indicated the presence of GO within the selective layer had significant

effect on the membrane performance. The addition of GO into the Pebax layer would form high efficient nanoscale flow channels for CO_2 gas, which further improved the membrane gas separation performance. However, the detailed mechanism of the GO sheets inside the membrane was still poorly understood and further investigation is thus required.

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Presenting author biography

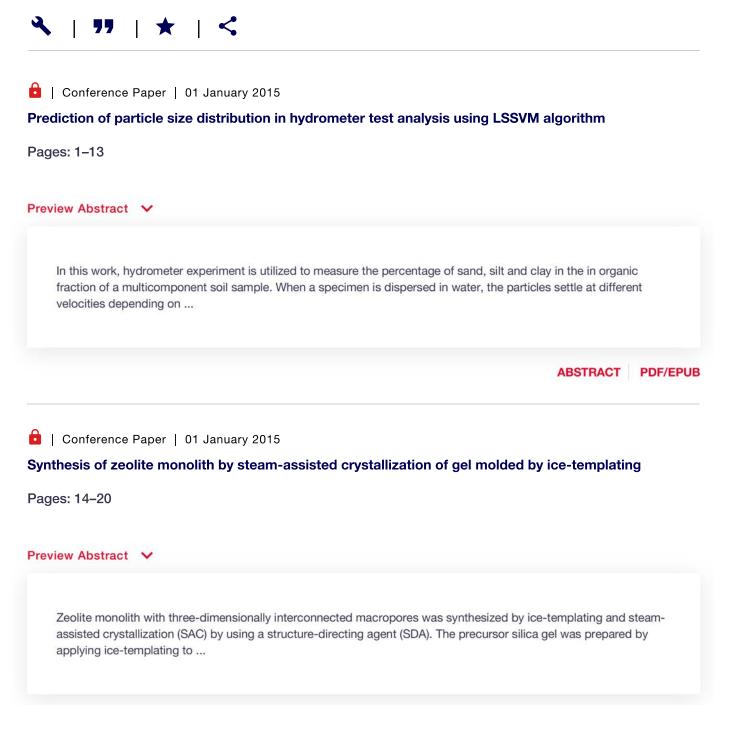
Jingwei Hou received his Ph.D. degree at the University of New South Wales in the area of biocatalytic membrane for wastewater treatment and gas separation in 2015. After completion of his Ph.D., he joined the UNESCO Centre for Membrane Science and Technology at the University of New South Wales as a research associate. His current research interests include gas separation membrane, biocatalytic membrane reactor and gas-liquid membrane contactor.

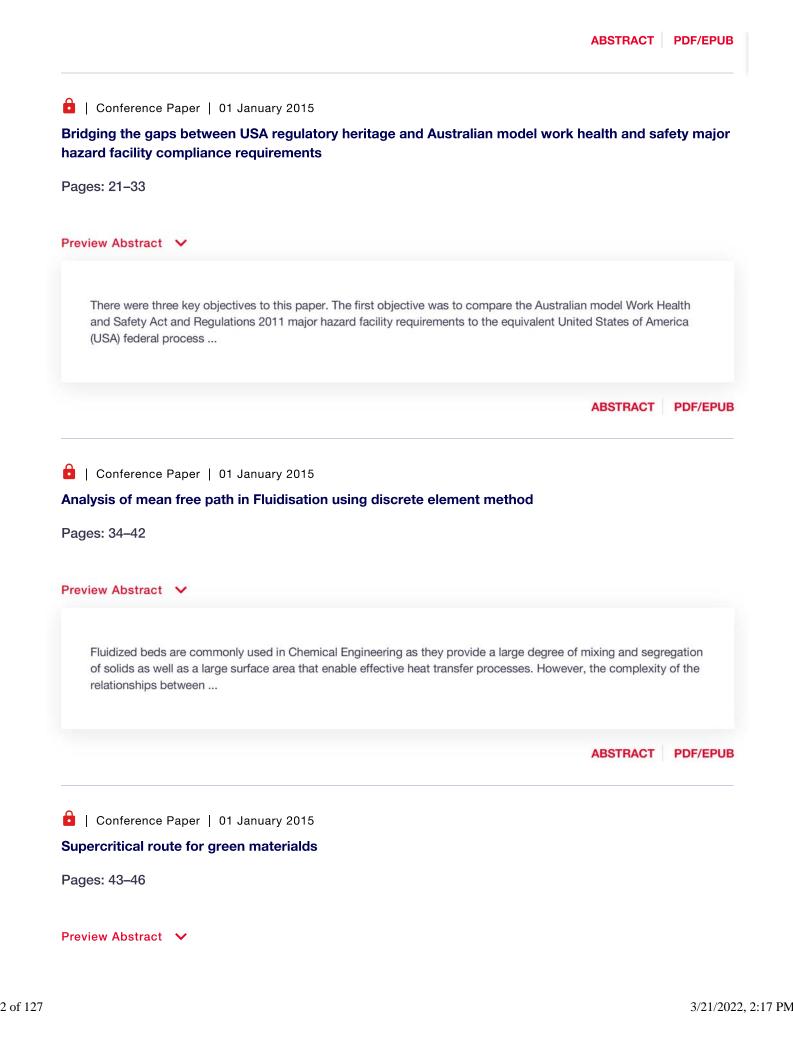


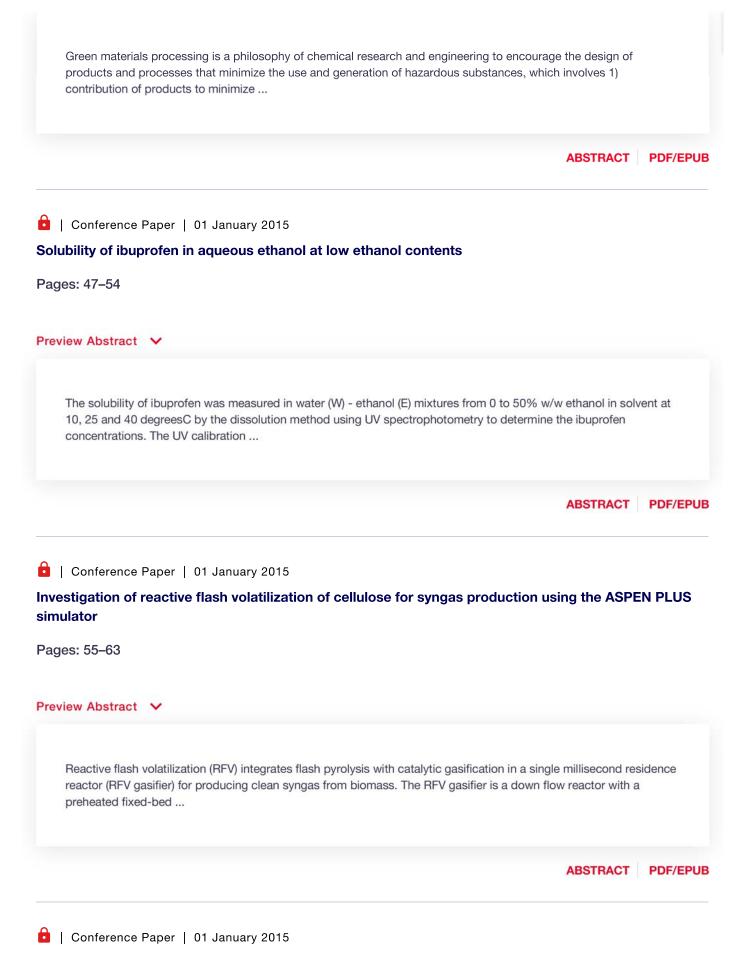
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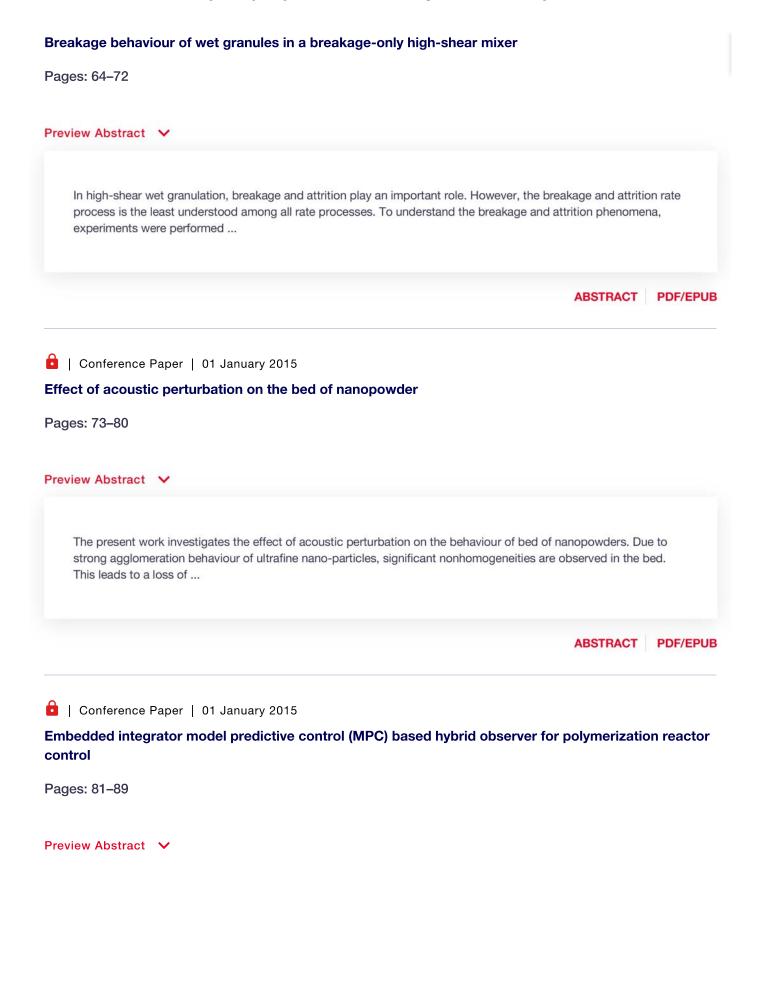
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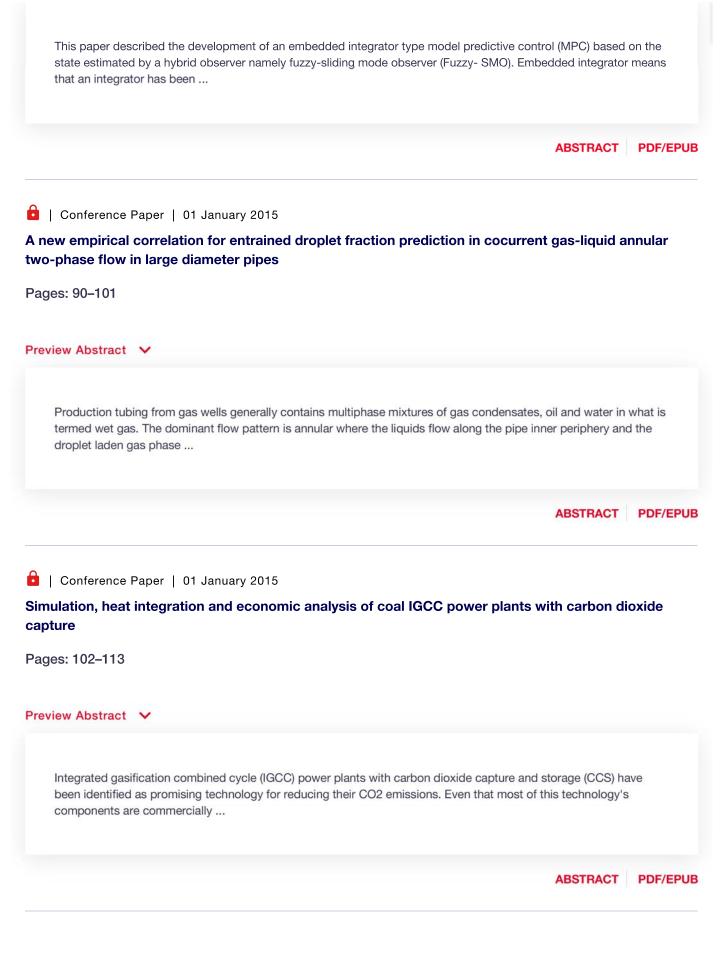




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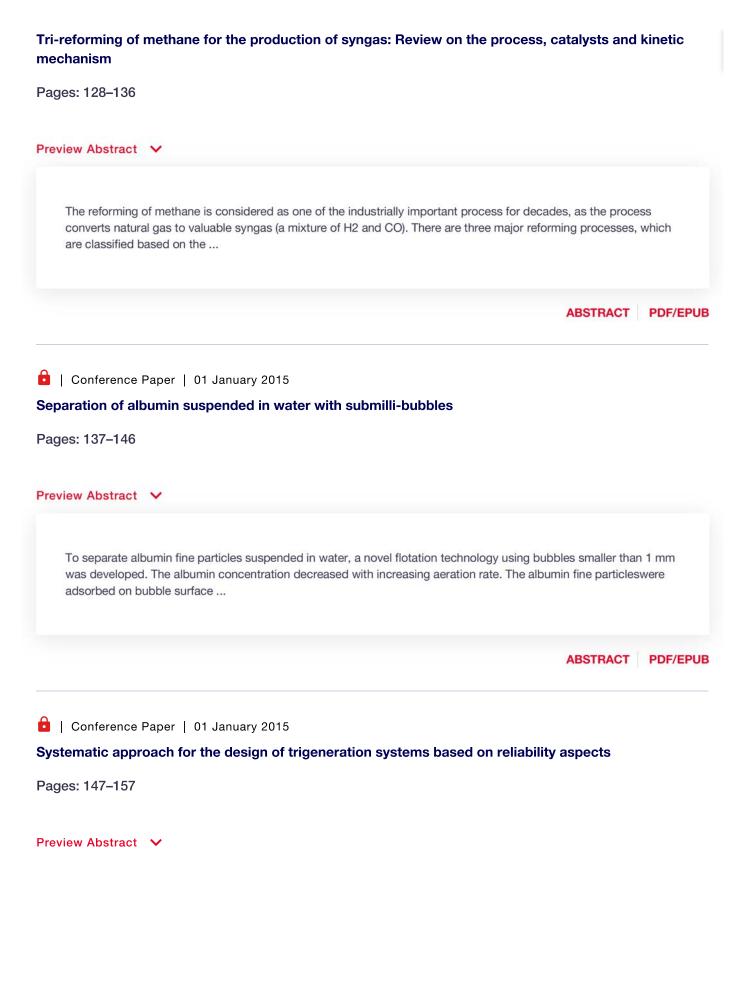
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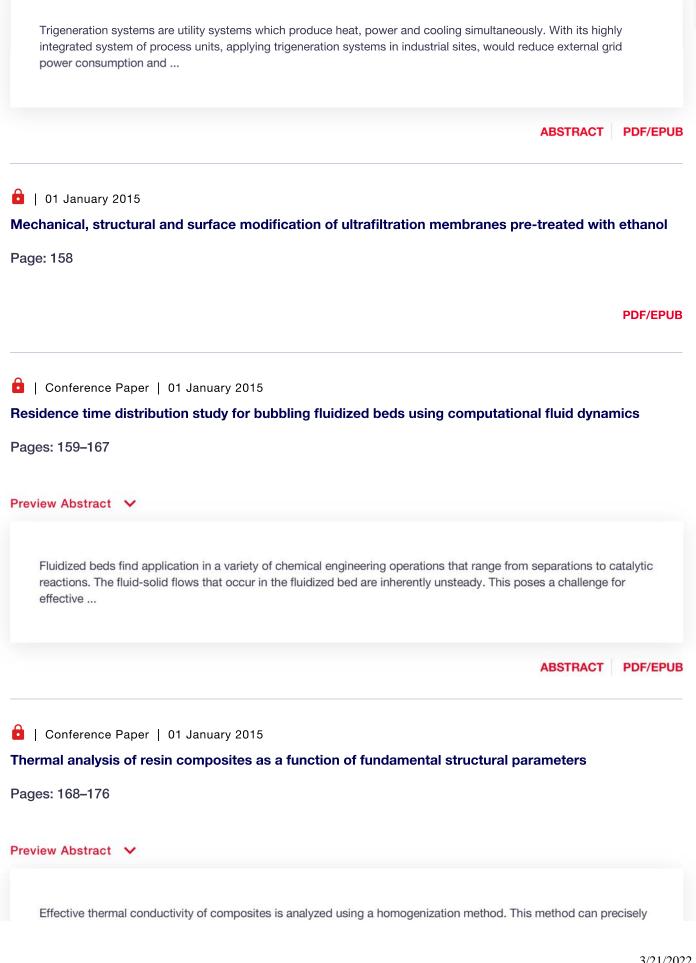
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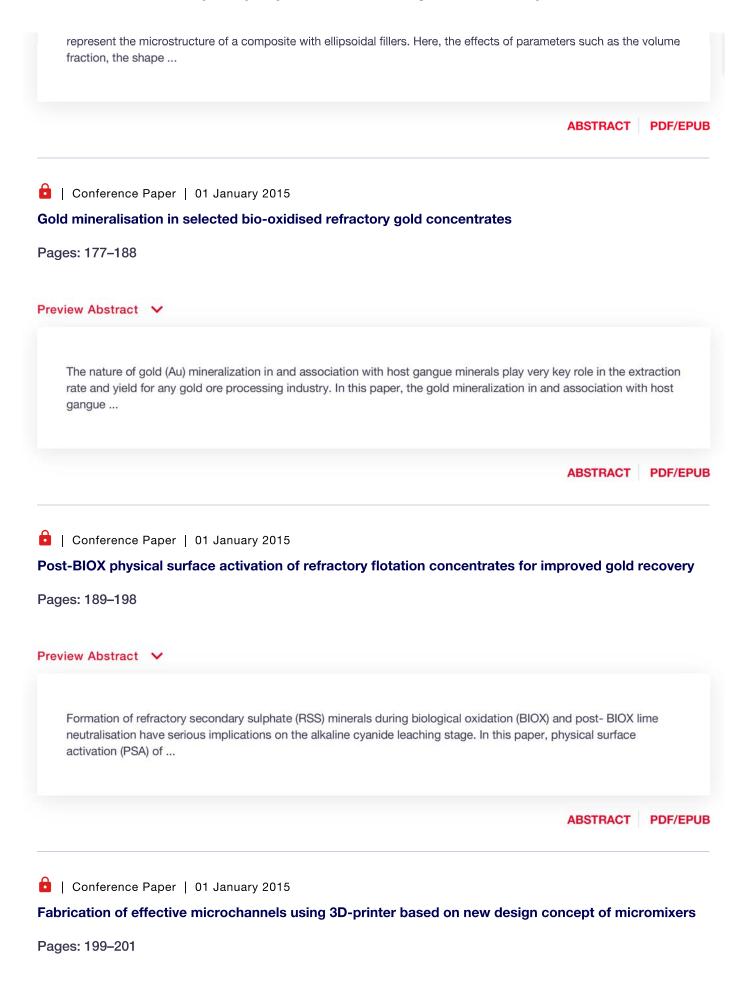
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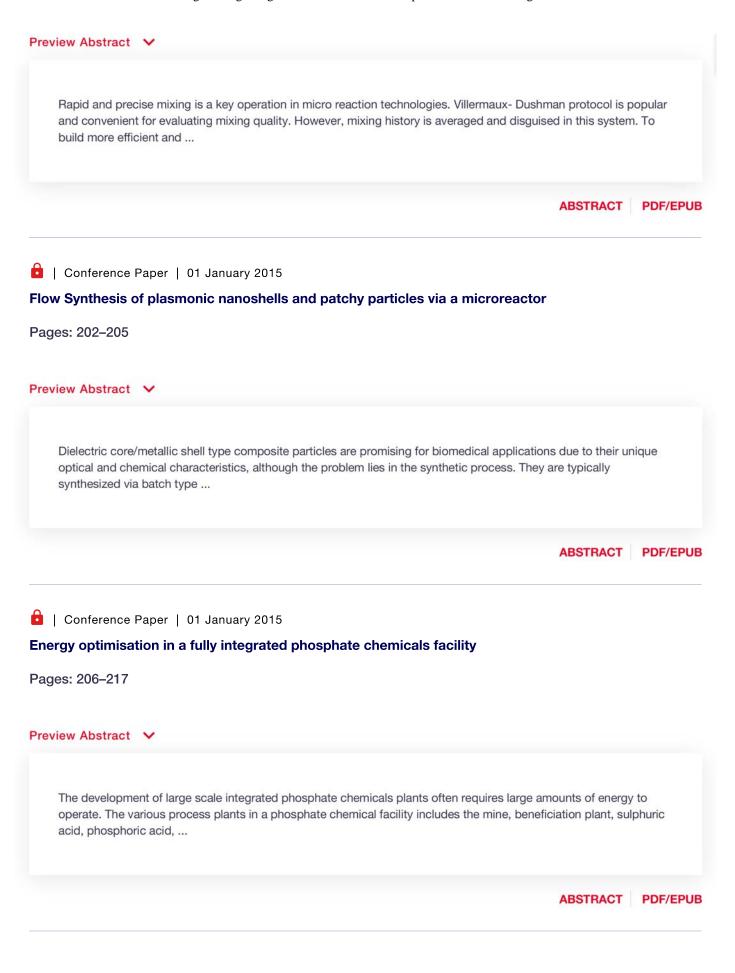
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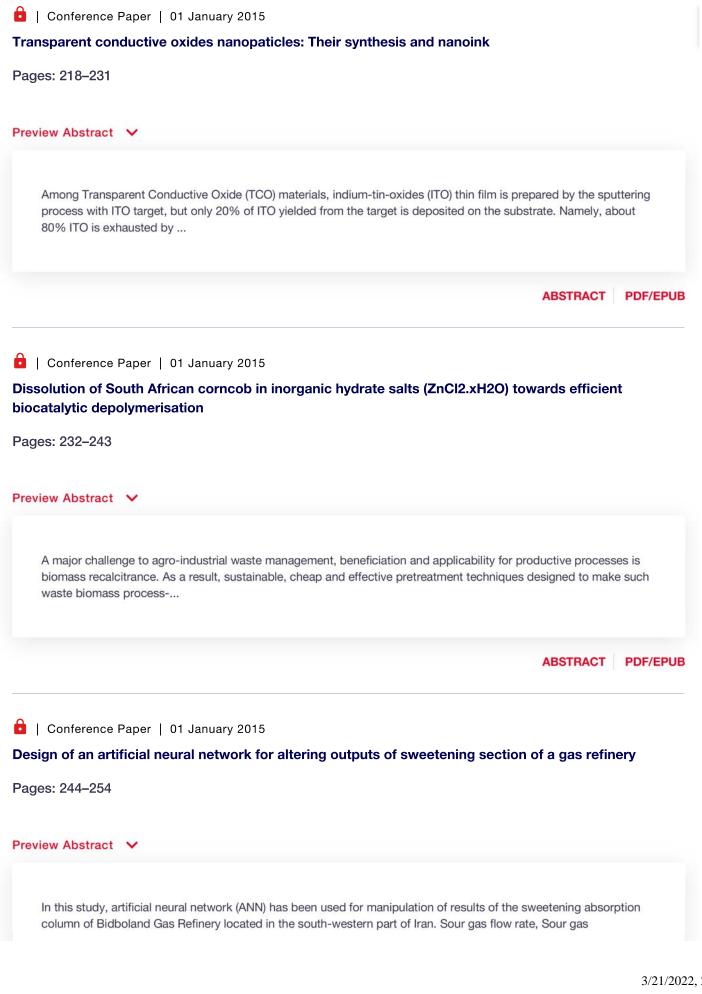






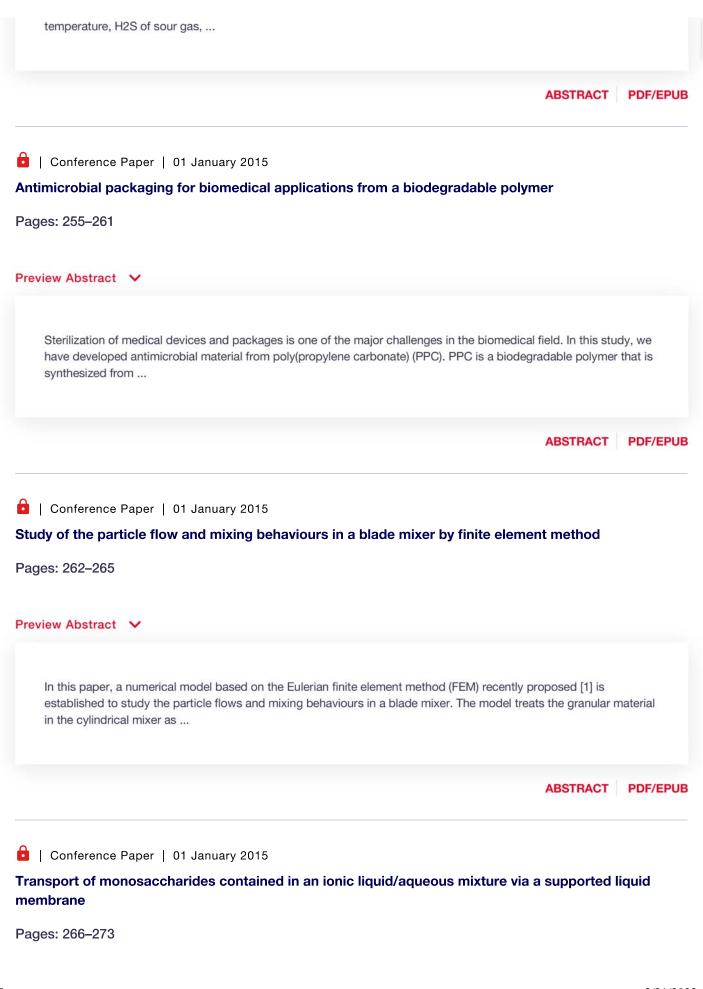


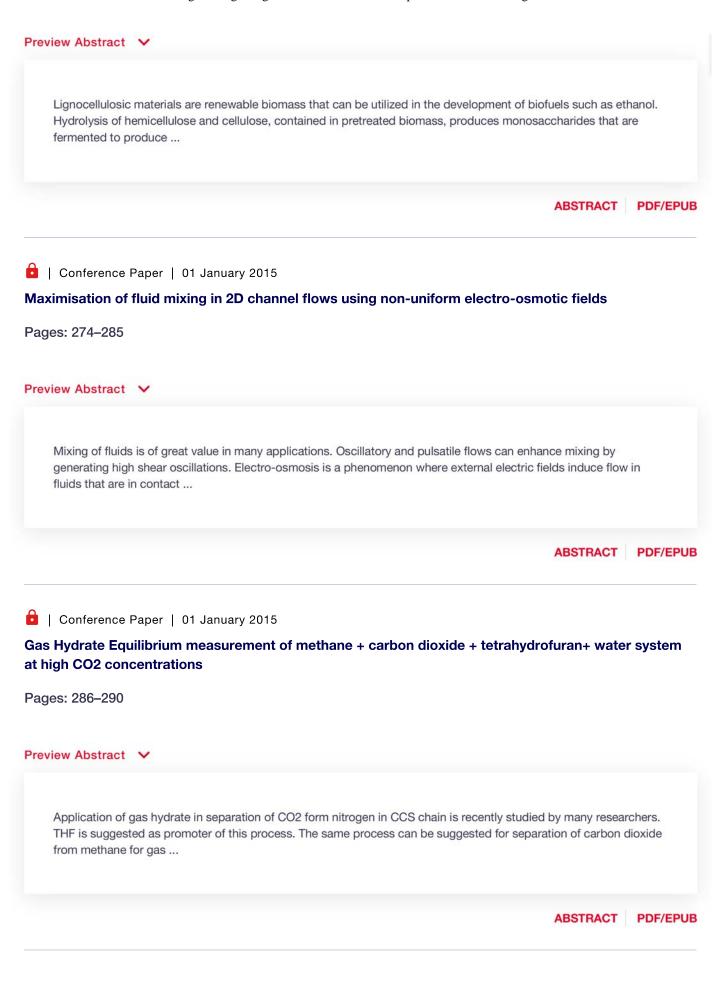
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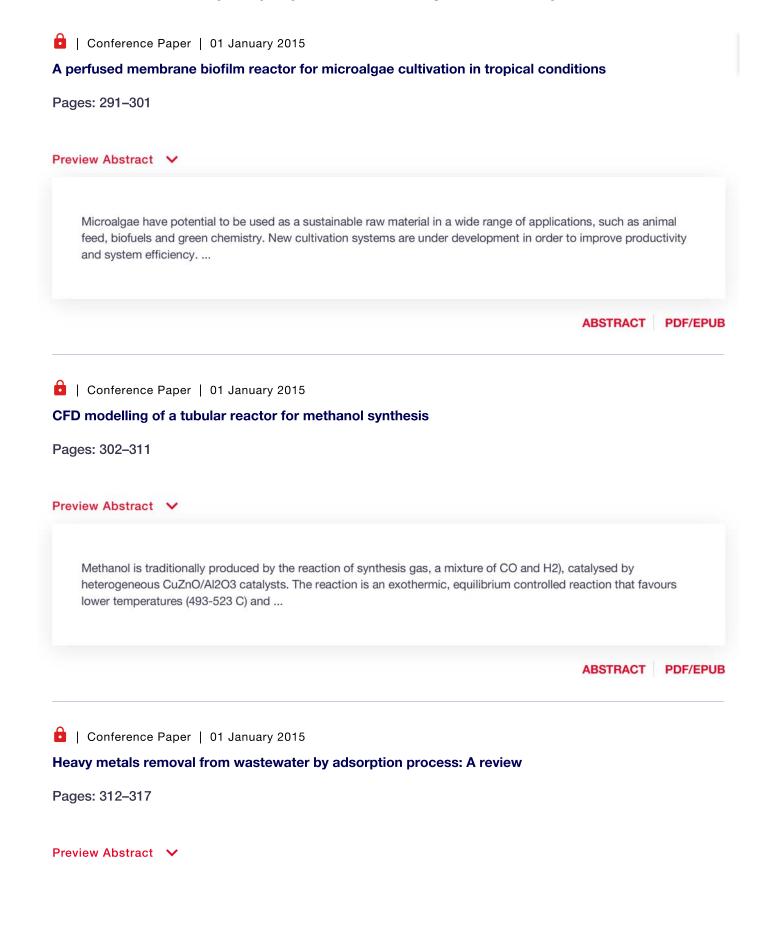


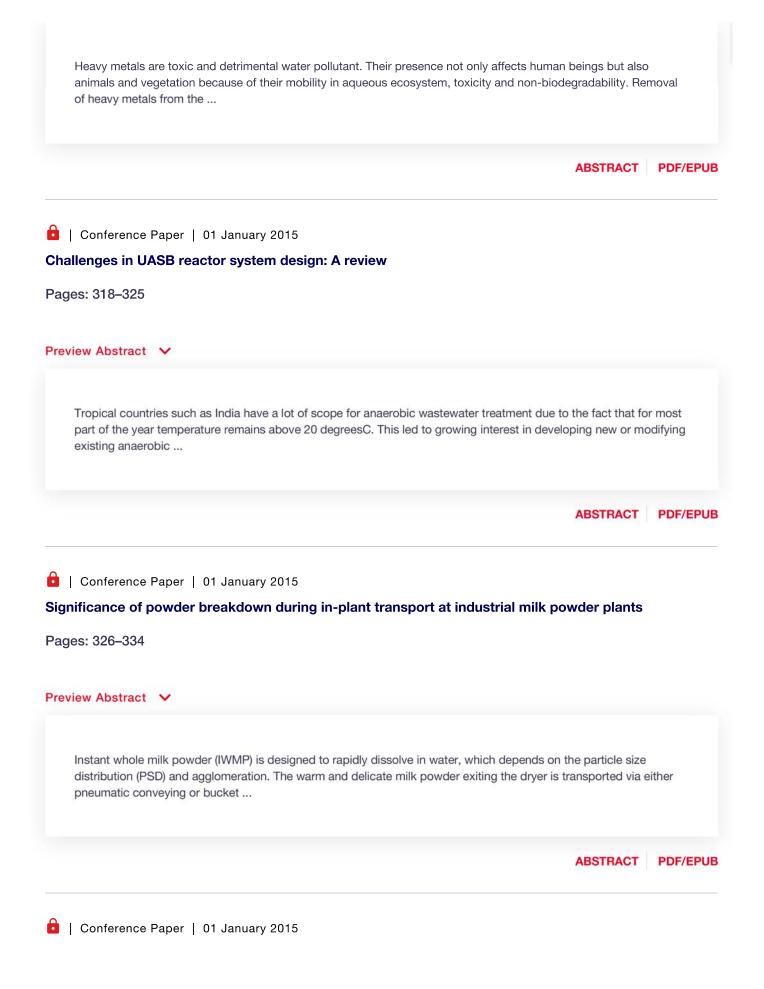
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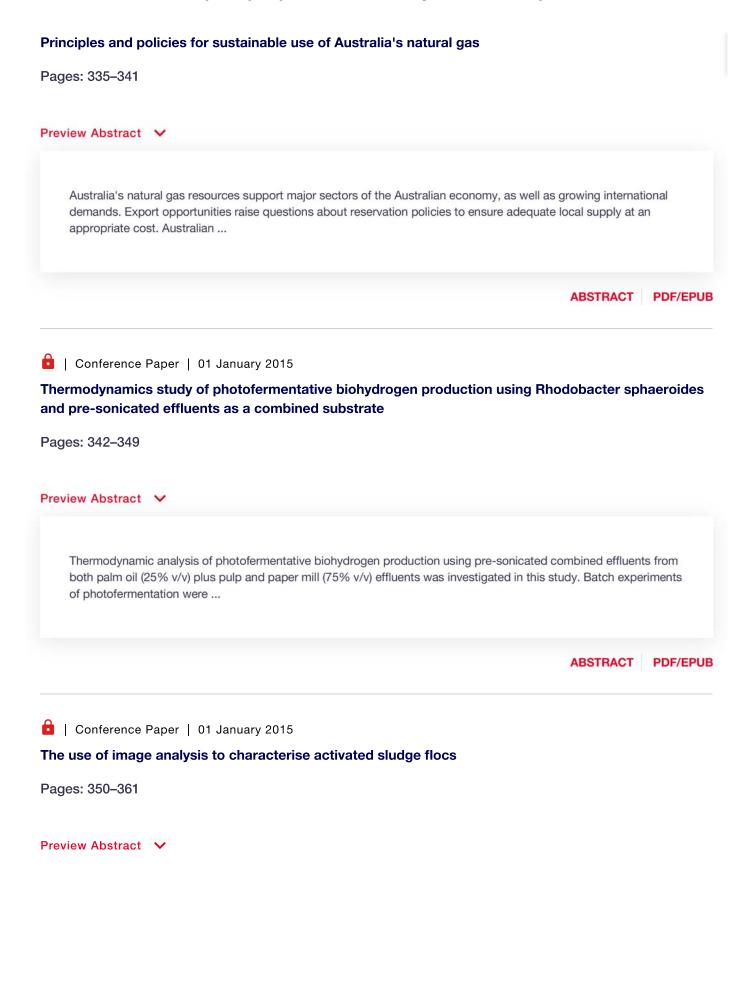




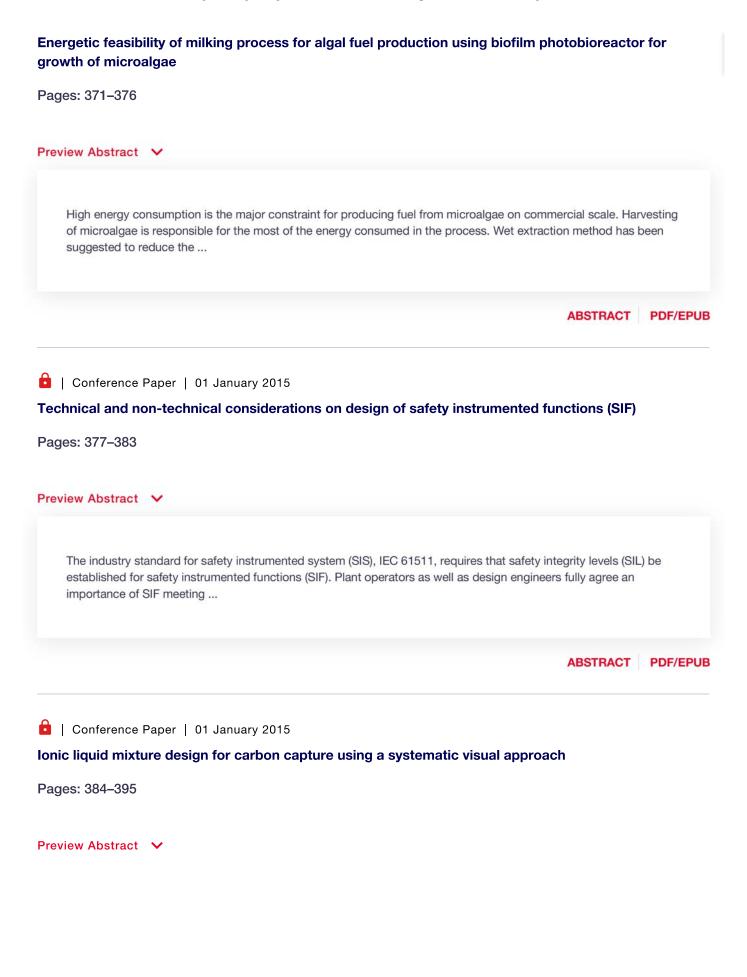


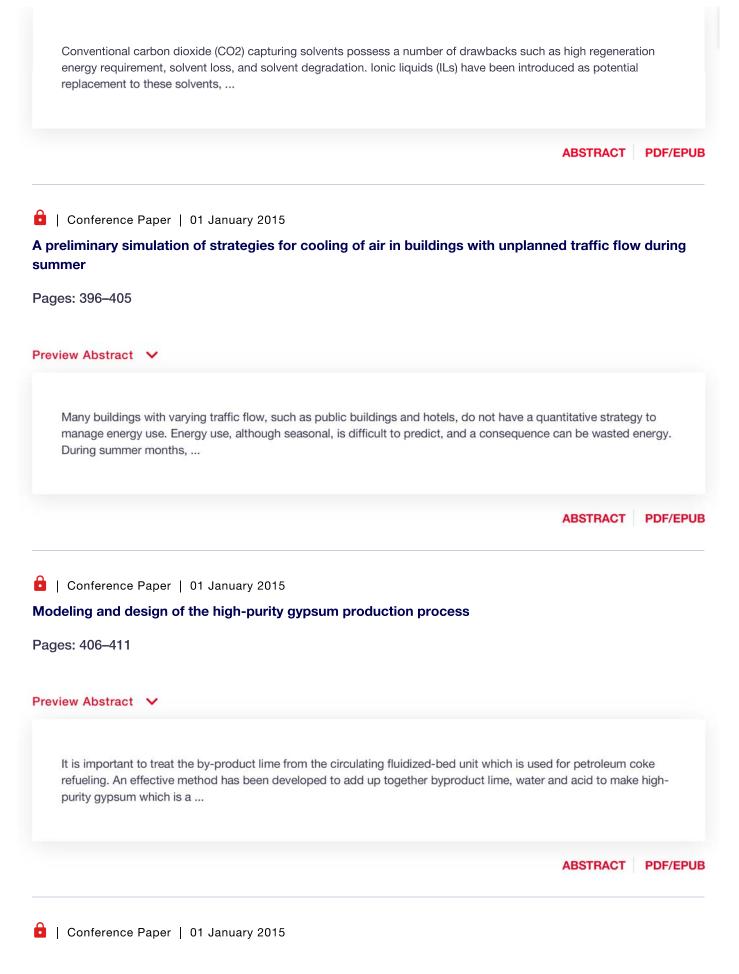


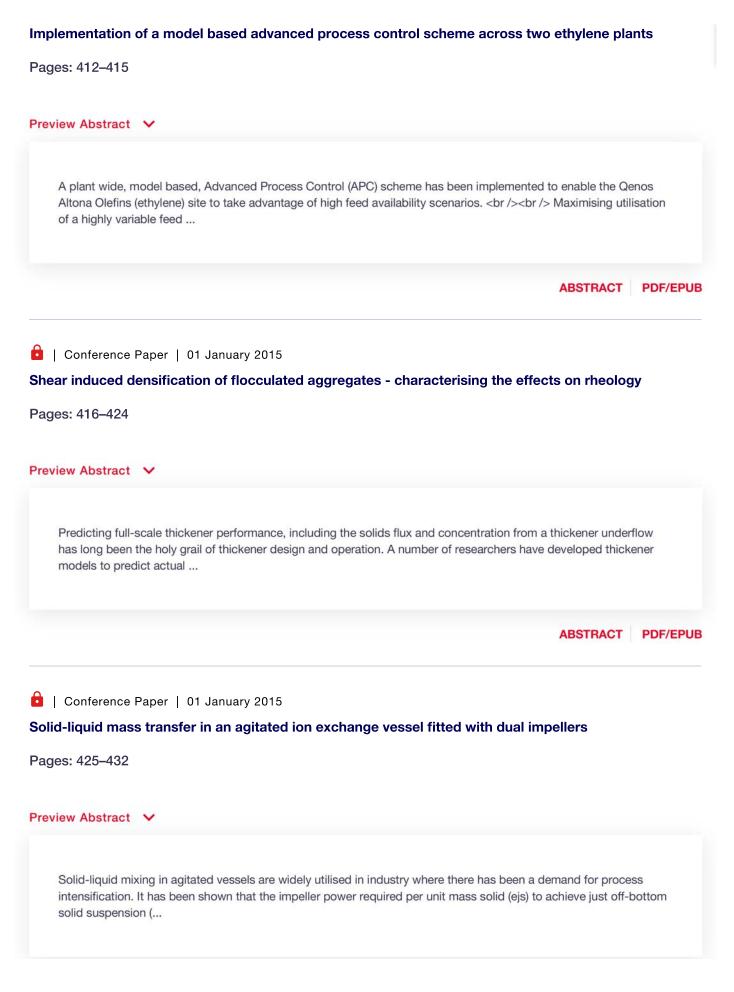
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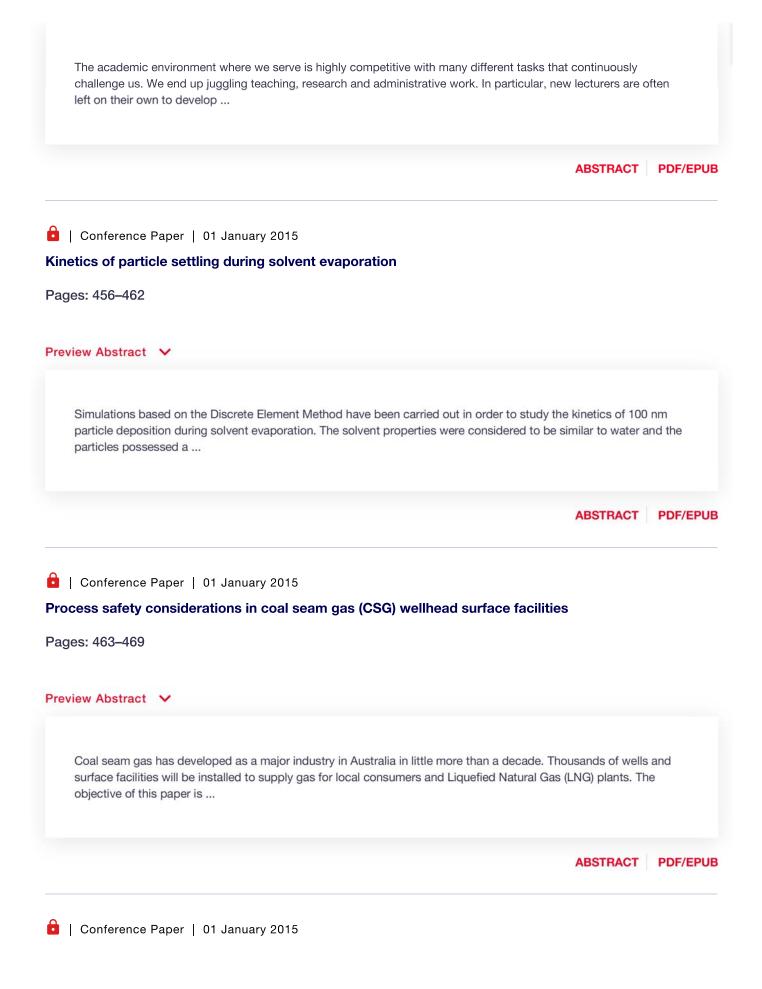
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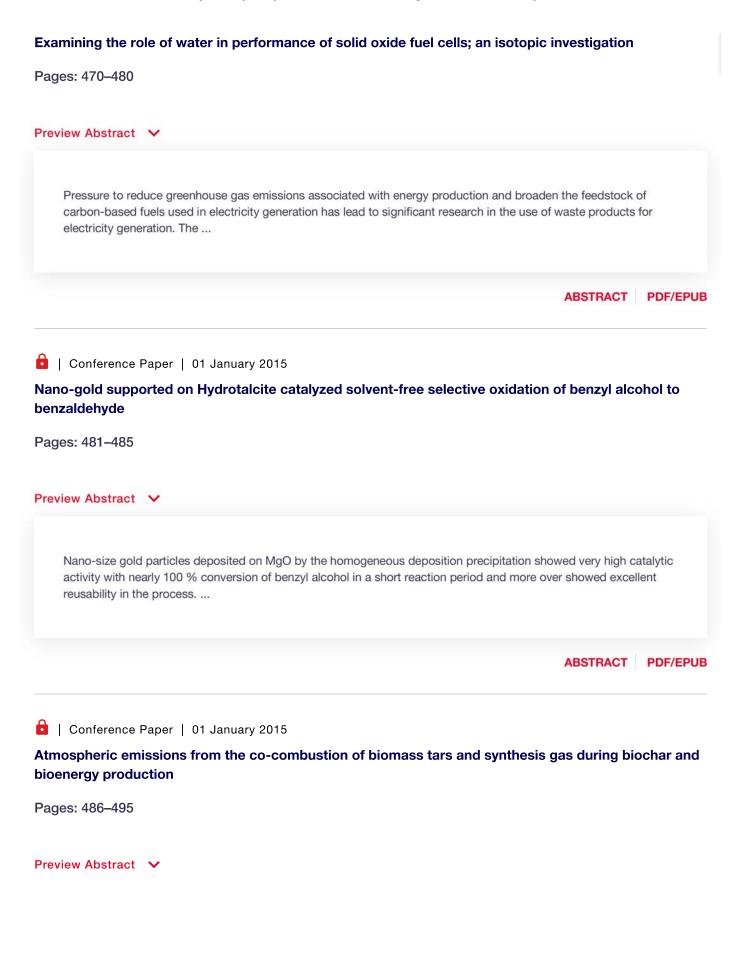


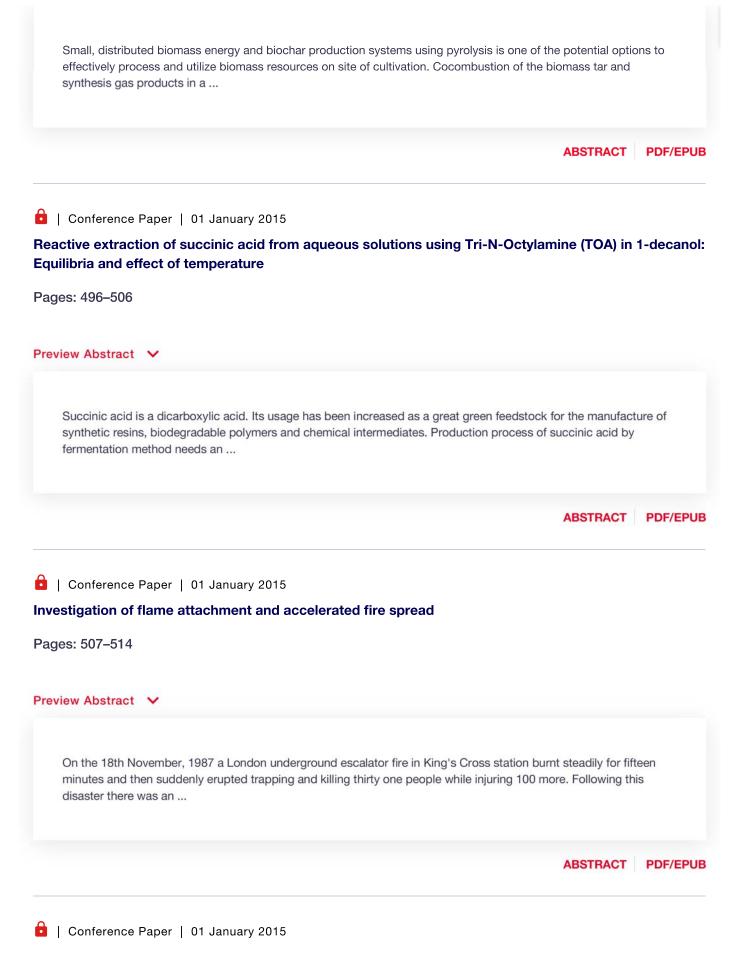


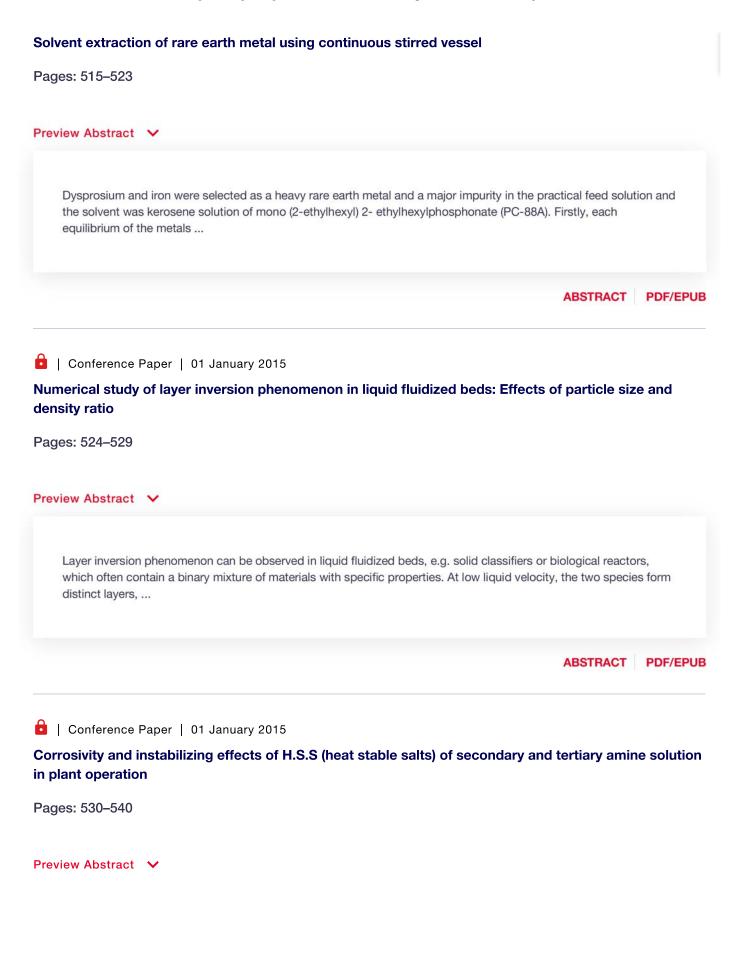
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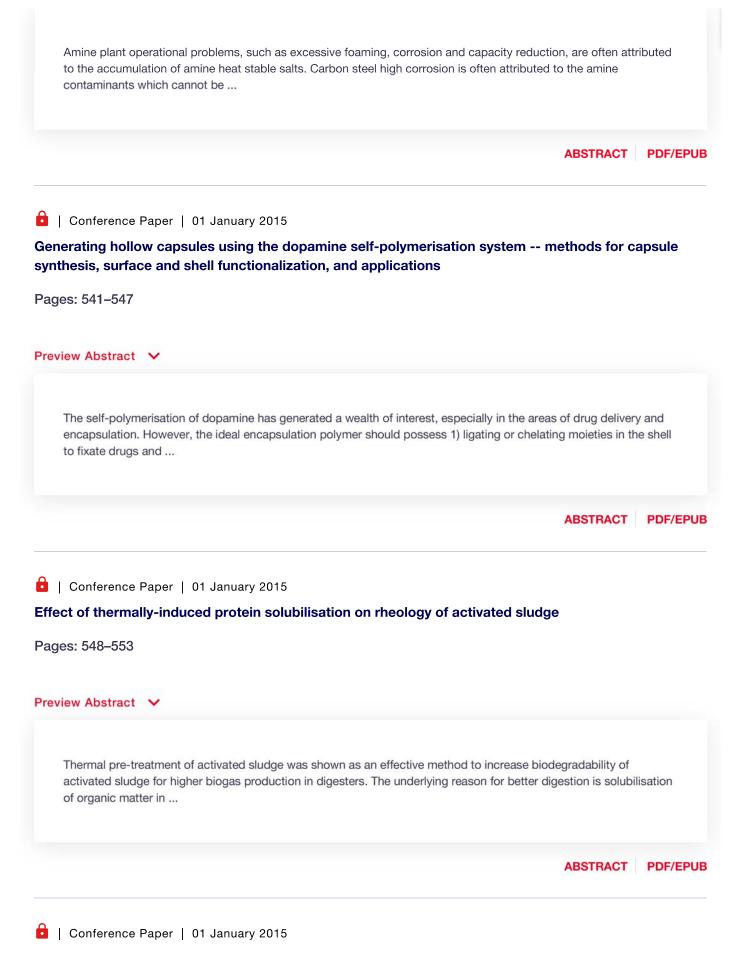


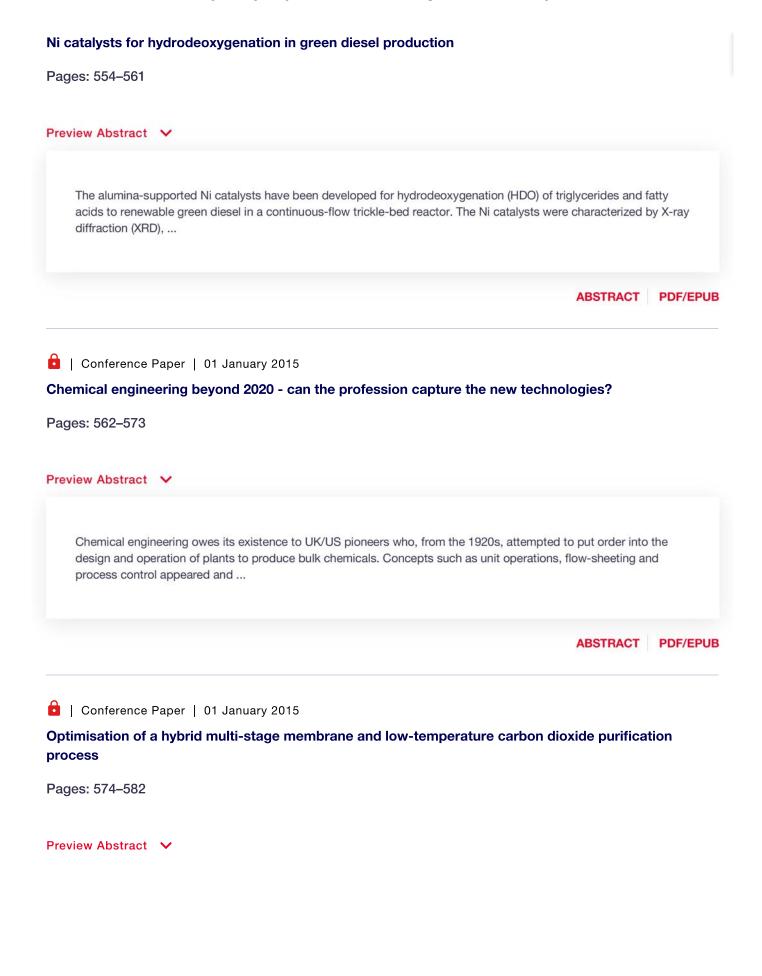
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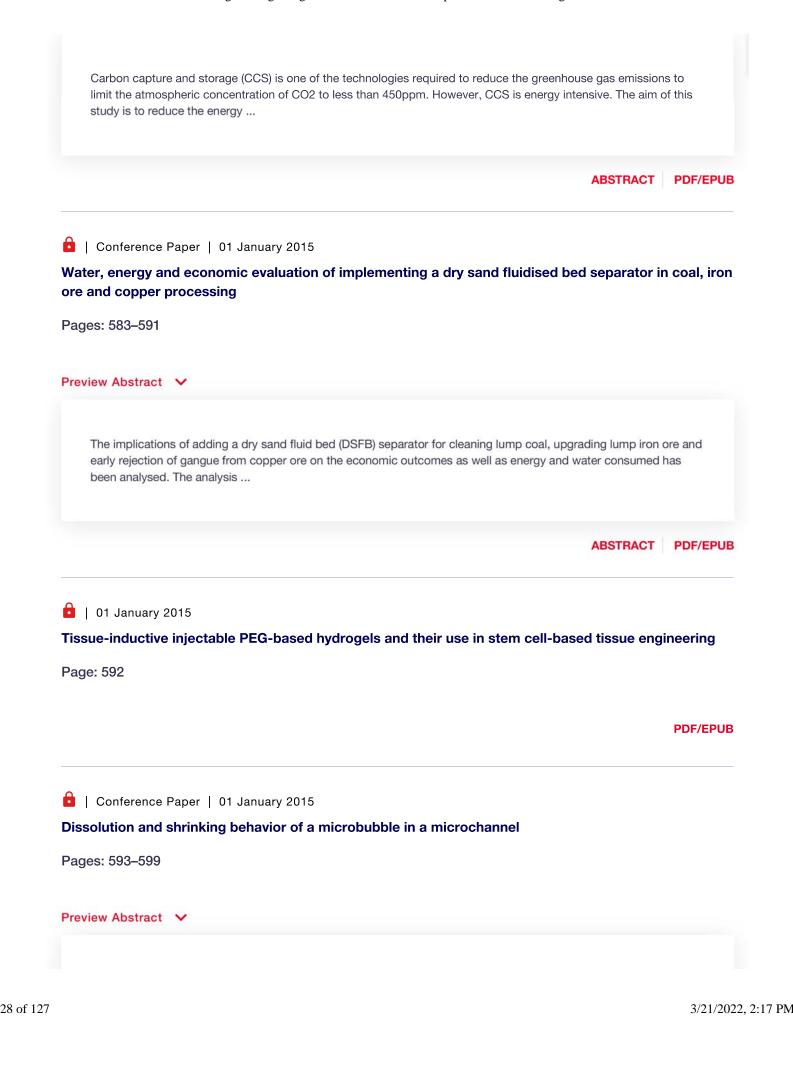


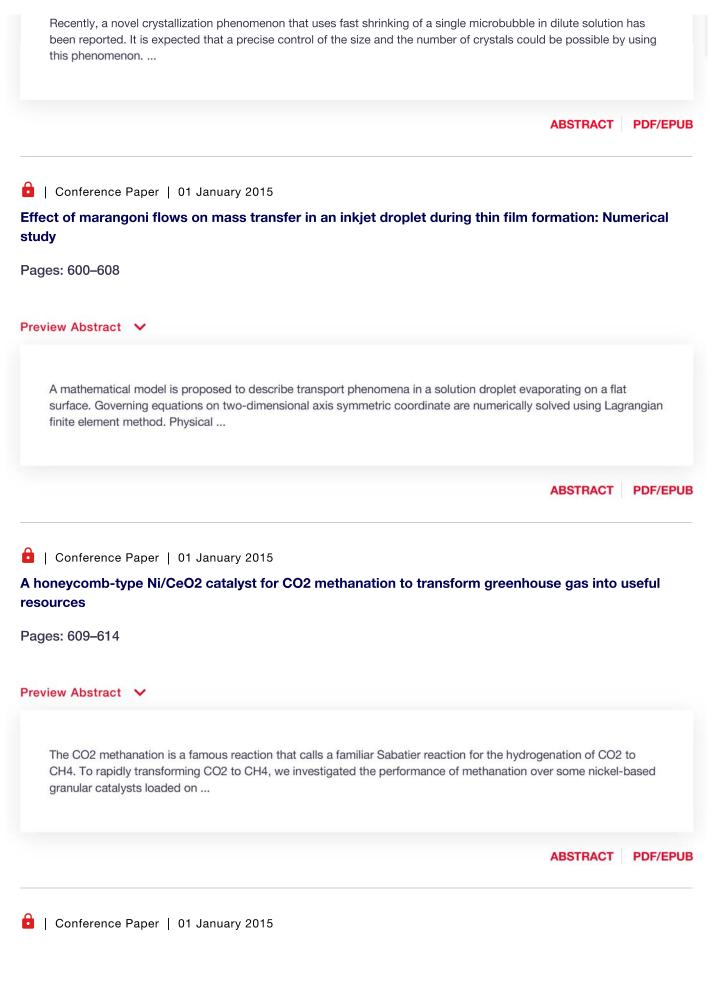


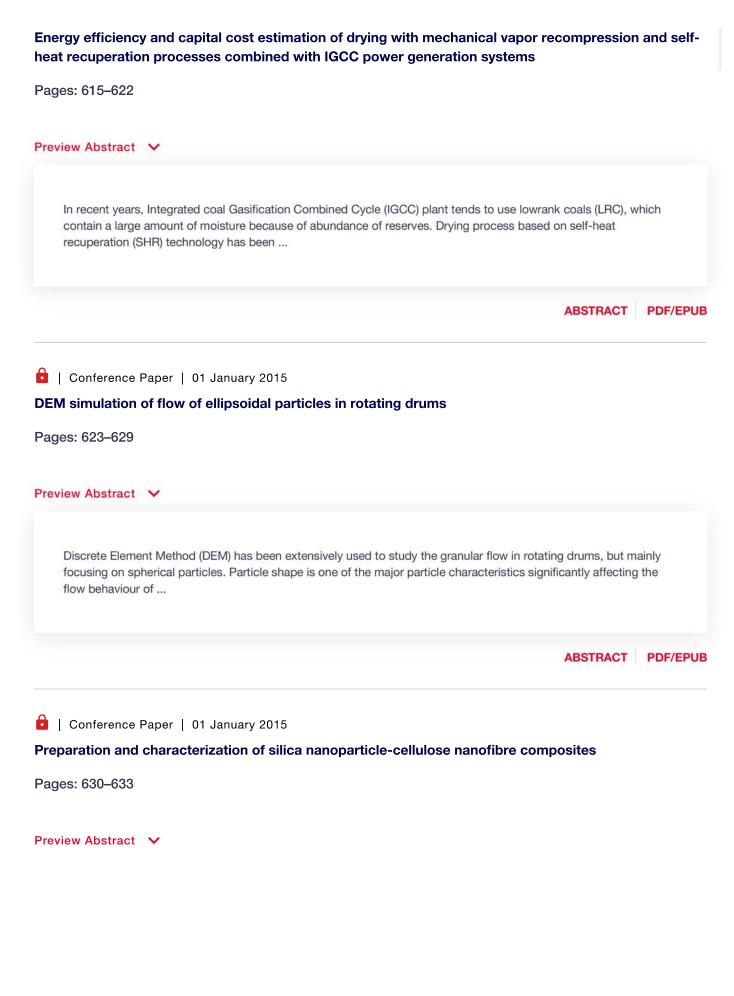




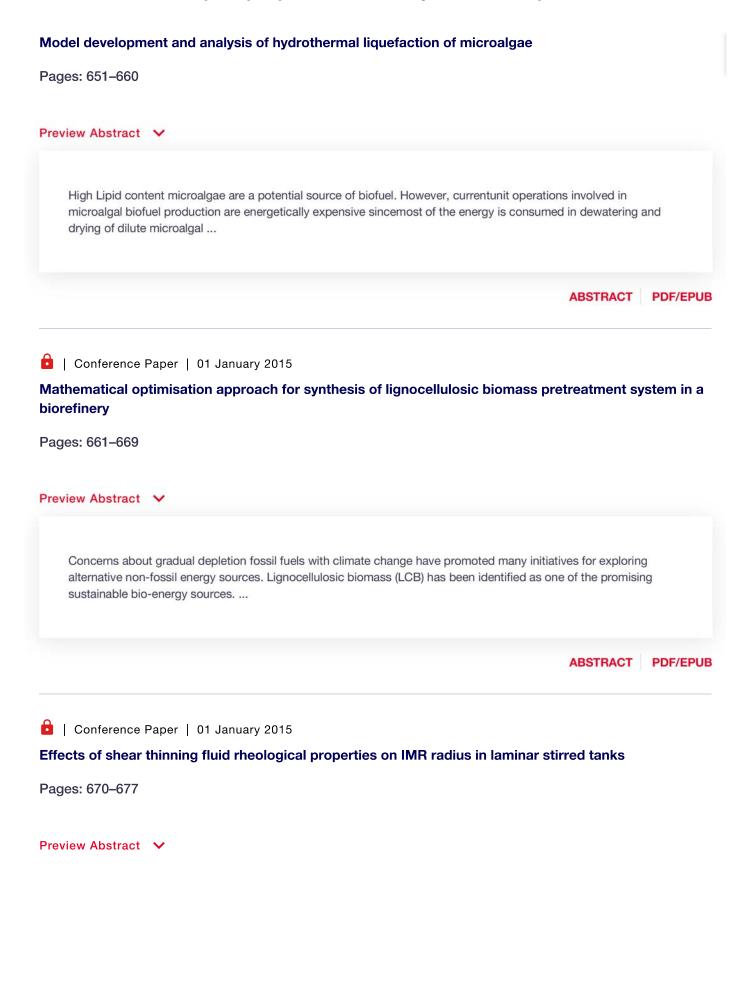


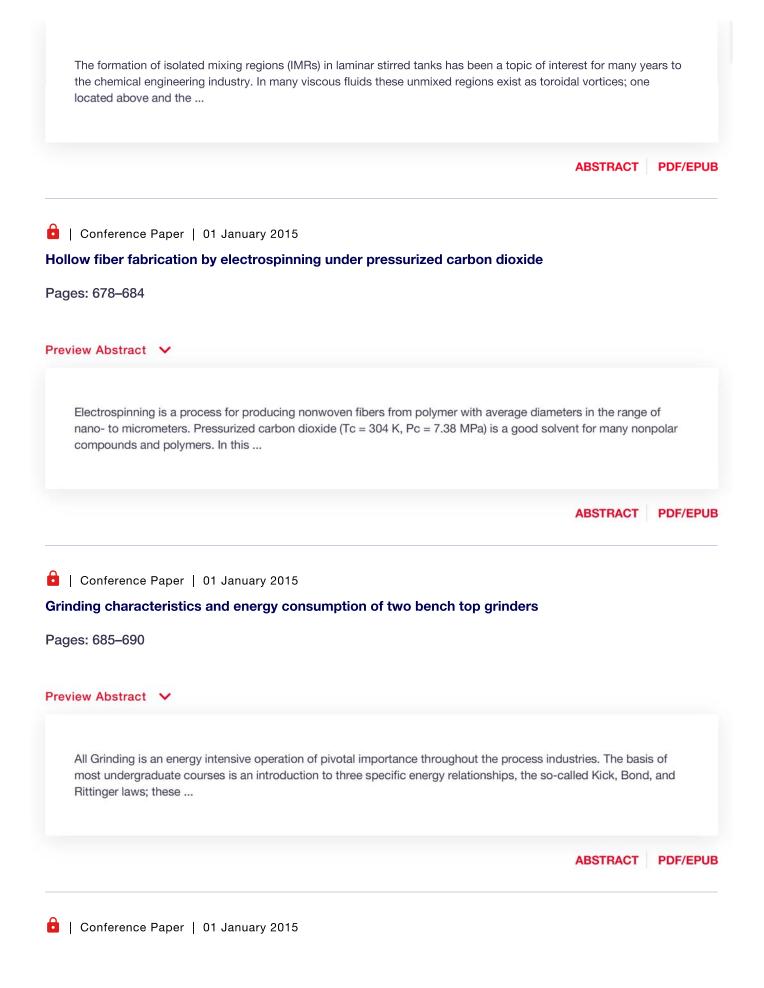


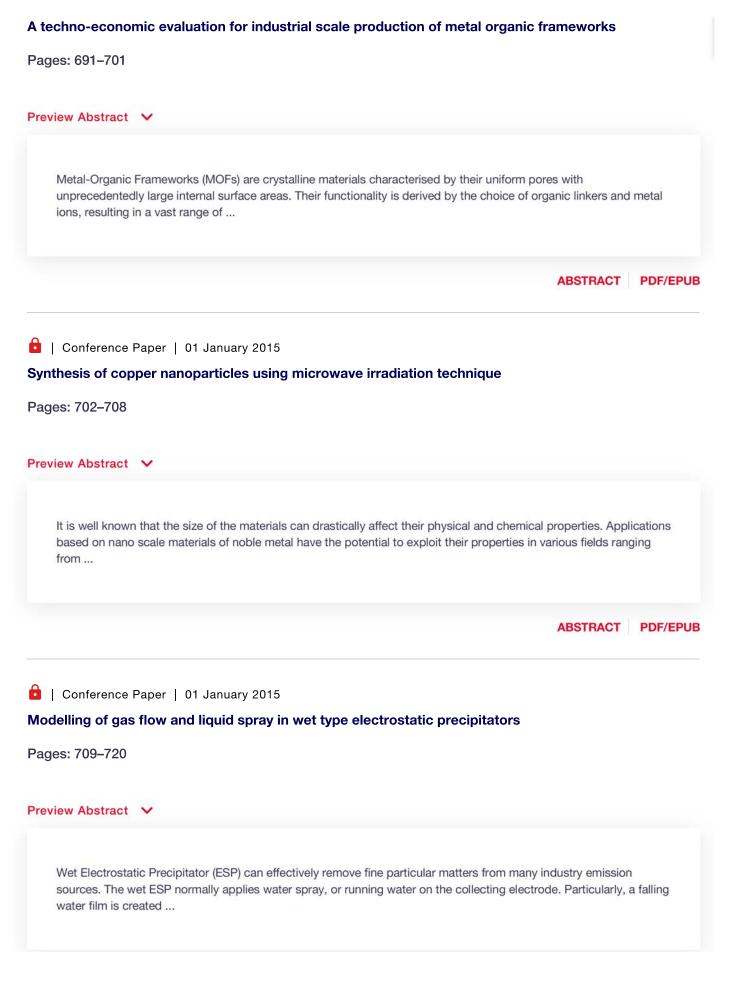


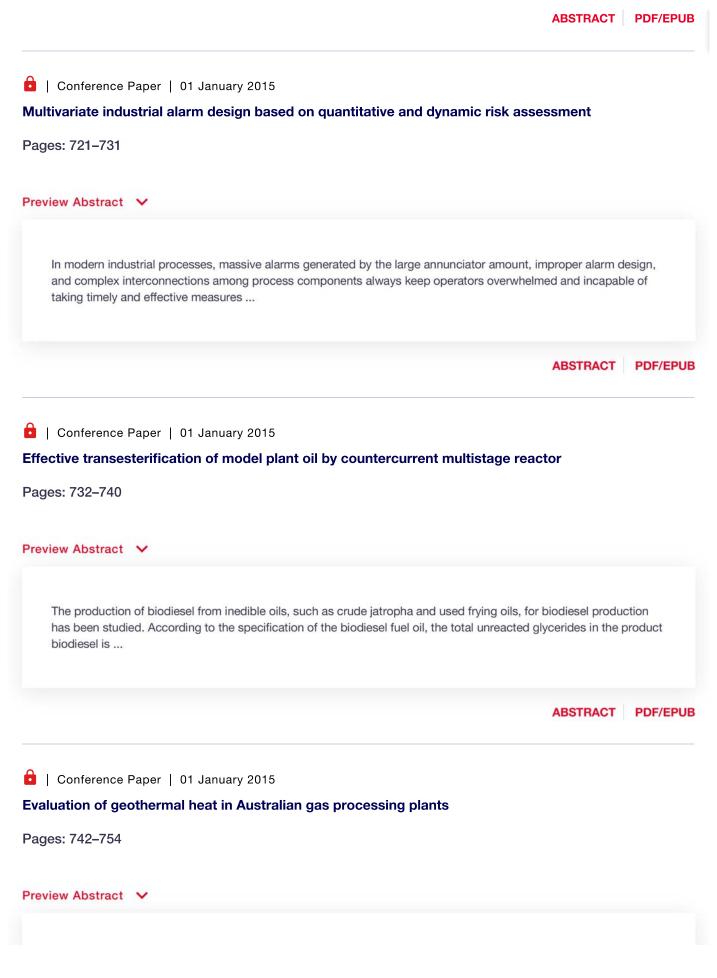


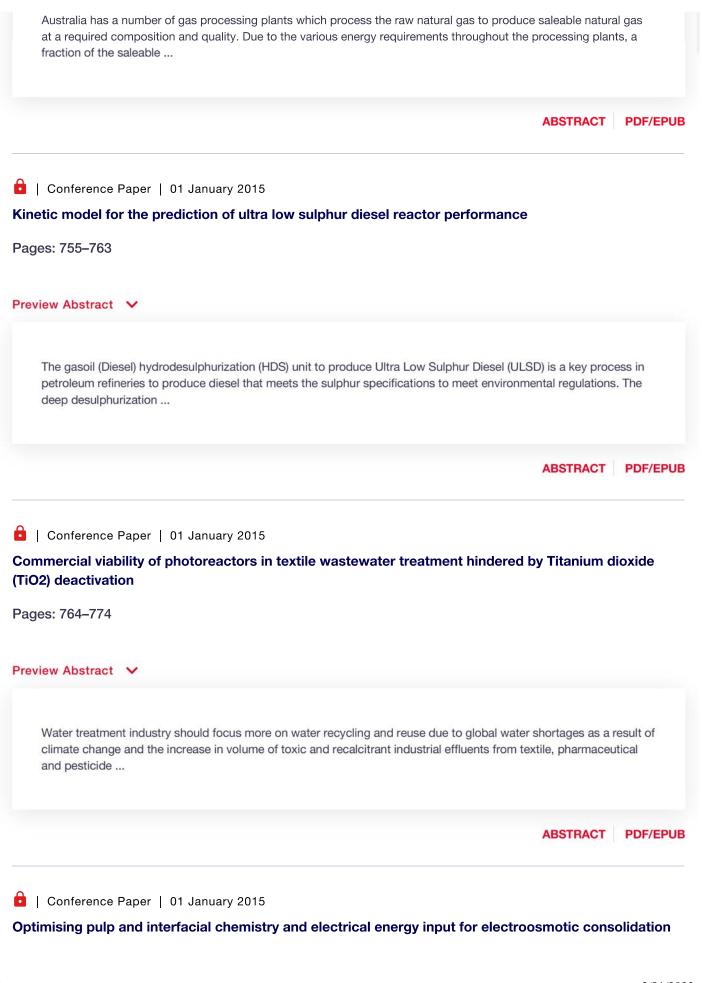
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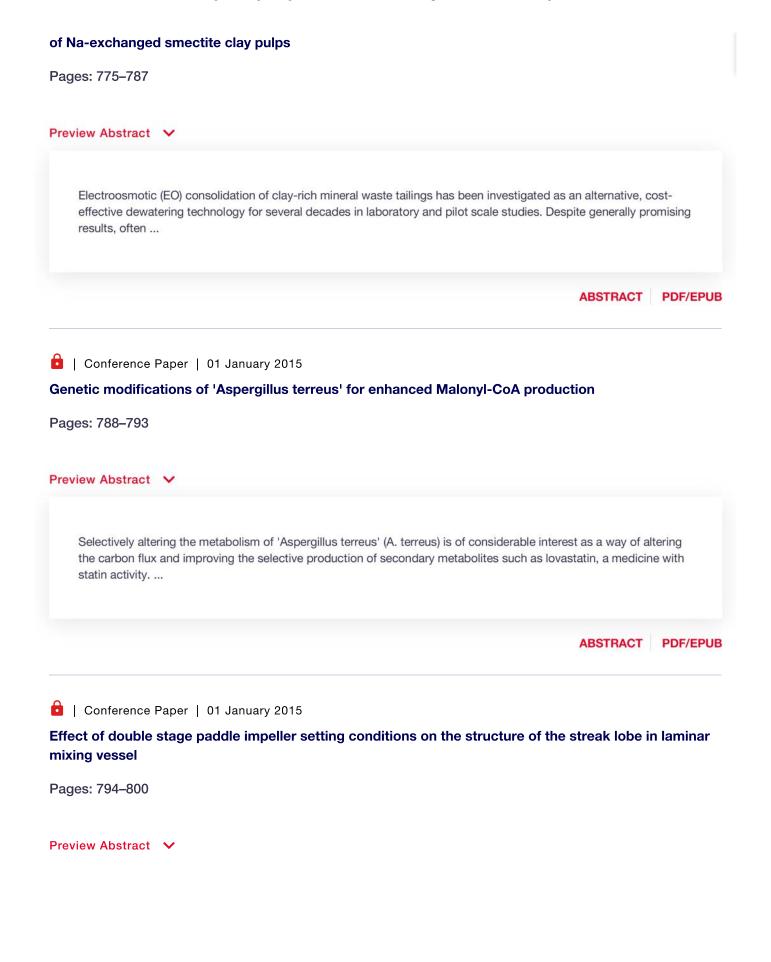




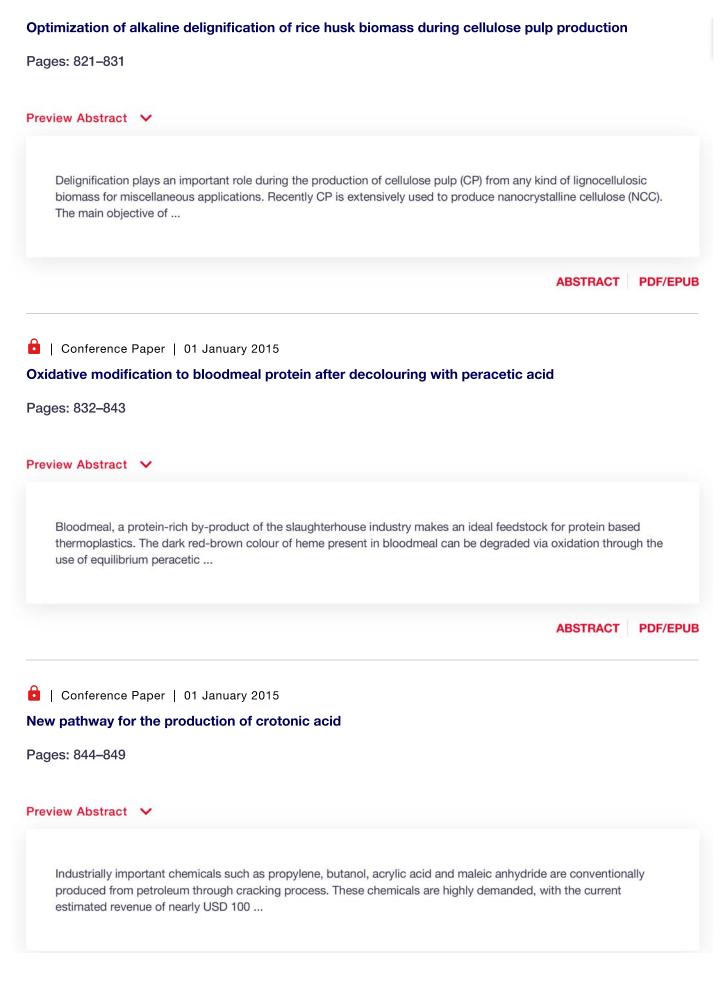


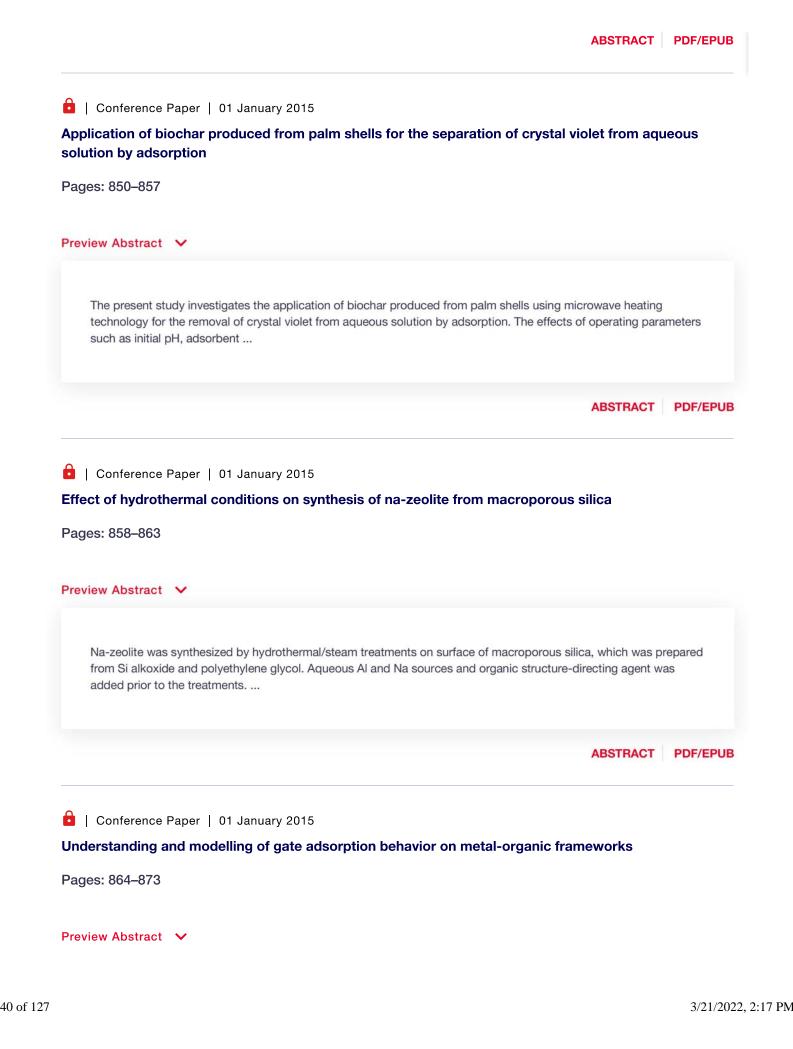


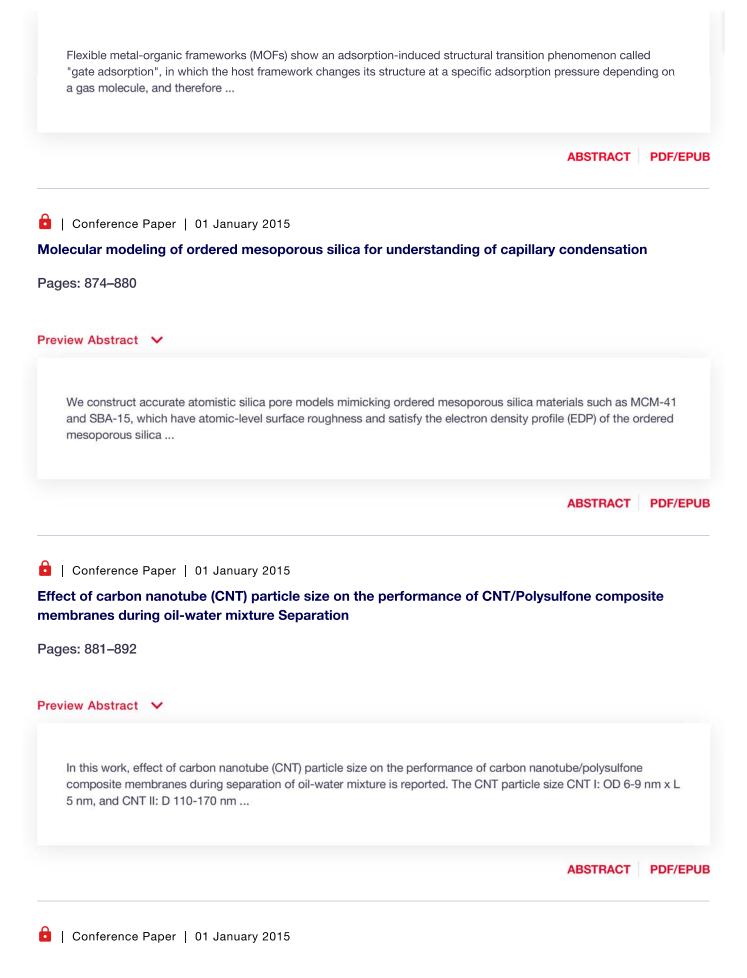


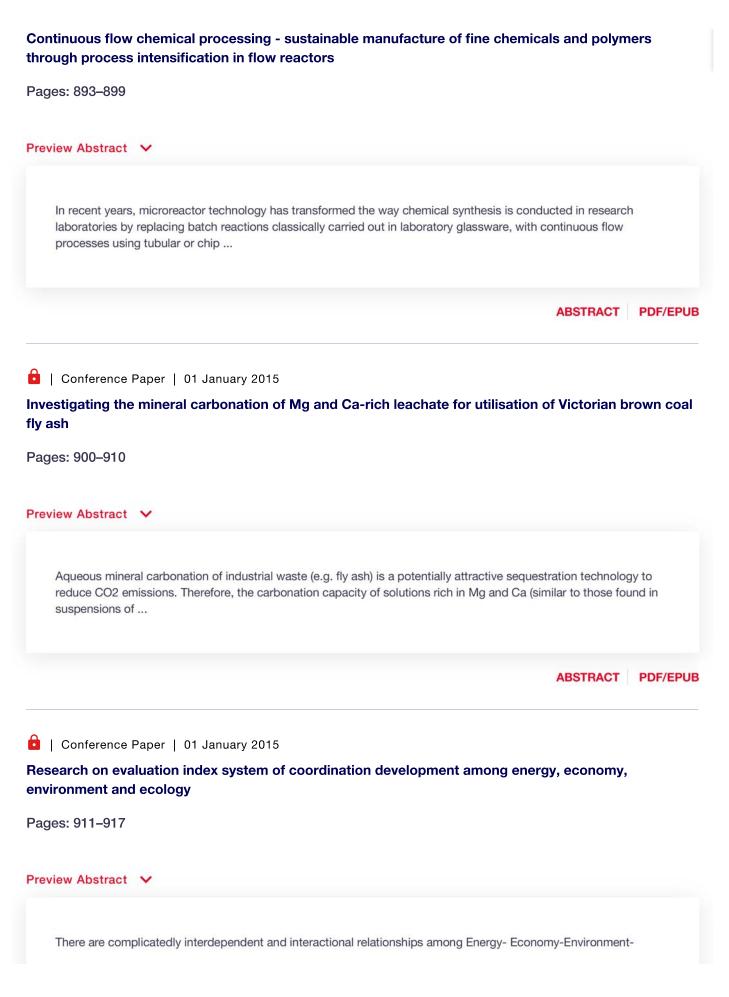


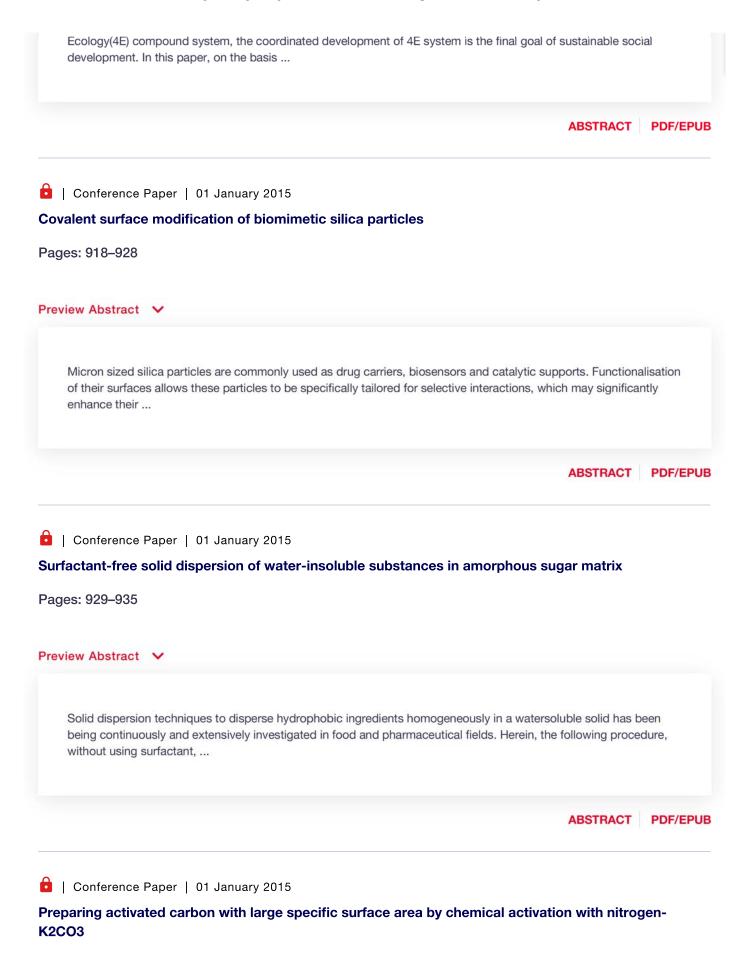
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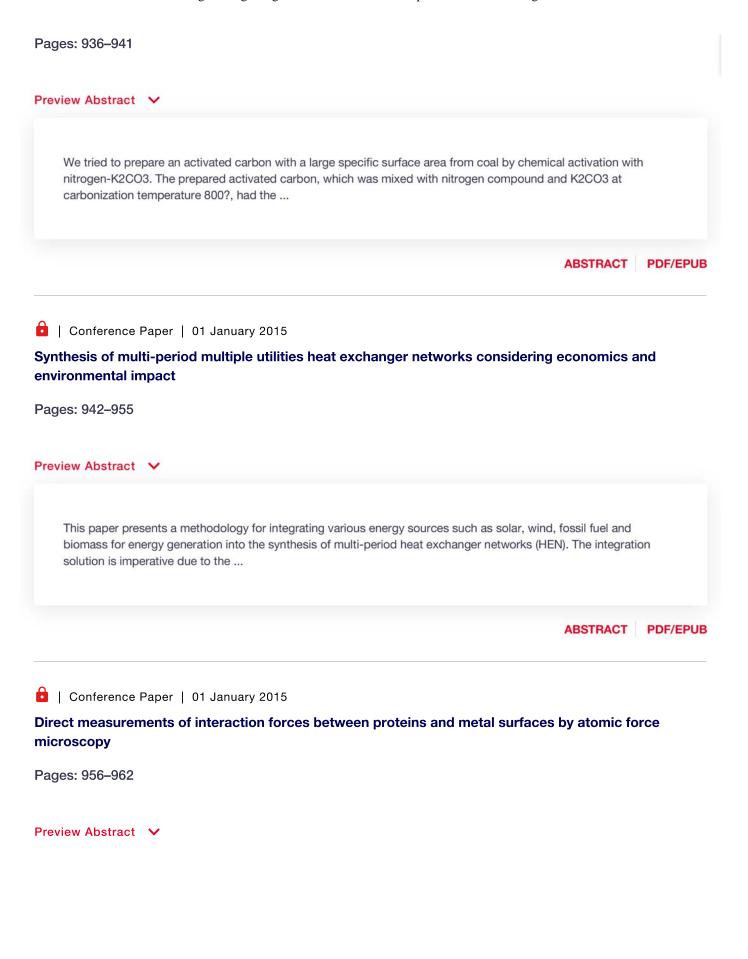


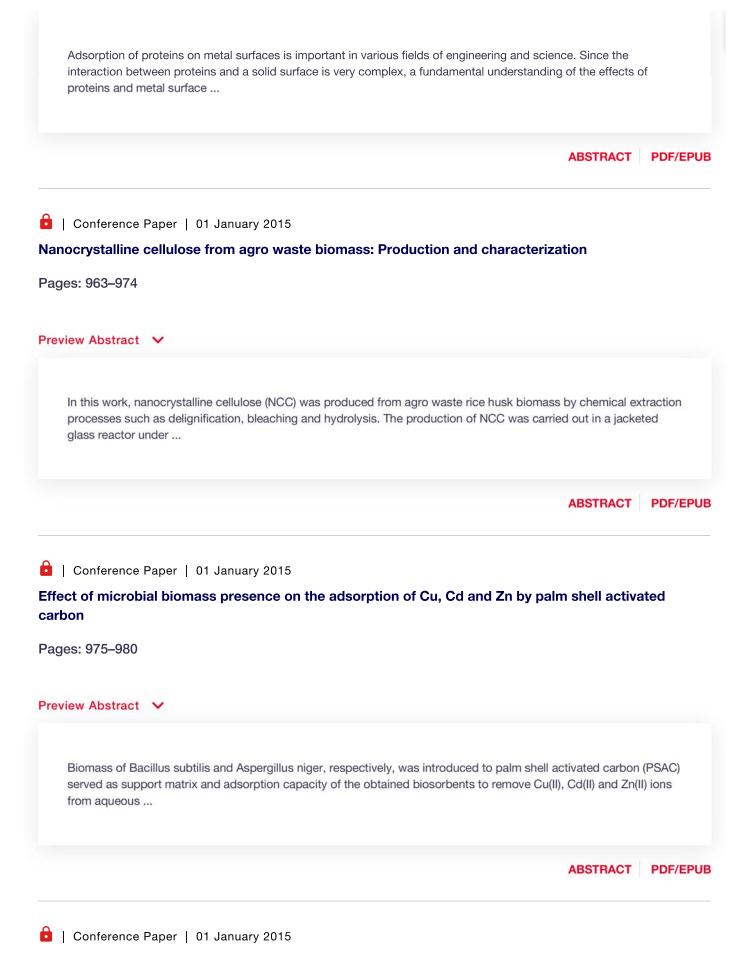


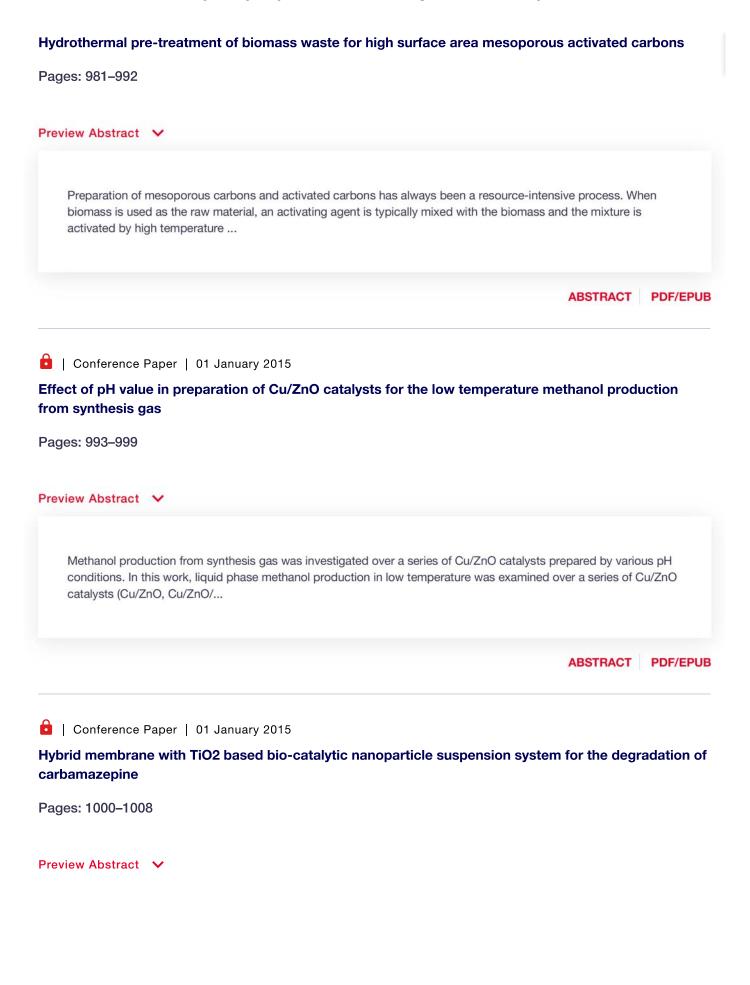


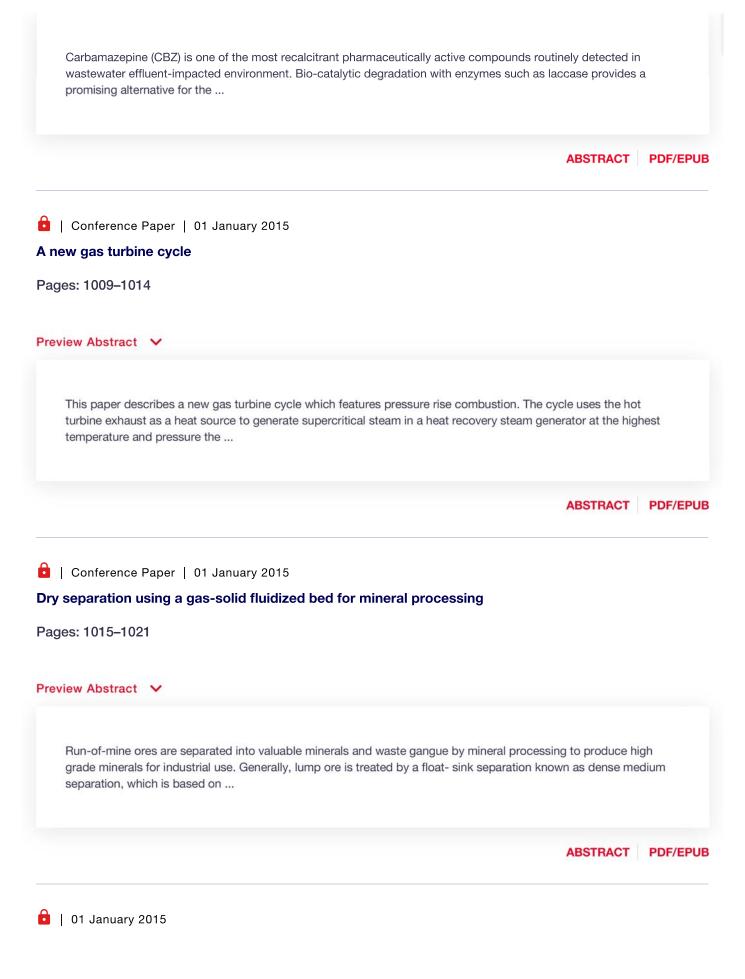


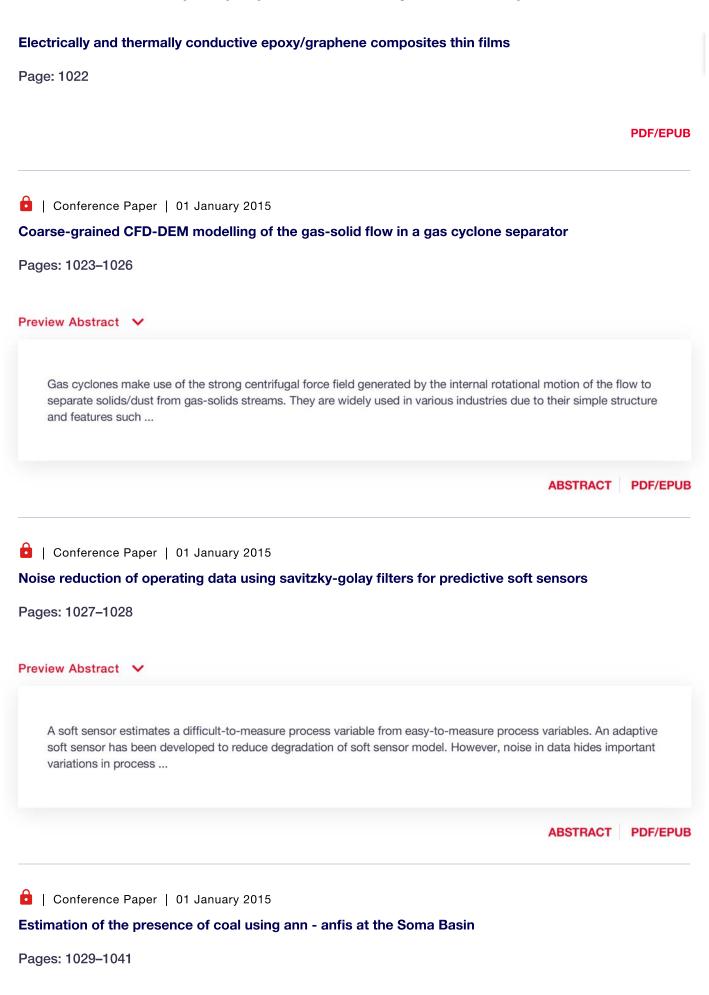




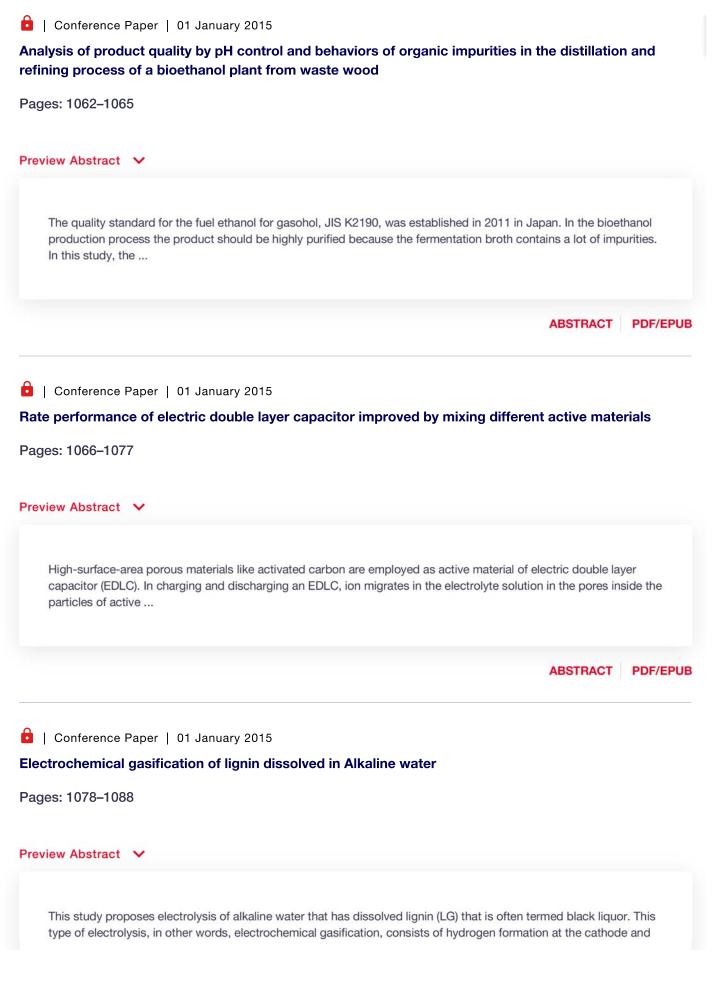




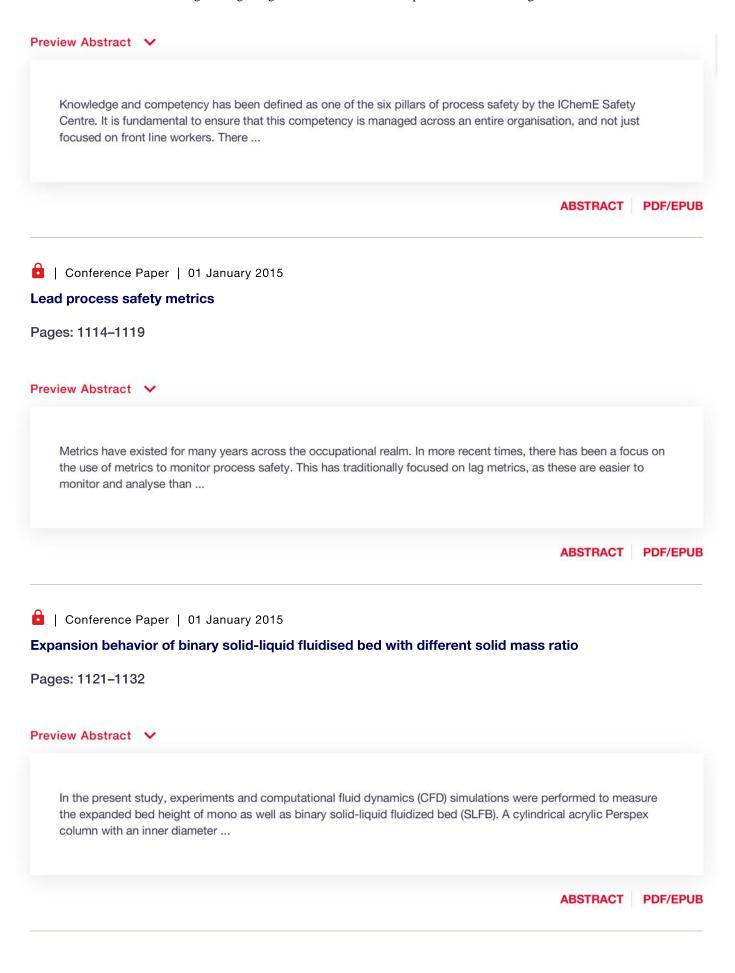


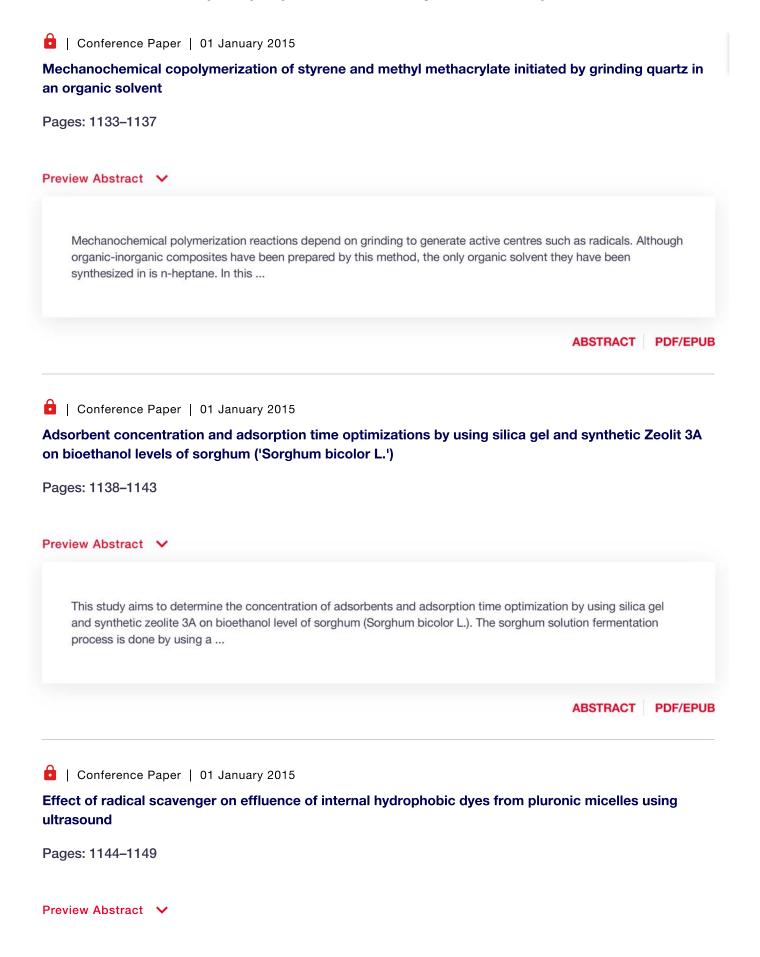


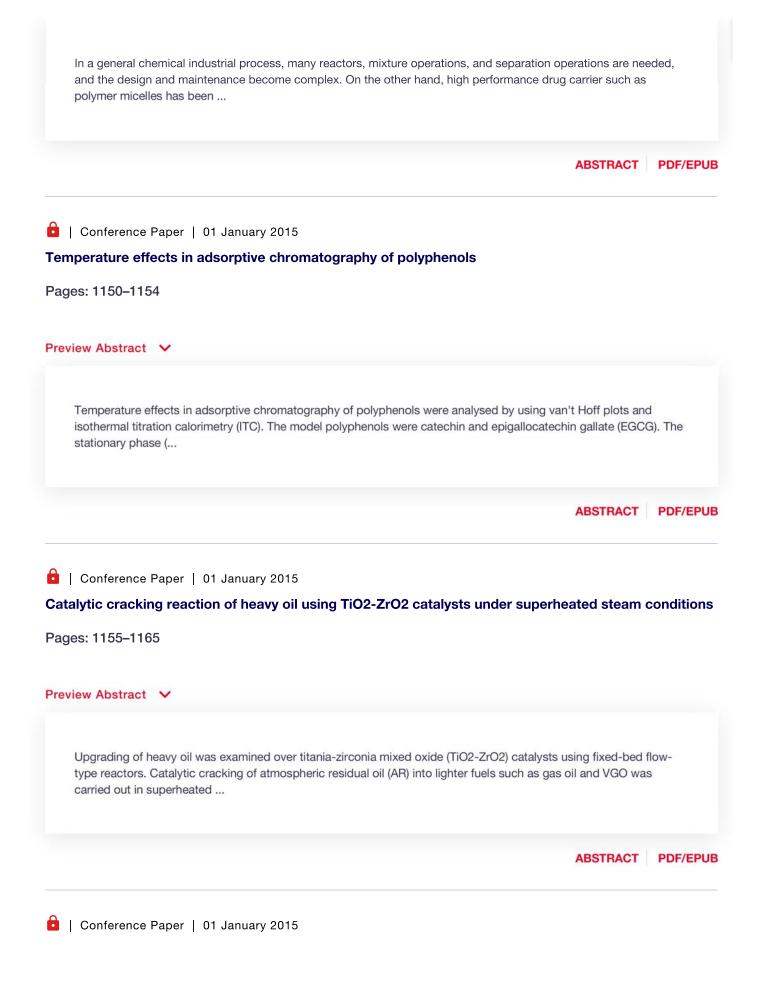
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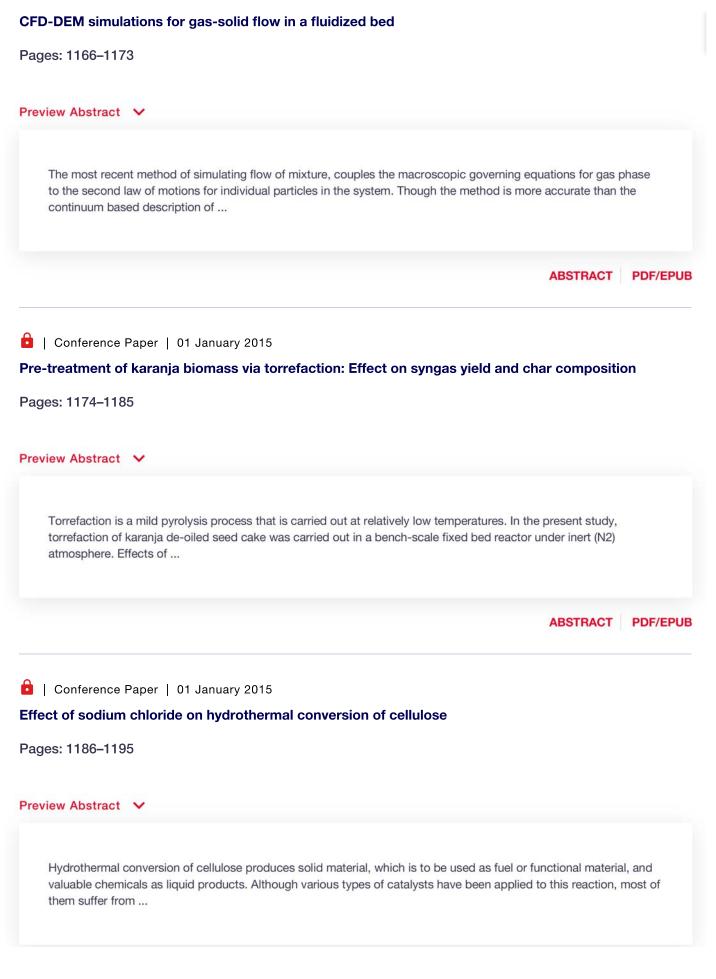


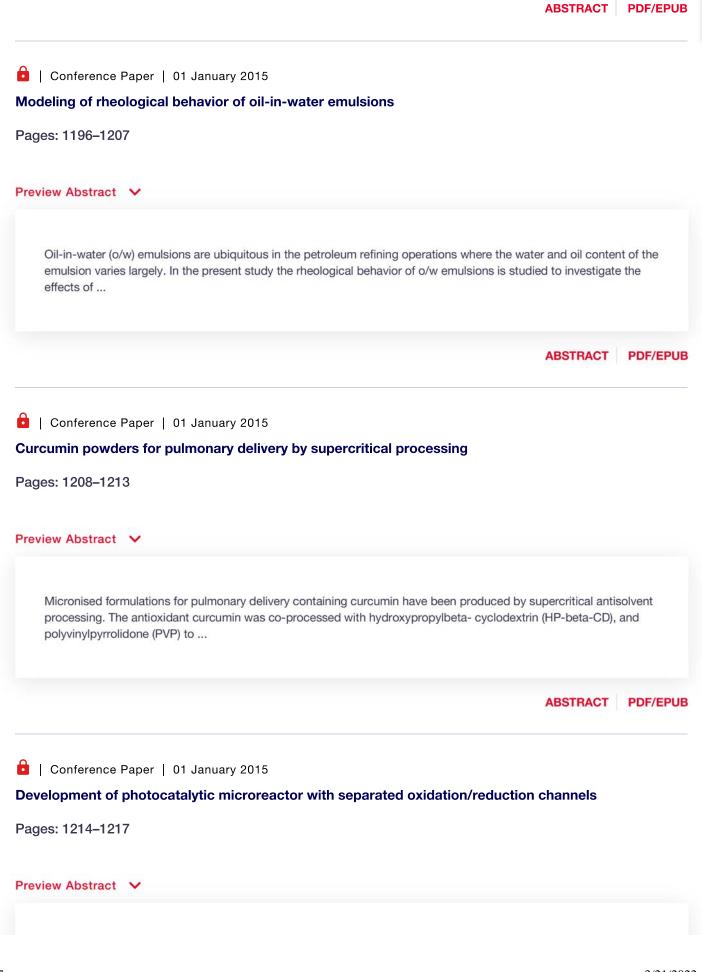
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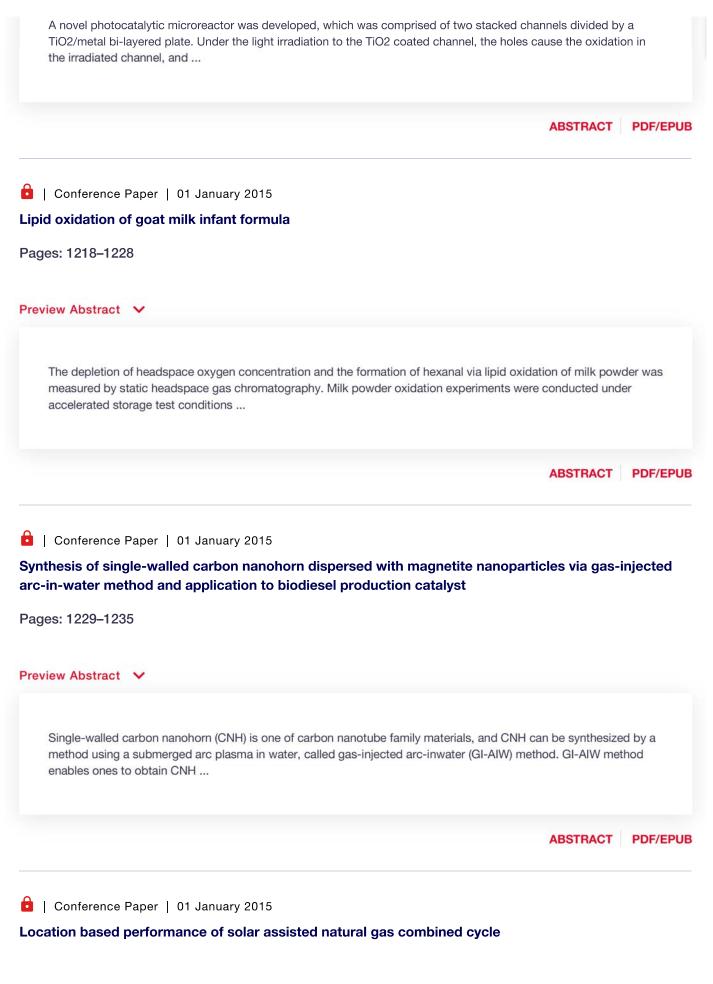


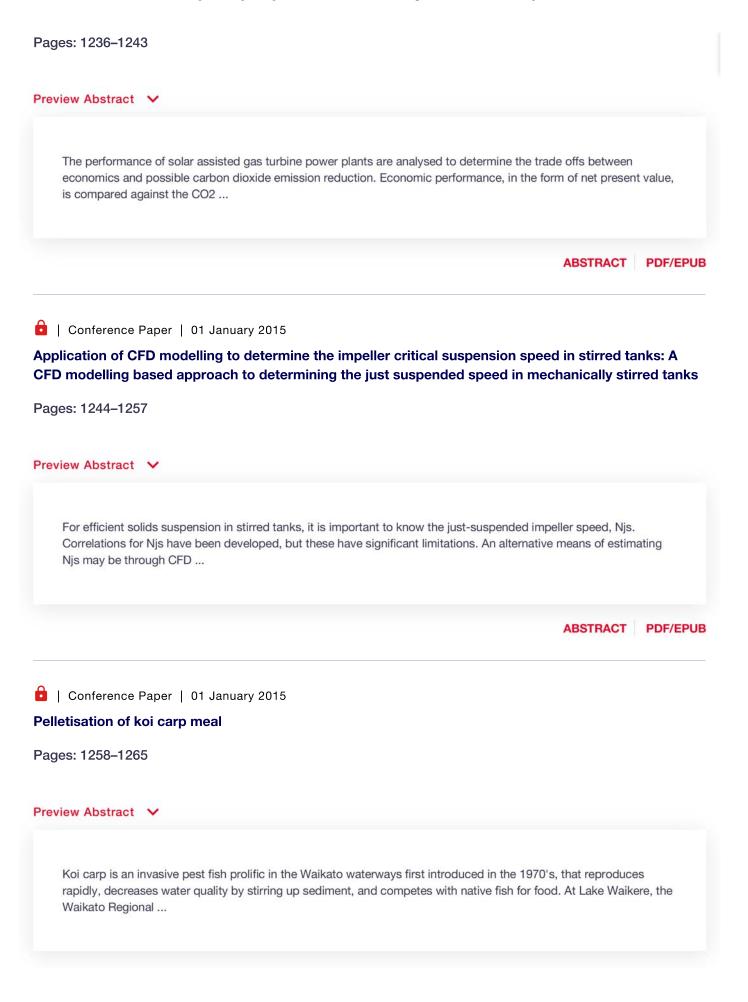


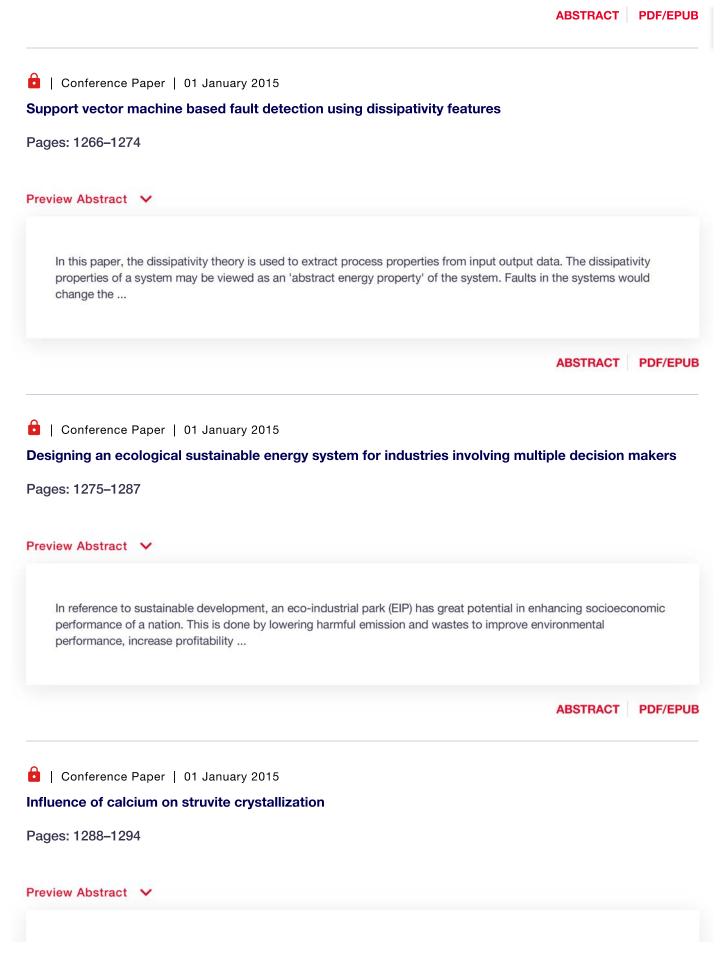


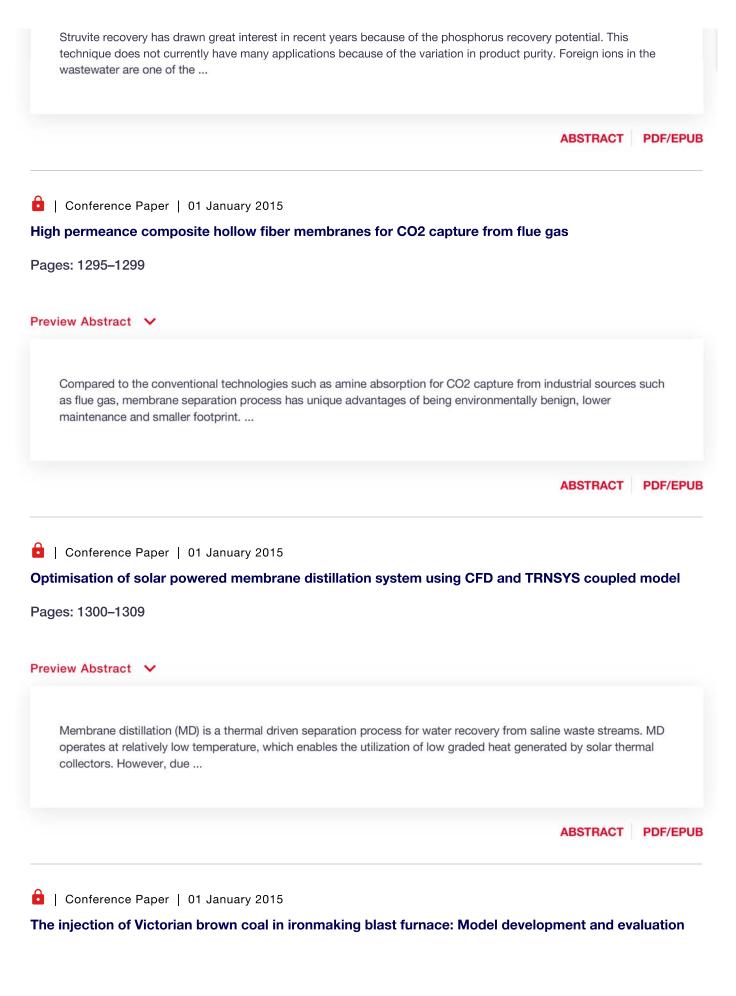


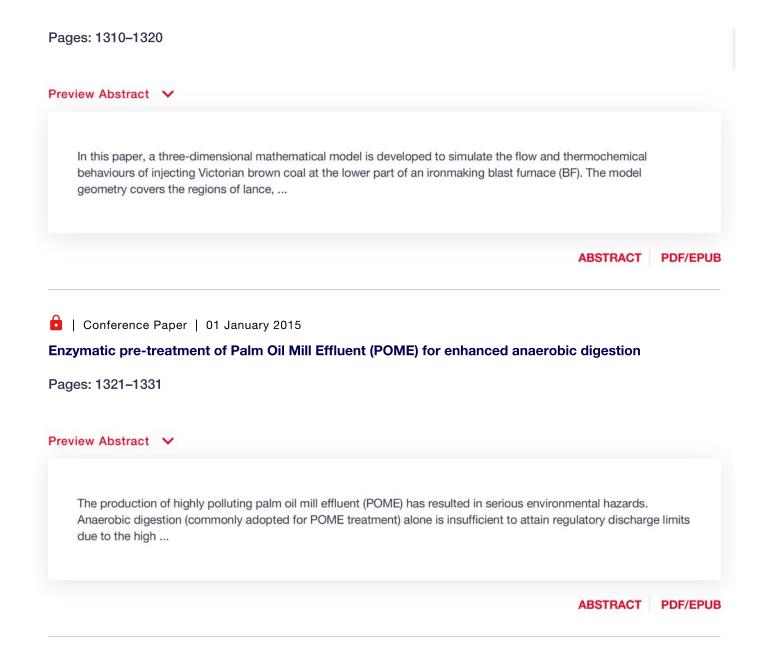


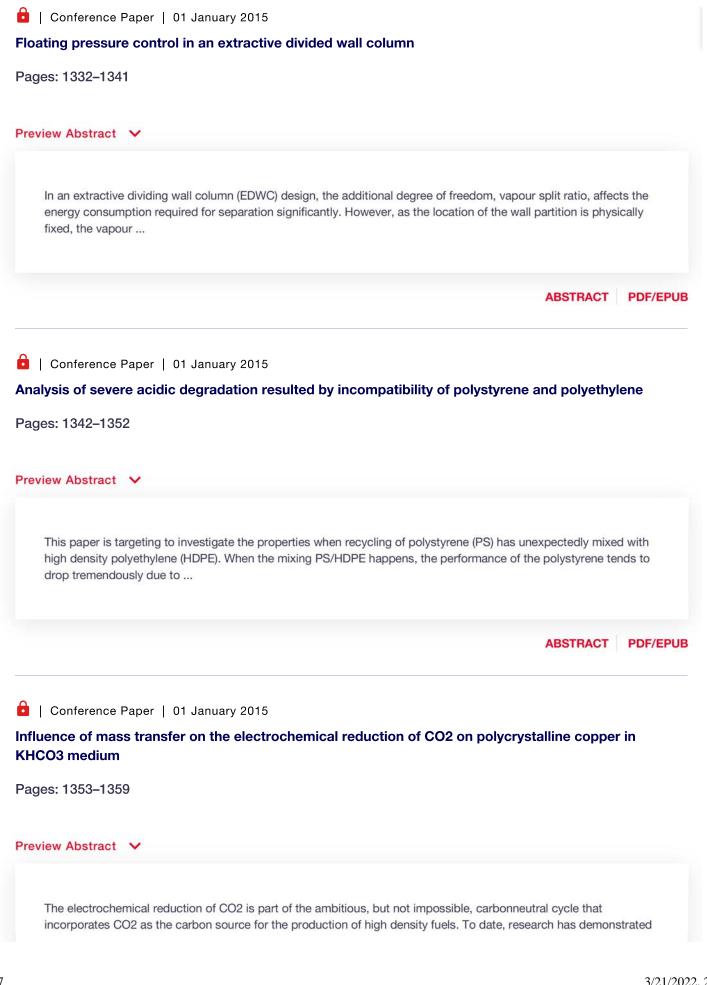


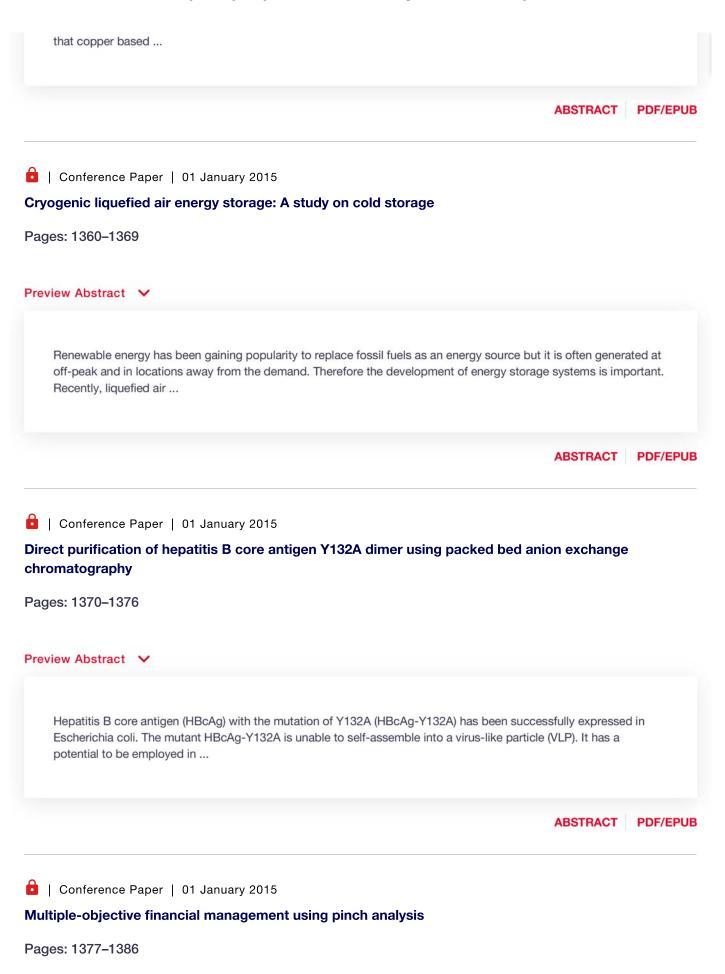


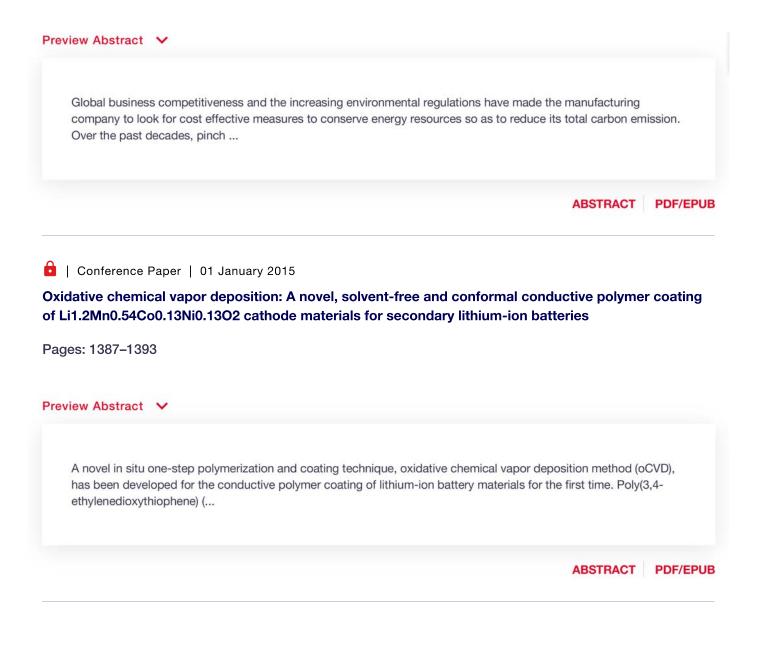


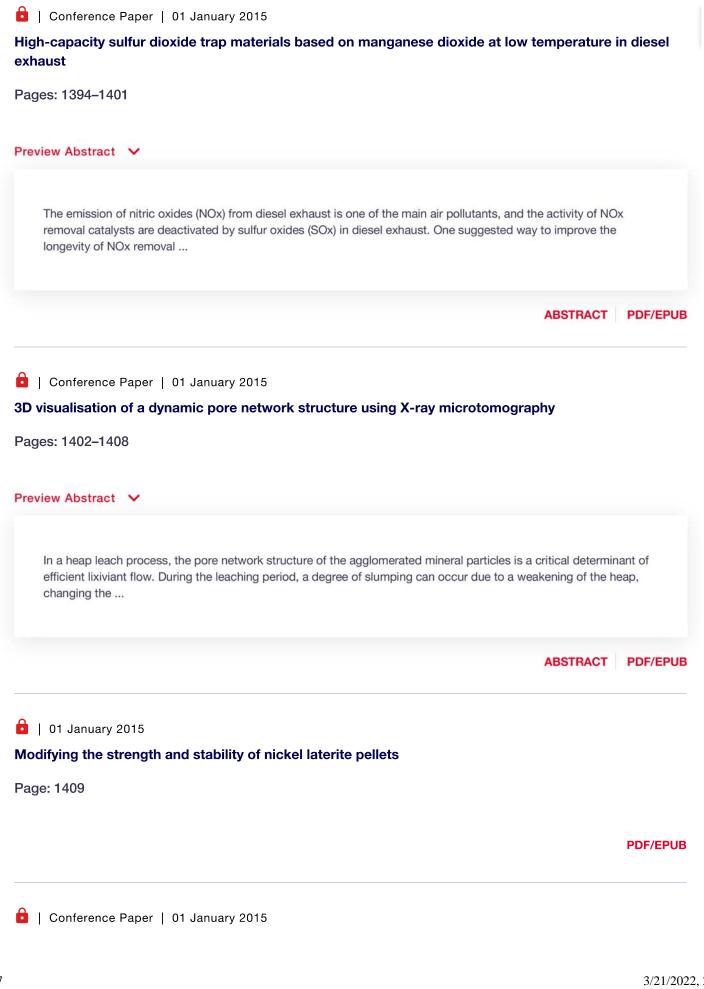


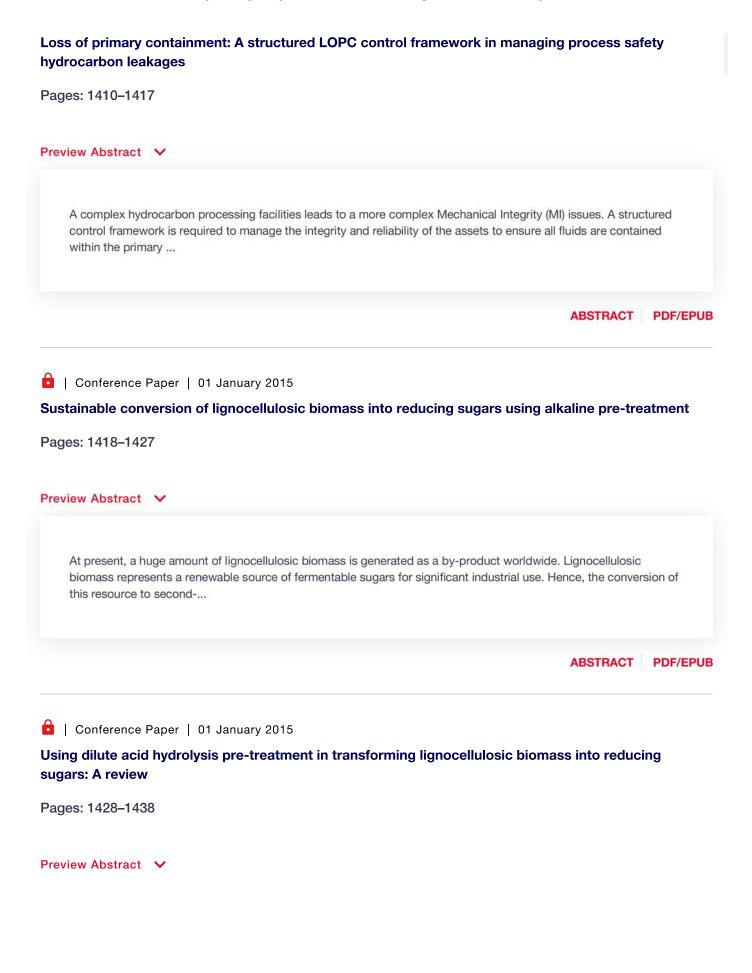


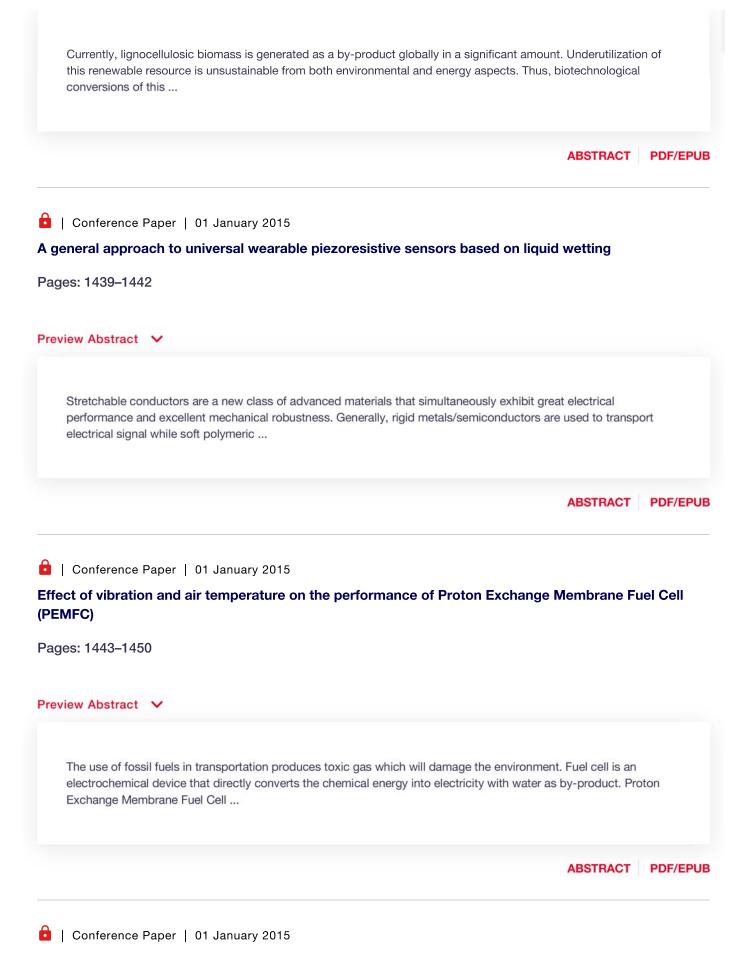


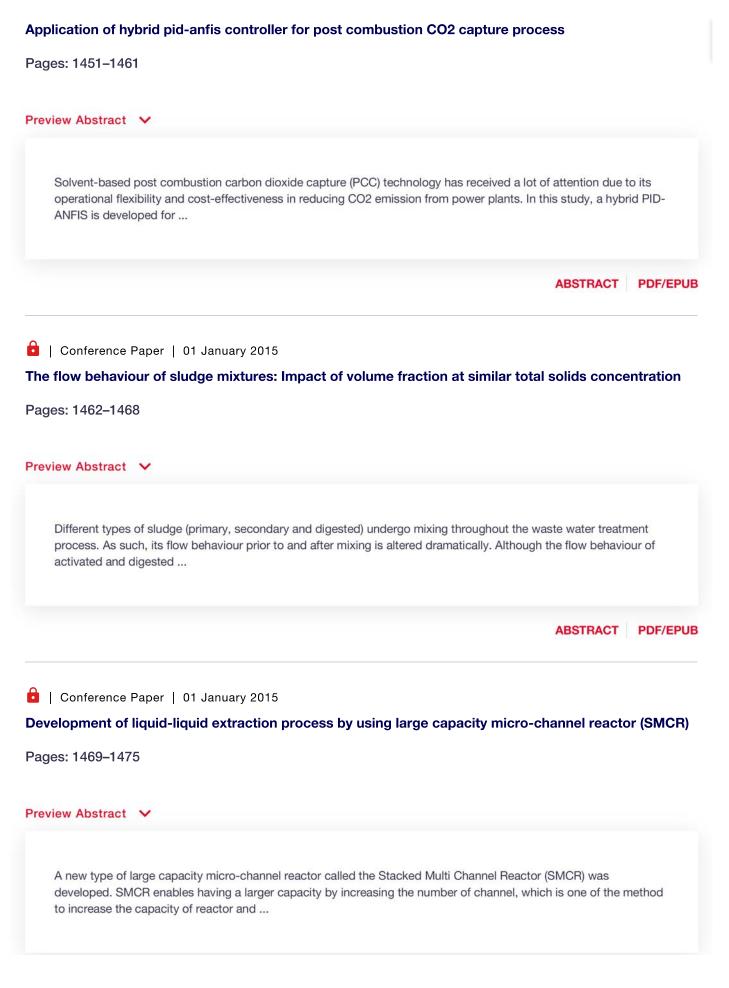


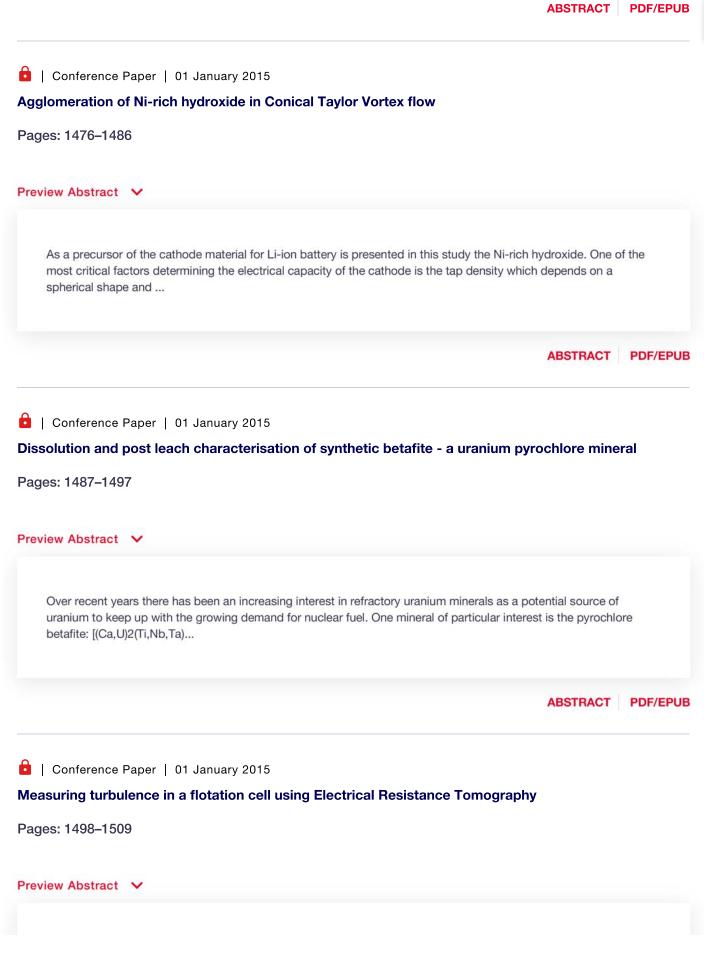


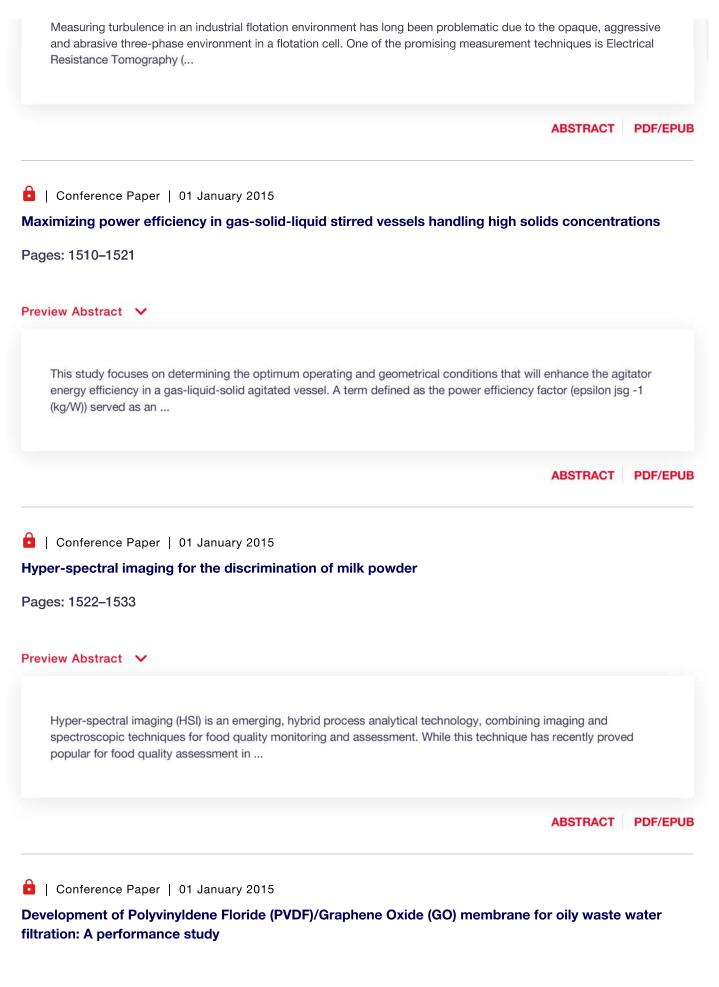




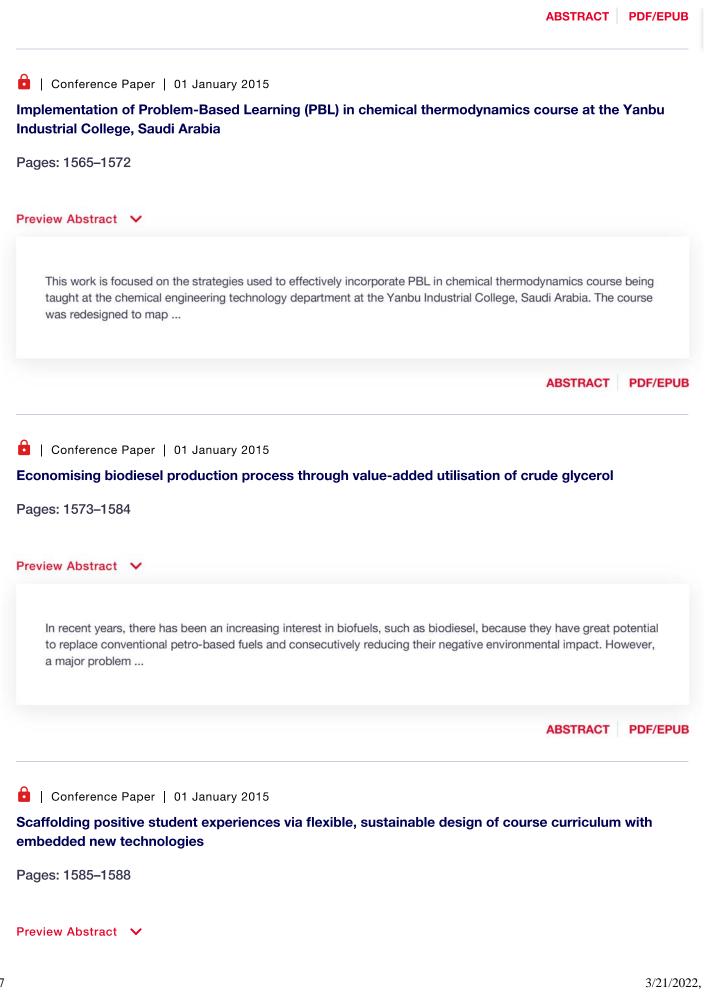


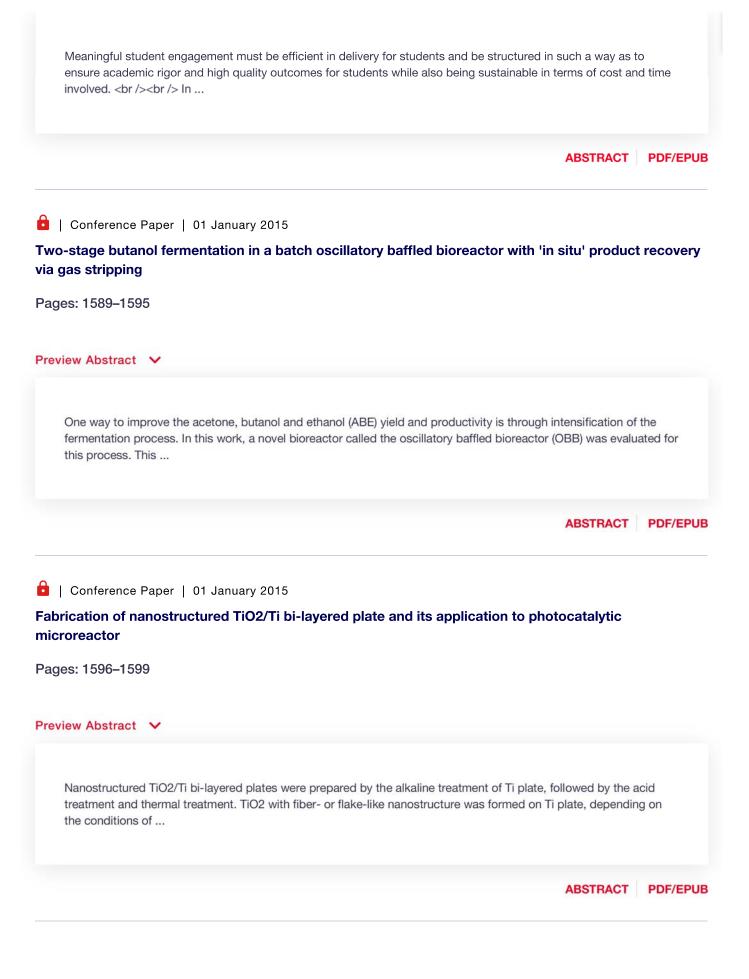


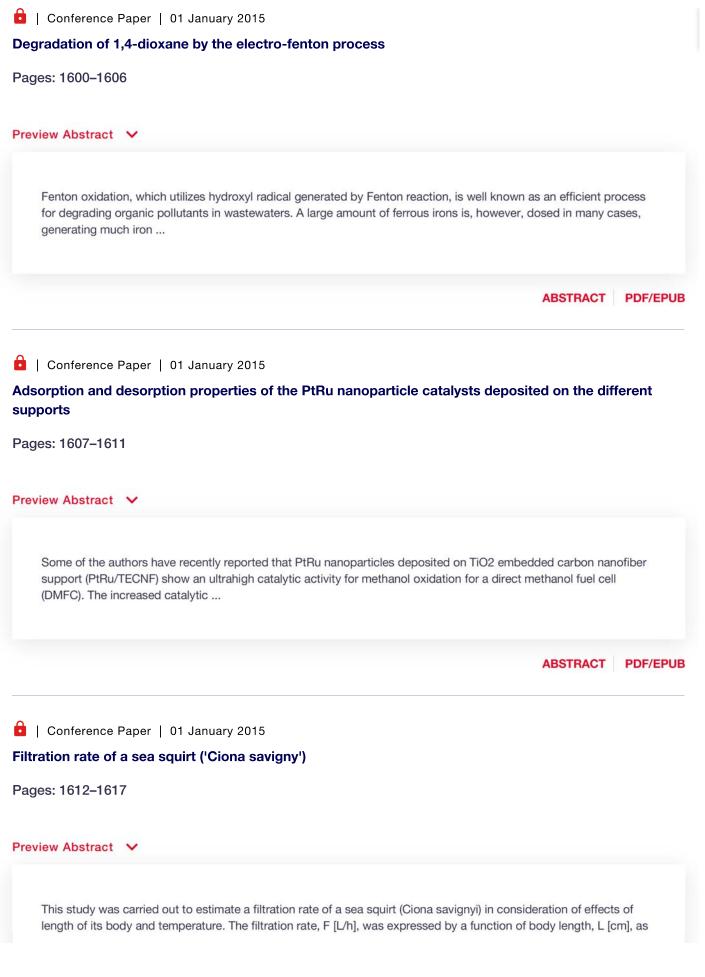




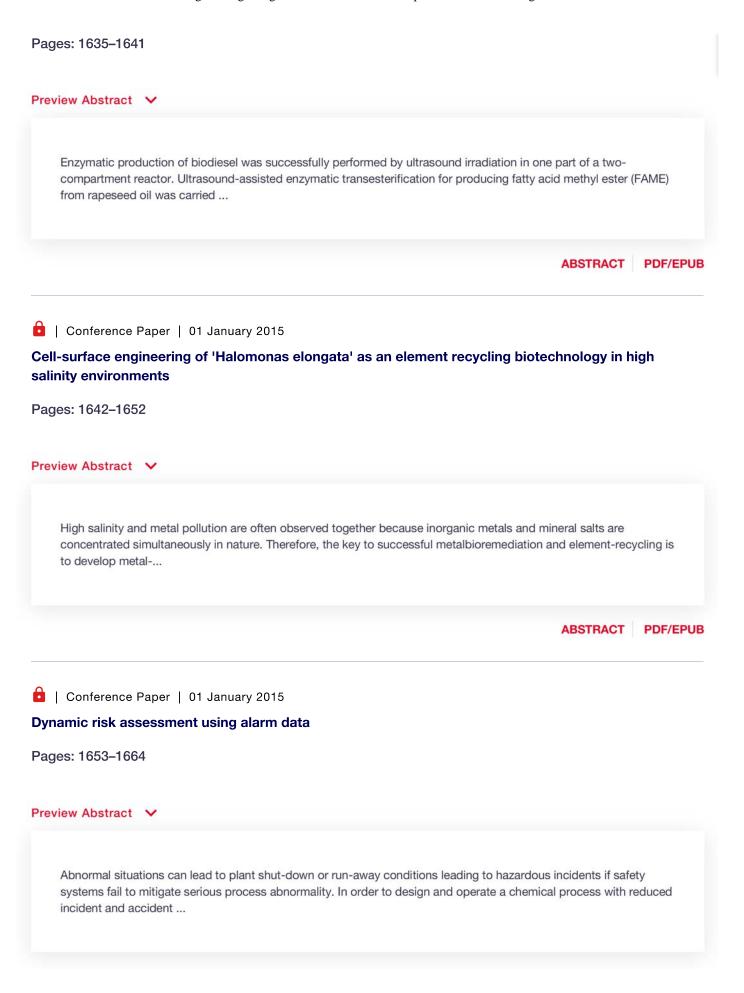
review Abstract 🗸	
Polyvinyldene floride (PVDF)/ graphene oxide (GO) membrane was developed for oily waste water filtration purposes at different pH. Incorporation of GO at 2wt% loading was found to decrease the overall crystallinity of the membrane as indicated by	
ABSTRACT PDF/E	PUE
Conference Paper 01 January 2015	
Production of lignin resin material from lignocellulosic biomass combining acidic saccharification an cetone treatment	ıd
Pages: 1546–1552	
review Abstract 🗸	
The structure of lignin, which is one of the main components of lignocellulosic biomass, is basically formed by the cross-linking of phenolic compounds. Taking the advantage of this structural property, the conversion method of lignin to resin material	
	PUE
ABSTRACT PDF/E	
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Conference Paper 01 January 2015	
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Conference Paper 01 January 2015 Synthesis, optimisation and characterisation of thermoresponsive polymer brushes Pages: 1553–1564	





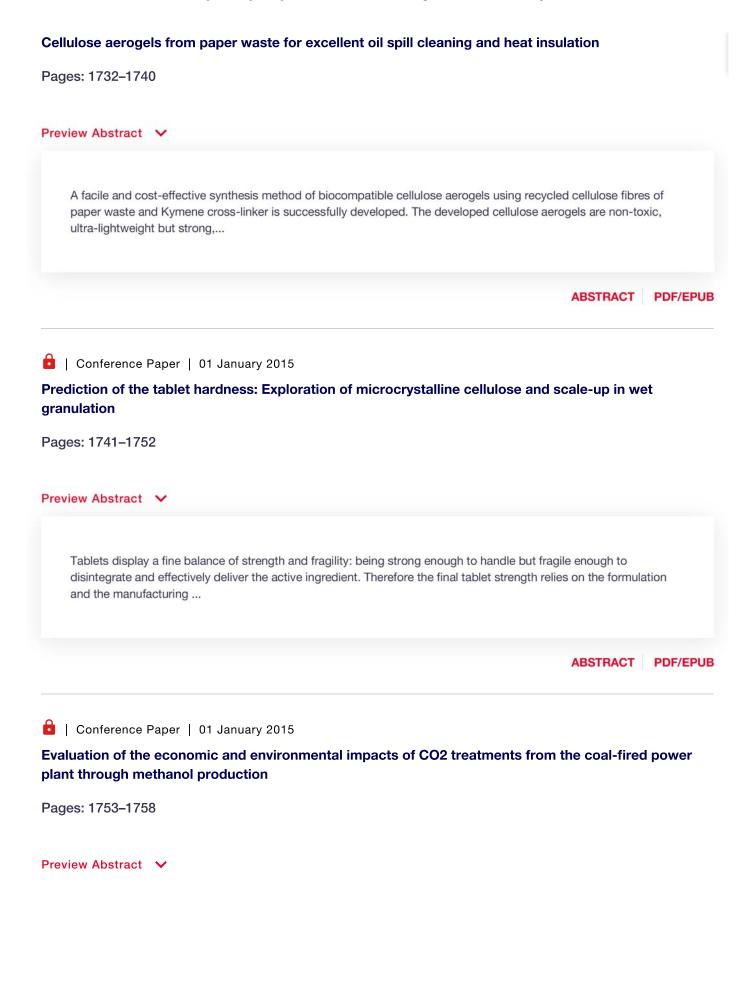


	ABSTRACT	PDF/EPU
Conference Paper 01 January 2015		
study on non-thermal effect of microwave through molecular orbital me f bubble formation	ethod and in-situ obs	ervation
ages: 1618–1622		
review Abstract 🗸		
Non-thermal effect of microwave on water has been a subject of interest, however, its understood. This study, therefore, aims to report the experimental and simulation invermicrowave on water. Particularly,		f
	ABSTRACT	PDF/EPU
Conference Paper 01 January 2015	ABSTRACT	PDF/EPU
lass transfer characterization of aluminum carboxymethyl cellulose men		
Conference Paper 01 January 2015 Iass transfer characterization of aluminum carboxymethyl cellulose mennking reaction		
lass transfer characterization of aluminum carboxymethyl cellulose men nking reaction		
lass transfer characterization of aluminum carboxymethyl cellulose men nking reaction ages: 1623–1634	nbrane involved in cr	VIC)
Iass transfer characterization of aluminum carboxymethyl cellulose mennking reaction ages: 1623–1634 review Abstract Superior molecular size screening and mass transfer flux of aluminum cross-linked ca membrane were demonstrated. The effect of cross-linking time by aluminum cation or some strain of the effect of cross-linking time by aluminum cation or some strain o	nbrane involved in cr	°OSS- MC)



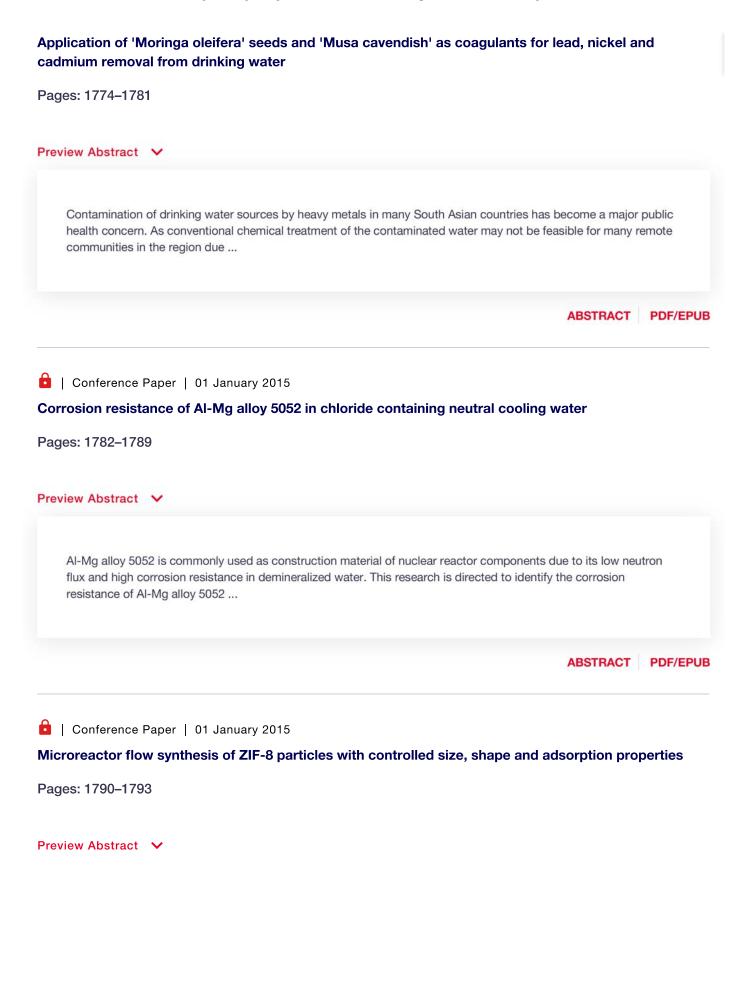
Conference	e Paper 01 January 2015	
A continuum si	mulation model for the Reflux classifier	
Pages: 1665–16	375	
Preview Abstract	t 🗸	
multicompon	for continuous processing has been developed to study the segregation and dispersion of ent systems in the Reflux Classifier (RC). The fluidized and inclined sections in the RC were divide Is and elements in the	ed into
	ABSTRACT	PDF/EPUE
	e Paper 01 January 2015 production of fatty acids by immobilized lipase in supercritical carbon dioxide	toward
Biocompatible industrial proce	production of fatty acids by immobilized lipase in supercritical carbon dioxide ess design	toward
Biocompatible industrial proce Pages: 1688–16	production of fatty acids by immobilized lipase in supercritical carbon dioxide ess design	toward
Biocompatible industrial proce Pages: 1688–16 Preview Abstrac Biocompatibl designed for	production of fatty acids by immobilized lipase in supercritical carbon dioxide ess design	toward
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Biocompatible industrial proce Pages: 1688–16 Preview Abstrac Biocompatibl designed for SCCO2 are it	production of fatty acids by immobilized lipase in supercritical carbon dioxide ess design 398 t v	
Biocompatible industrial proce Pages: 1688–16 Preview Abstract Biocompatible designed for SCCO2 are it	production of fatty acids by immobilized lipase in supercritical carbon dioxide ess design iss t le production of fatty acids using immobilized lipase in supercritical carbon dioxide (SCCO2) was industrial processes. The advantages of biocompatible production with enzymatic reaction using its nontoxicity for human ABSTRACT	
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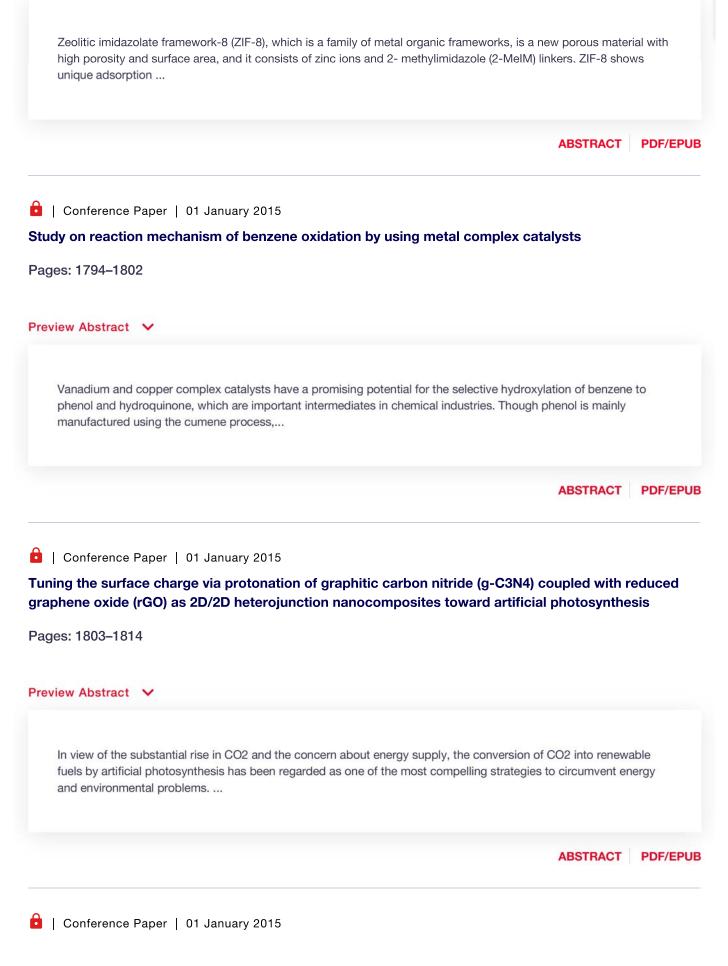
materials	
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owards processing waste green plant materials: A naterials and biochemical products	sustainable source of leaf proteins, raw biological
ages: 1711–1719	
review Abstract 🗸	
Waste green plant materials (WGPMs), such as lawn clippi materials with potential value-added applications. This stur protein recovery process to	
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Conference Paper 01 January 2015 systematic multi-objective methodology for optime ages: 1720–1731	
ages: 1720–1731	nal mixture design in integrated biorefineries

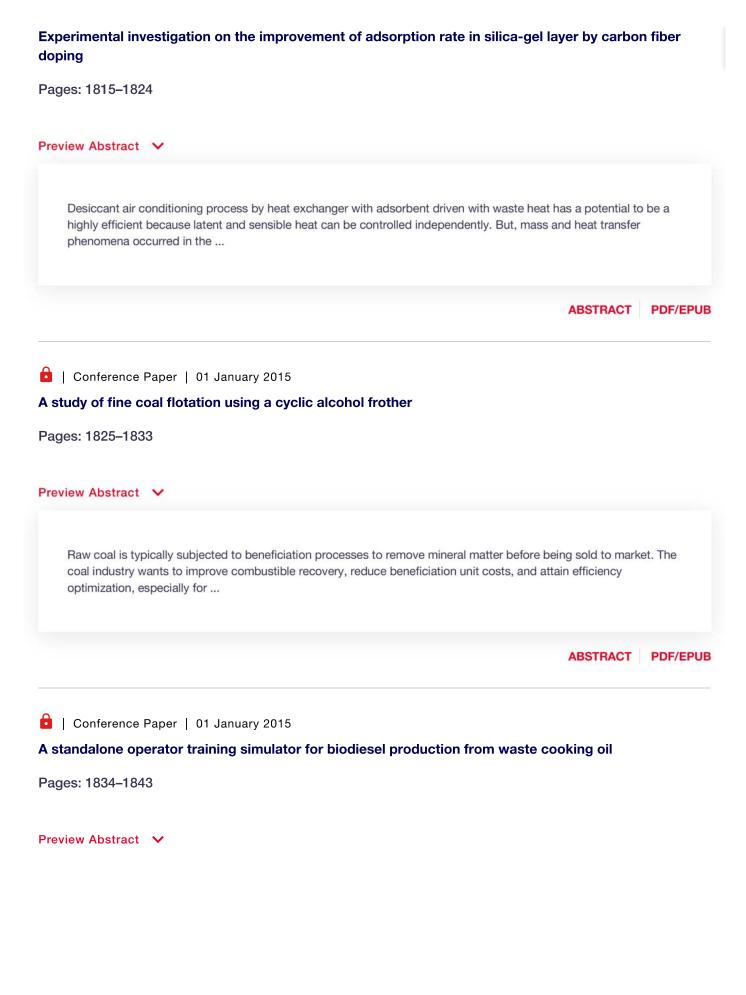


	e CO2. One viable solution is to recover and store CO2 underground through capture processes. However to capture CO2 is still	r, the
	ABSTRACT	PDF/EPU
🔒 Con	ference Paper 01 January 2015	
Starting	torque and flow dynamics of vertical paddle impellers	
ages: 1	759–1766	
review A	bstract 🗸	
	n rotation of an impeller is started, the torque is larger than that at a steady state. This torque is important n of the impeller. However, the relationship between the starting torque and the rotational speed and the e	
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n situ m emoval Pages: 1	ference Paper 01 January 2015 odifications of bacterial cellulose film with 'Pandanus amaryllifolius' extract for heavy	
n situ m emoval Pages: 1 Preview A Bacte prope	ference Paper 01 January 2015 odifications of bacterial cellulose film with 'Pandanus amaryllifolius' extract for heavy 767–1773	

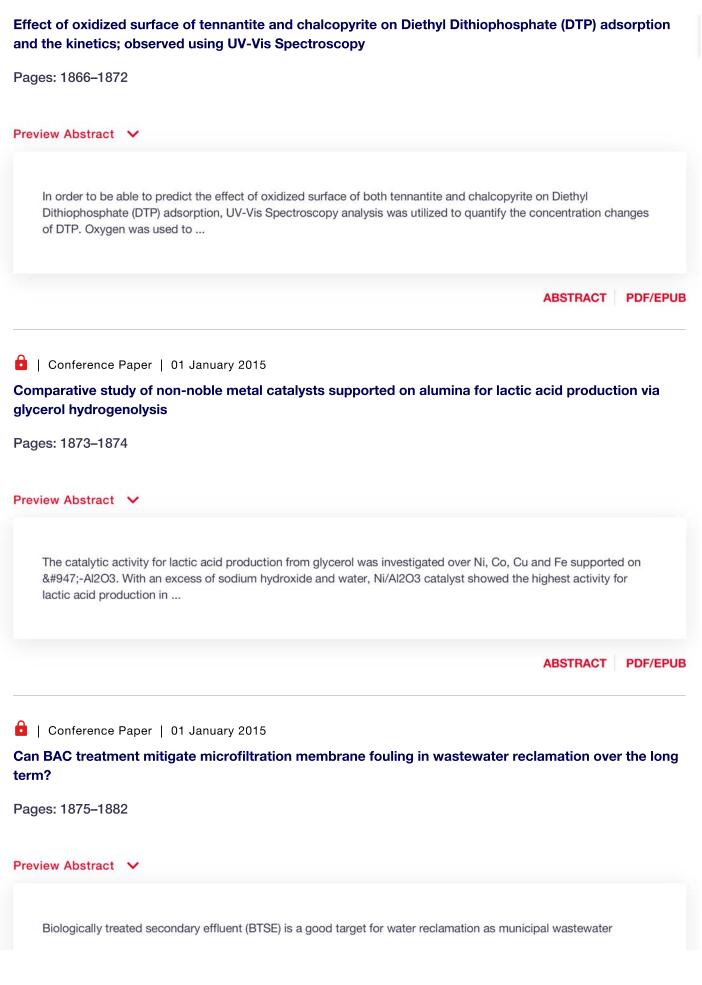
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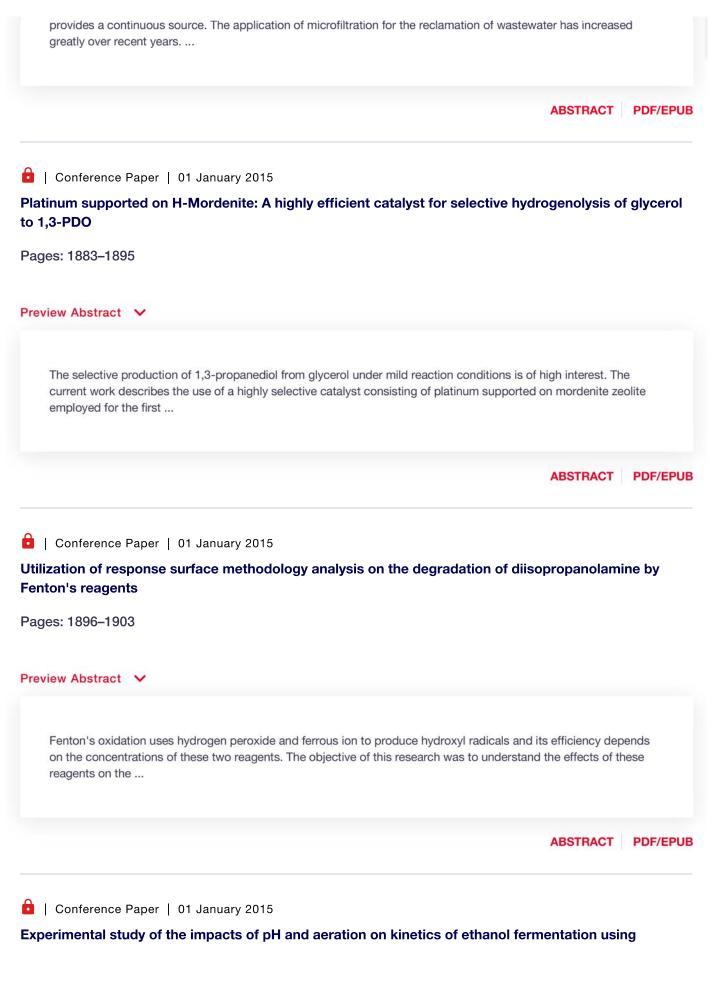




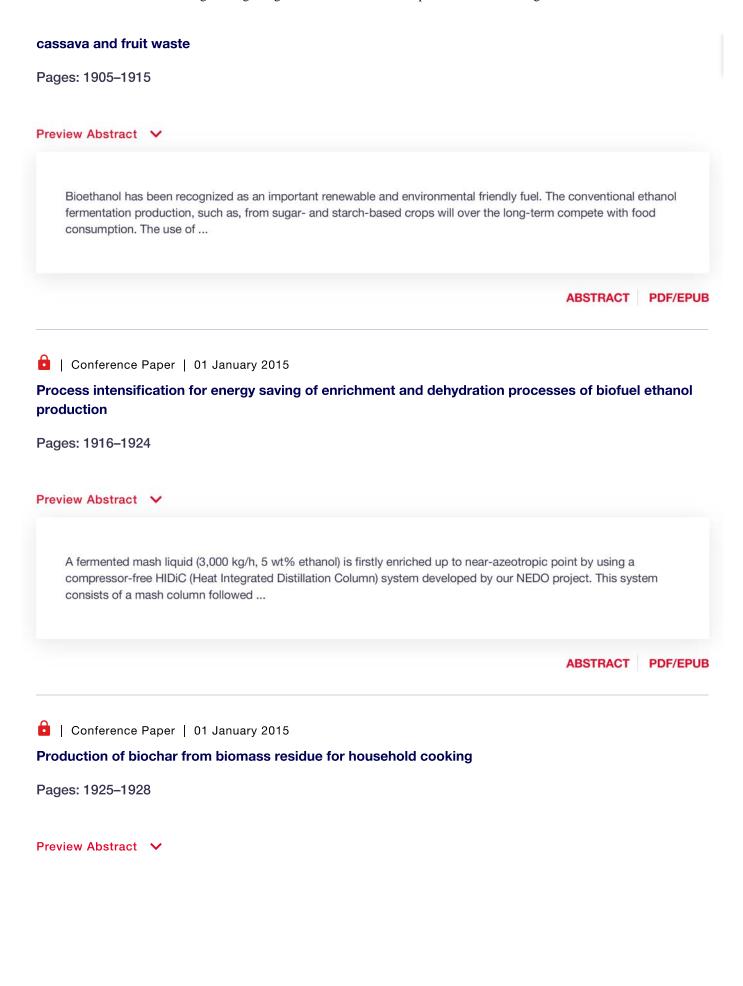


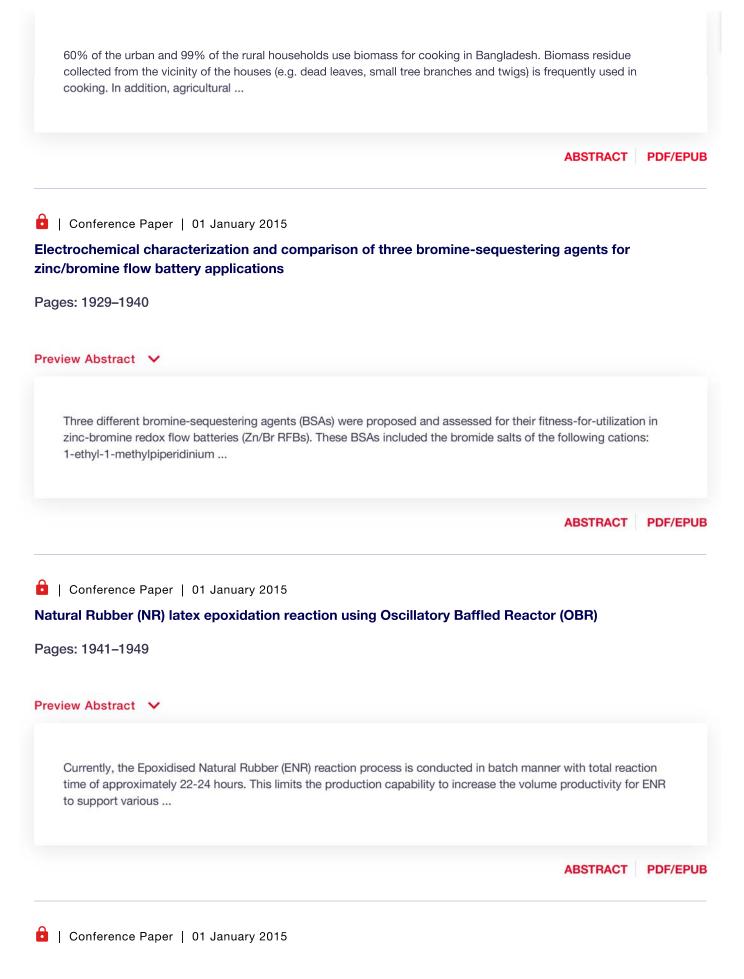
an alternative fuel in the future. Using waste cooking oil (WCO) enables cost effective facilitates	l concerns, biodiesel is seen as biodiesel production and also
	ABSTRACT PDF/EPUB
B Conference Paper 01 January 2015	
CFD-DEM simulation of binary systems of particles in liquid fluidised bed	s: Segregation and dispersion
Pages: 1844–1853	
Preview Abstract 🗸	
Solid liquid fluidised beds (SLFB) are widely used in industrial processes, where solid densities are often encountered. The differences in the physical properties of particles complete	
	ABSTRACT PDF/EPUB
Conference Paper 01 January 2015	
Conference Paper 01 January 2015 Analysis of interaction forces for predicting the transition from segregation a miniaturised gas fluidised bed	on to mixing of binary solids in
Analysis of interaction forces for predicting the transition from segregation a miniaturised gas fluidised bed	on to mixing of binary solids in
Analysis of interaction forces for predicting the transition from segregation a miniaturised gas fluidised bed Pages: 1854–1865	on to mixing of binary solids in
Analysis of interaction forces for predicting the transition from segregation	biochemical and powder
Analysis of interaction forces for predicting the transition from segregation a miniaturised gas fluidised bed Pages: 1854–1865 Preview Abstract V Gas-solid fluidised beds are widely used in chemical, petrochemical, pharmaceutical, industries. Particles used in gas-solid fluidised beds often differ in size and/or density	biochemical and powder

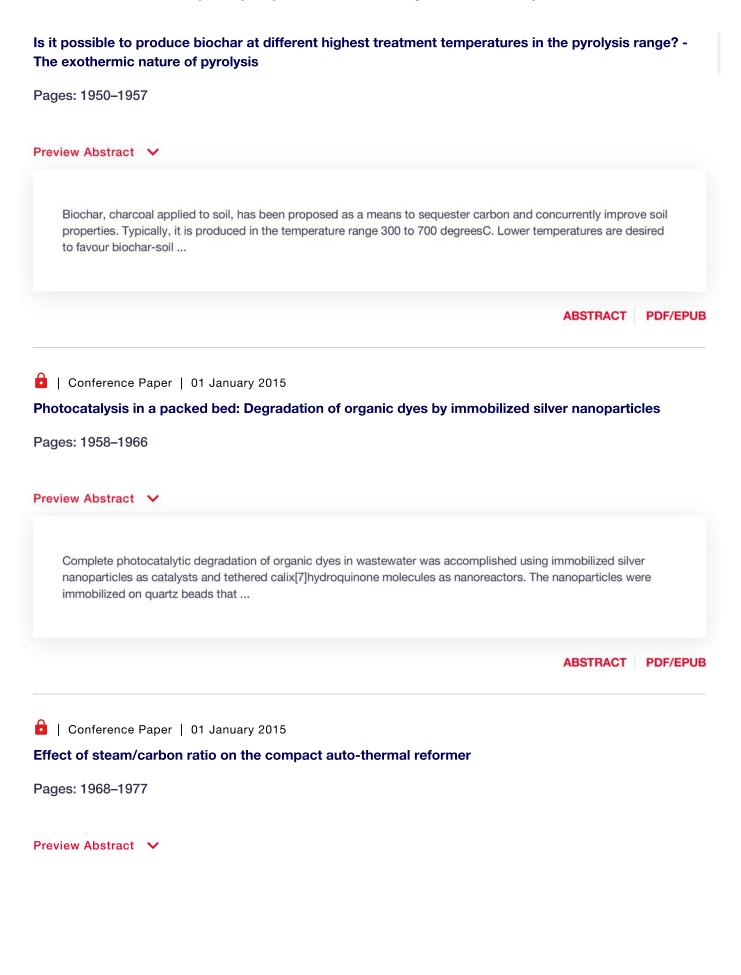




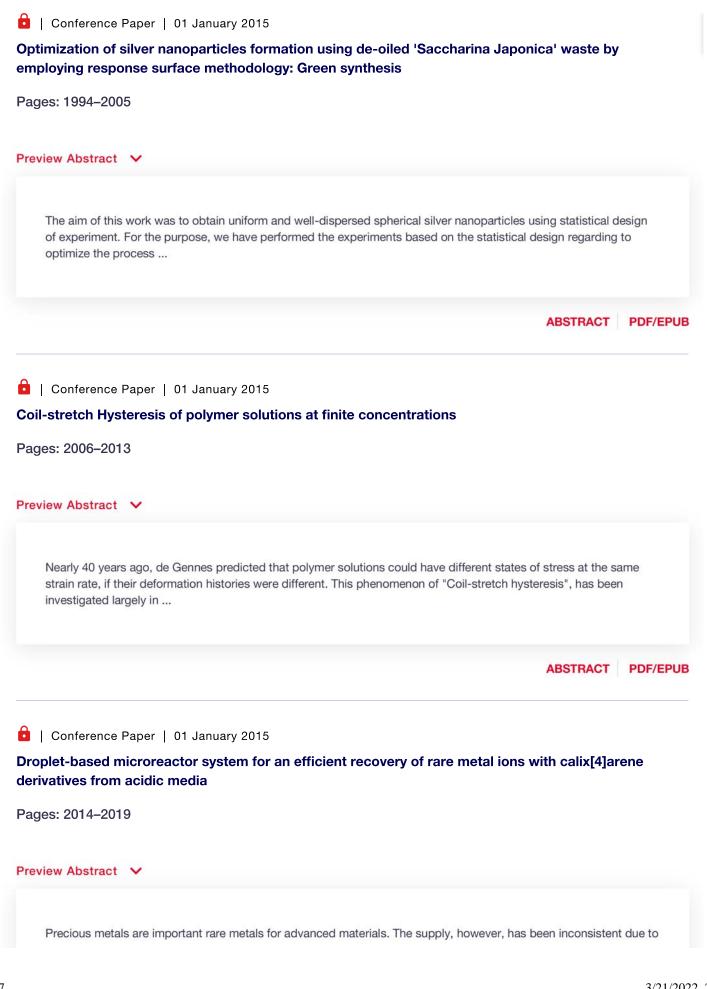
Asia Pacific Confederation of Chemical Engineering Congress 2015: A...

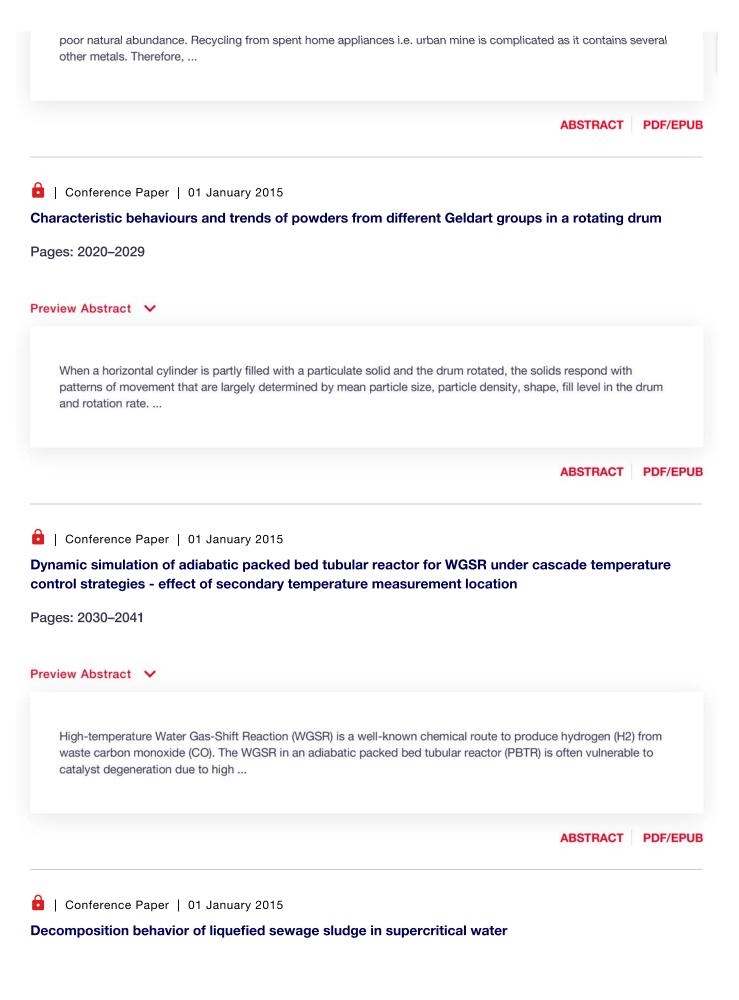




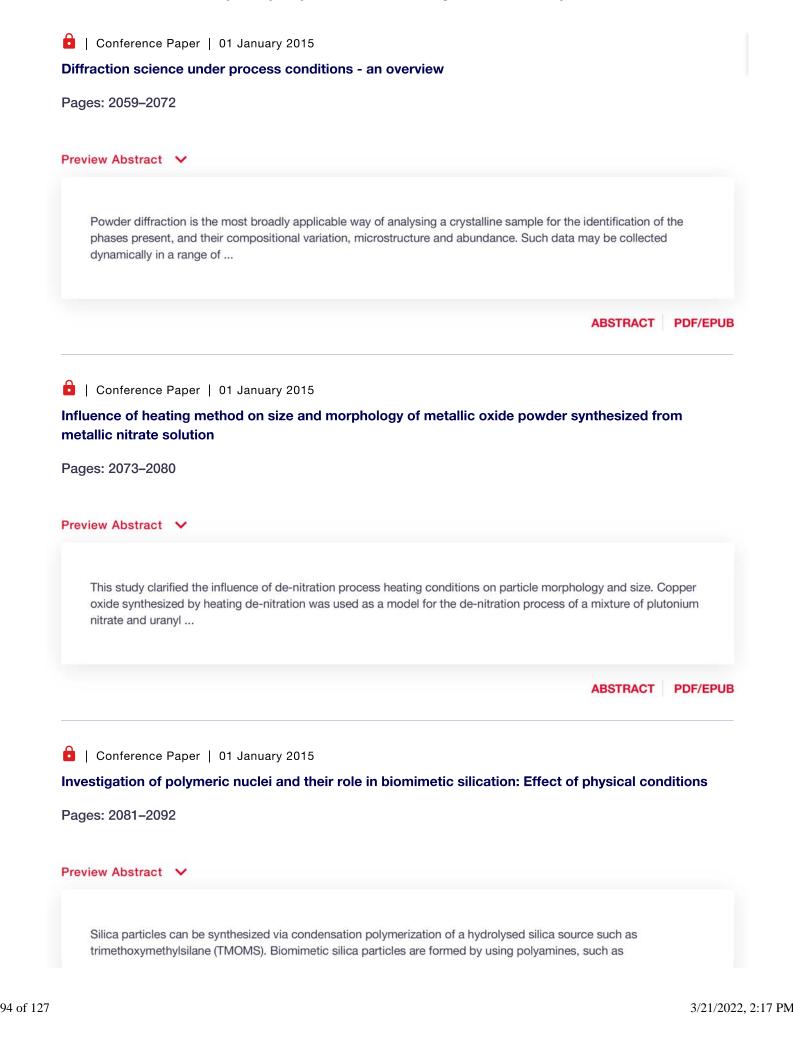


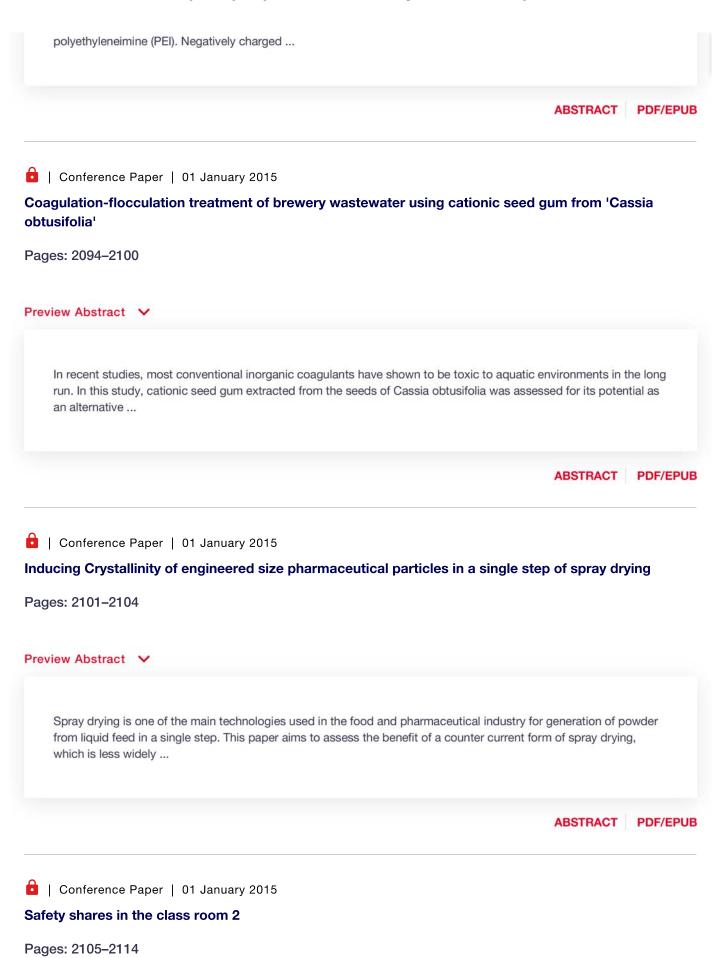
	ABSTRACT	PDF/EP
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athematical modelling novel burden profile	of flow and heat transfer in COREX process by CFDDEM method: The	e effect (
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	is produced mainly in a conventional blast furnace (BF). New ironmaking processes ha ast two decades because of environmental concerns to reduce CO2 emission. One suc ch can operate,	
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Conference Paper		
pact of pretreatment	on supercritical carbon dioxide extraction from dried parsley ('Petrose	elinum
pact of pretreatment spum L.') leaves	on supercritical carbon dioxide extraction from dried parsley ('Petros	elinum
	on supercritical carbon dioxide extraction from dried parsley ('Petros	elinum
pact of pretreatment spum L.') leaves ges: 1989–1993 eview Abstract v The effect of pretreatme	ent on the extraction of bioactive components apigenin from dried parsley ('Petroselinur g supercritical carbon dioxide (SCCO2) was examined. Apigenin is a bioactive ingredier	m

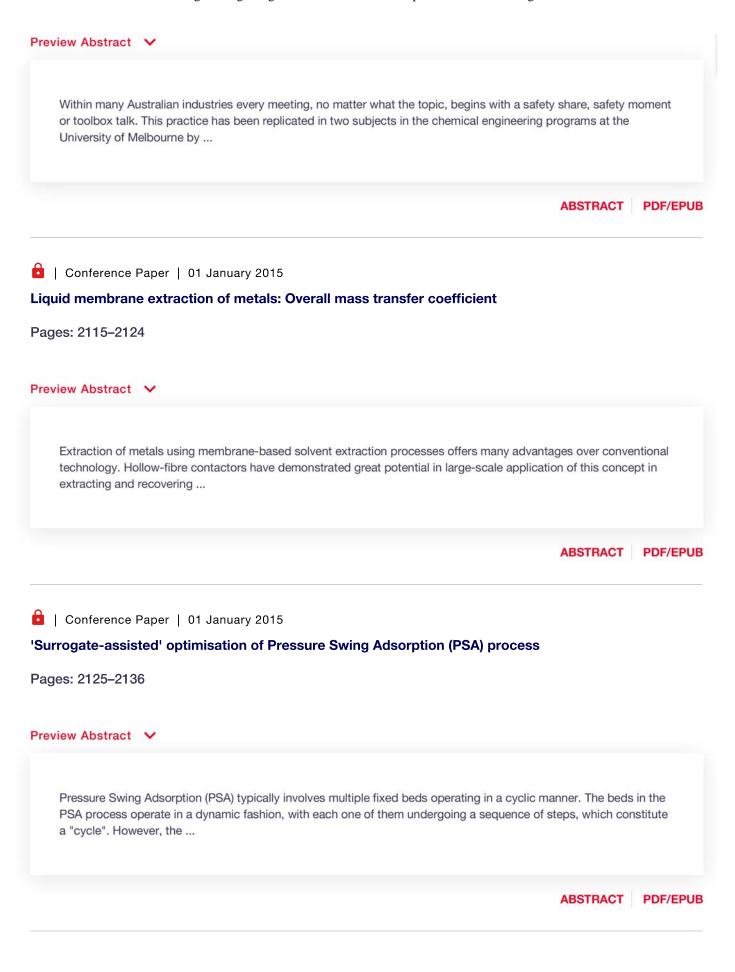


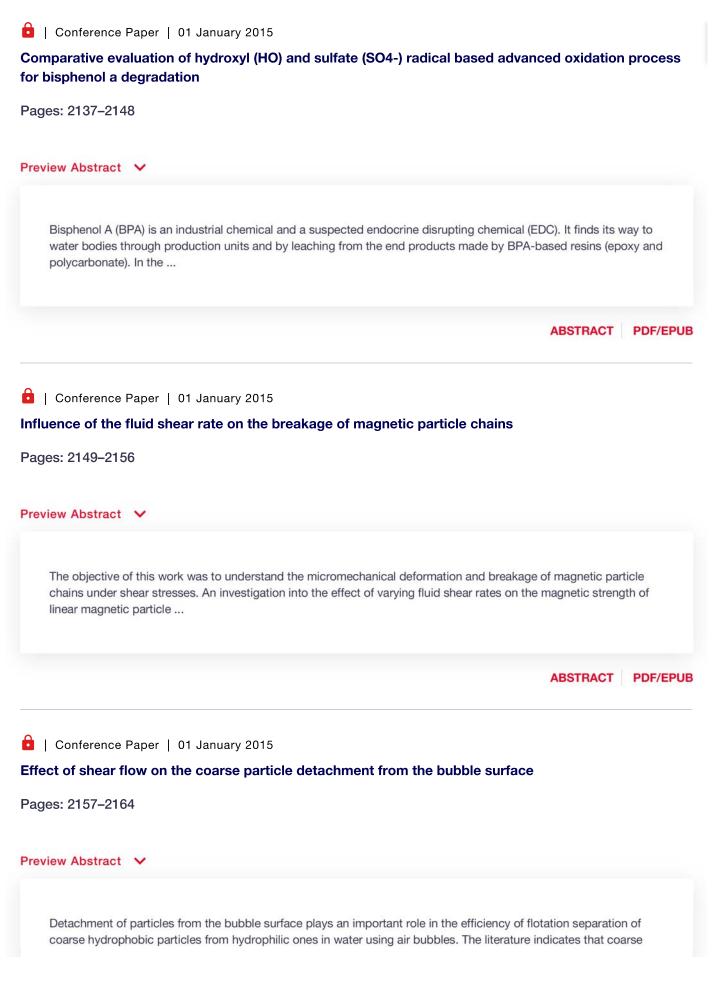


Prev	iew Abstract 🗸
	In this work, a fundamental study on the decomposition behavior of liquefied sewage sludge in supercritical water was considered. From gasification experiments using batch reactors, both gas production and efficiencies were in no way inferior to those
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6	Conference Paper 01 January 2015
	Conference Paper 01 January 2015 simulation of thorium extraction in membrane contactors
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CFE Pag	simulation of thorium extraction in membrane contactors

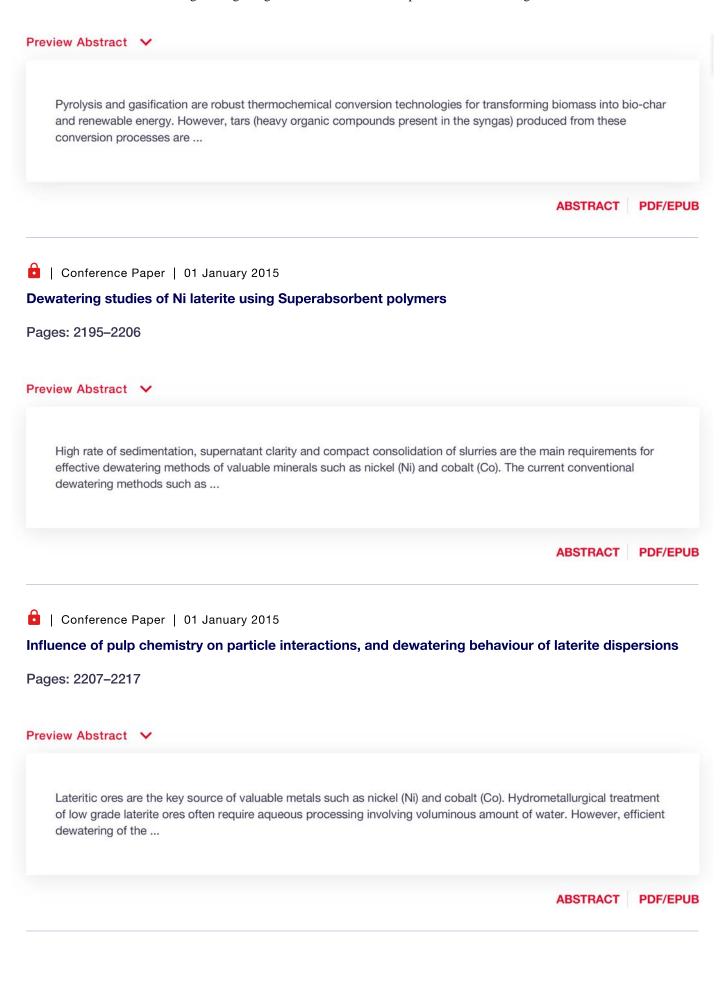


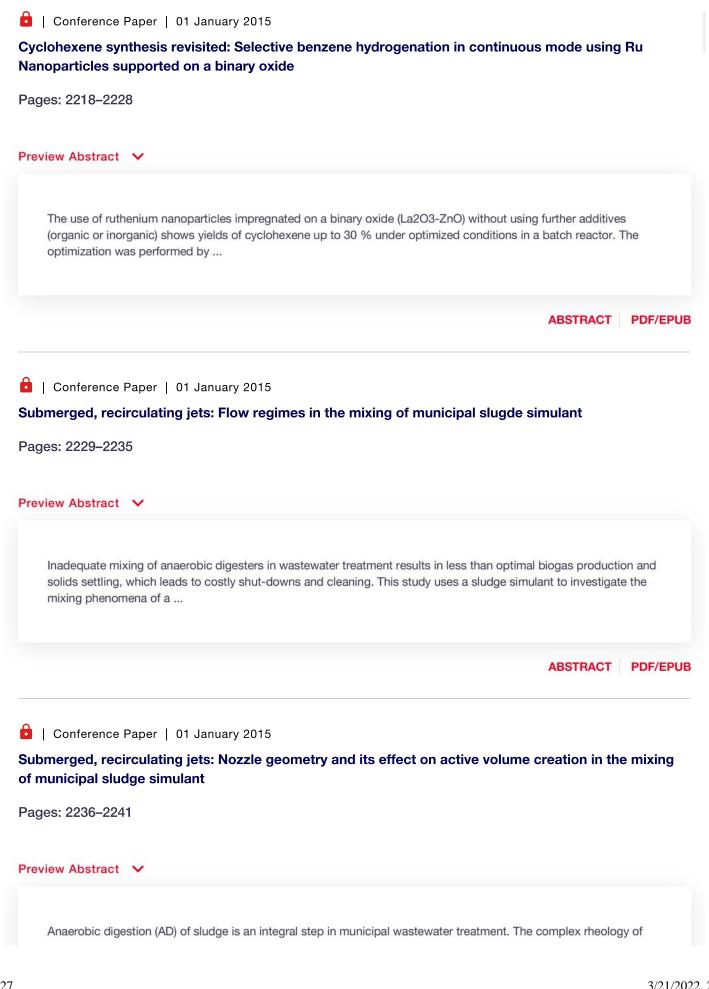


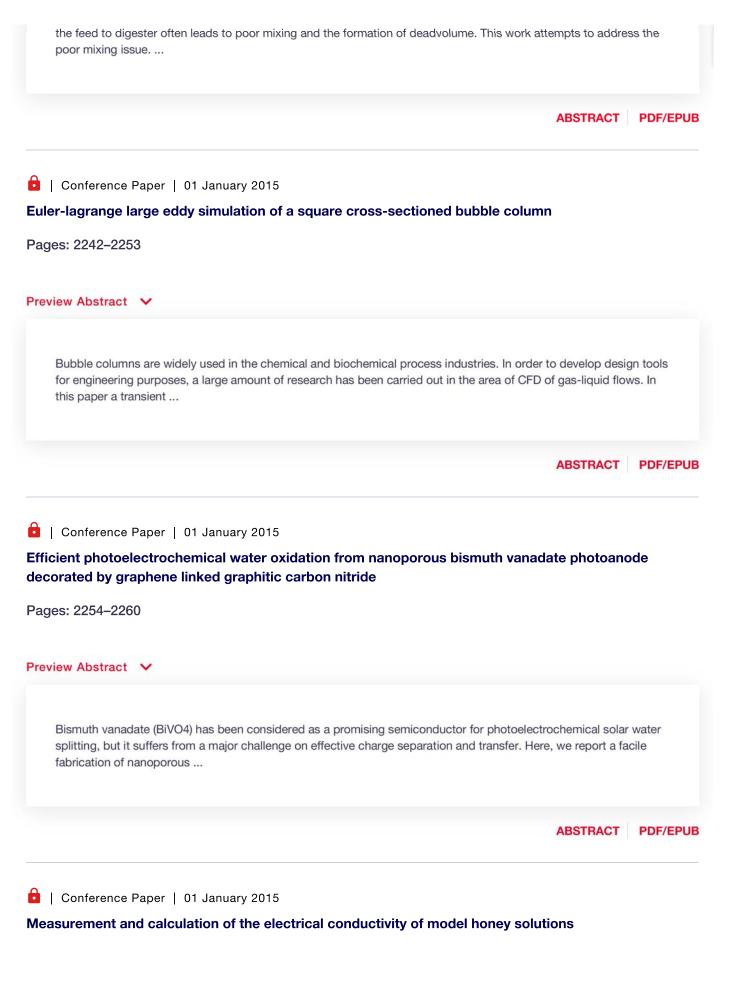


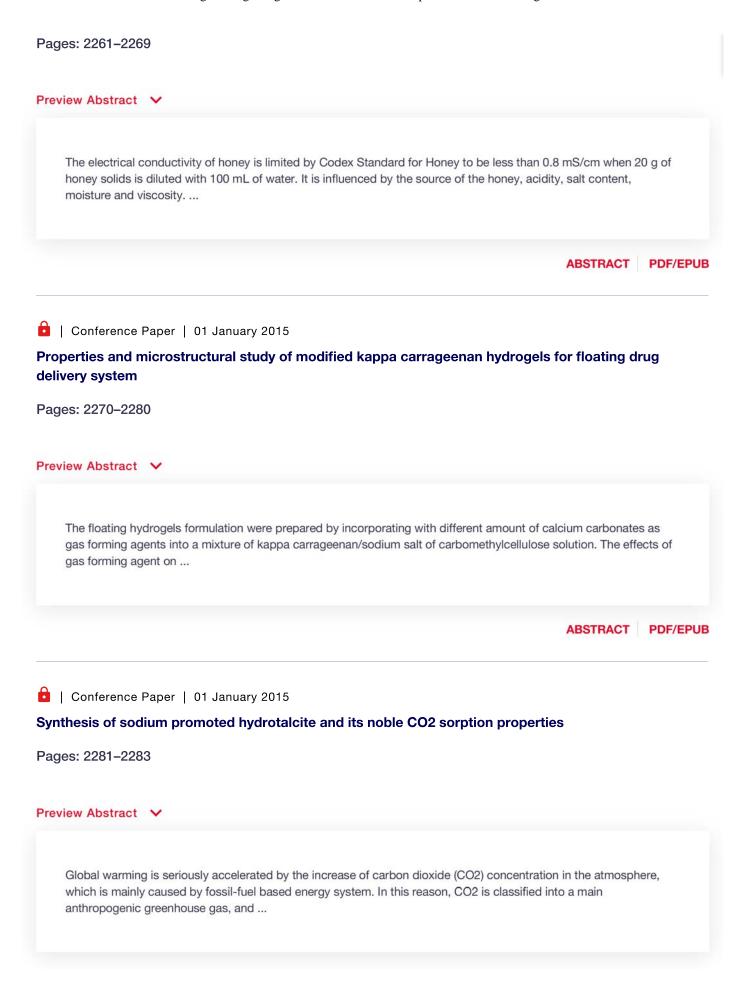


Terence Paper 01 January 2015 Ing yields from the extraction of different citrus peels and spray drying of the 65–2173 bstract ♥ work, a comparison between the quality and quantity of natural antioxidant powders production and spray drying of various citrus peels has been performed. The average total phenolisus (orange,	uced from cc	ombined TPC) of
Terence Paper 01 January 2015 Ing yields from the extraction of different citrus peels and spray drying of the 65–2173 bstract ♥ work, a comparison between the quality and quantity of natural antioxidant powders production and spray drying of various citrus peels has been performed. The average total phenolisus (orange,	he extract	S ombined TPC) of
ag yields from the extraction of different citrus peels and spray drying of the 65–2173 bestract work, a comparison between the quality and quantity of natural antioxidant powders prodution and spray drying of various citrus peels has been performed. The average total phenoli us (orange,	uced from co lic contents (ombined TPC) of
ag yields from the extraction of different citrus peels and spray drying of the 65–2173 bestract work, a comparison between the quality and quantity of natural antioxidant powders prodution and spray drying of various citrus peels has been performed. The average total phenoli us (orange,	uced from co lic contents (ombined TPC) of
65–2173 bstract work, a comparison between the quality and quantity of natural antioxidant powders prodution and spray drying of various citrus peels has been performed. The average total phenoli us (orange,	uced from co lic contents (ombined TPC) of
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work, a comparison between the quality and quantity of natural antioxidant powders produ tion and spray drying of various citrus peels has been performed. The average total phenoli us (orange,	lic contents (TPC) of
tion and spray drying of various citrus peels has been performed. The average total phenoli us (orange,	lic contents (TPC) of
tion and spray drying of various citrus peels has been performed. The average total phenoli us (orange,	lic contents (TPC) of
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liquid separation involving suspensions is important in a large range of industrial application ssing and wastewater treatment and disposal. The development of theoretical descriptions ation, or		
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ioxide reforming of biomass tar using recycled material as catalyst suppo 86–2194	7113	

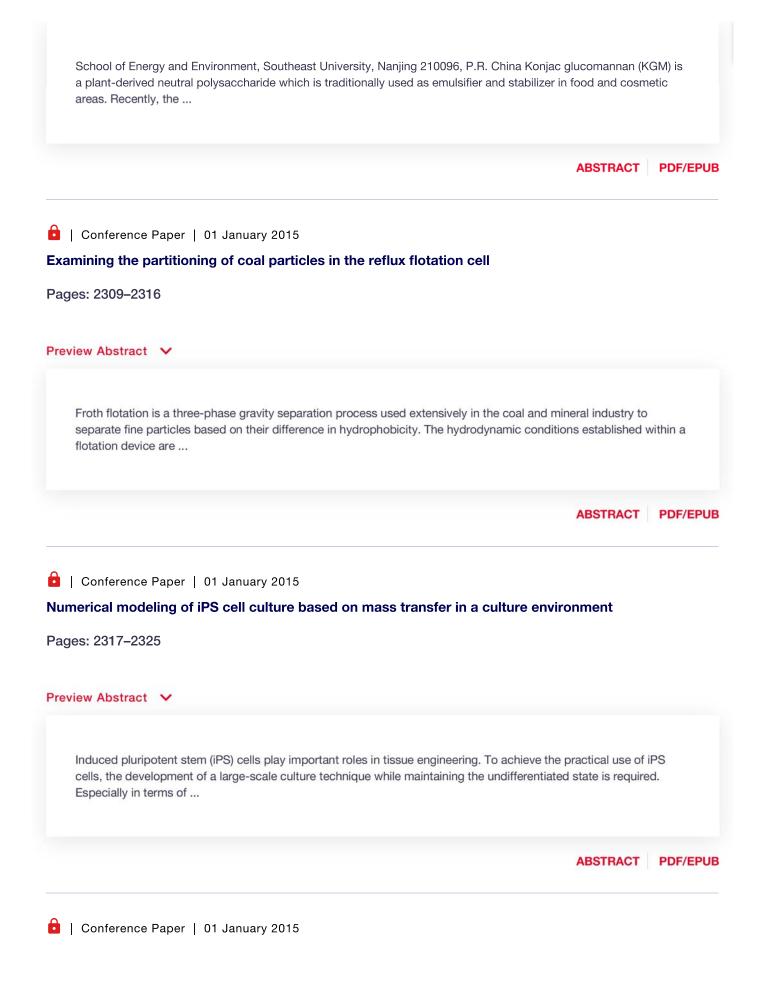


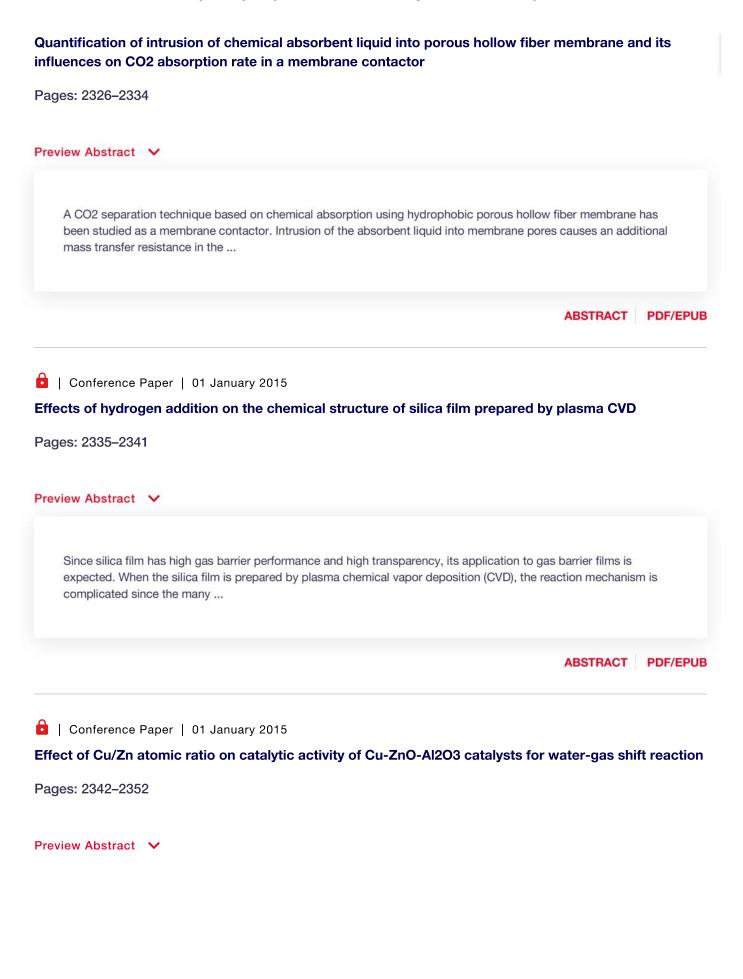


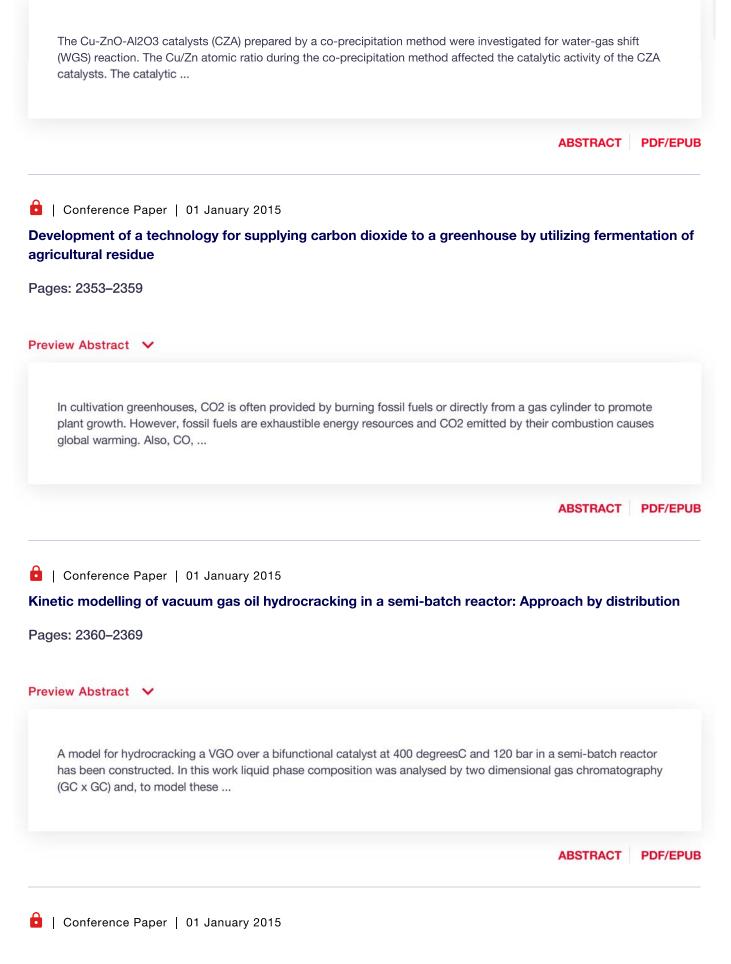


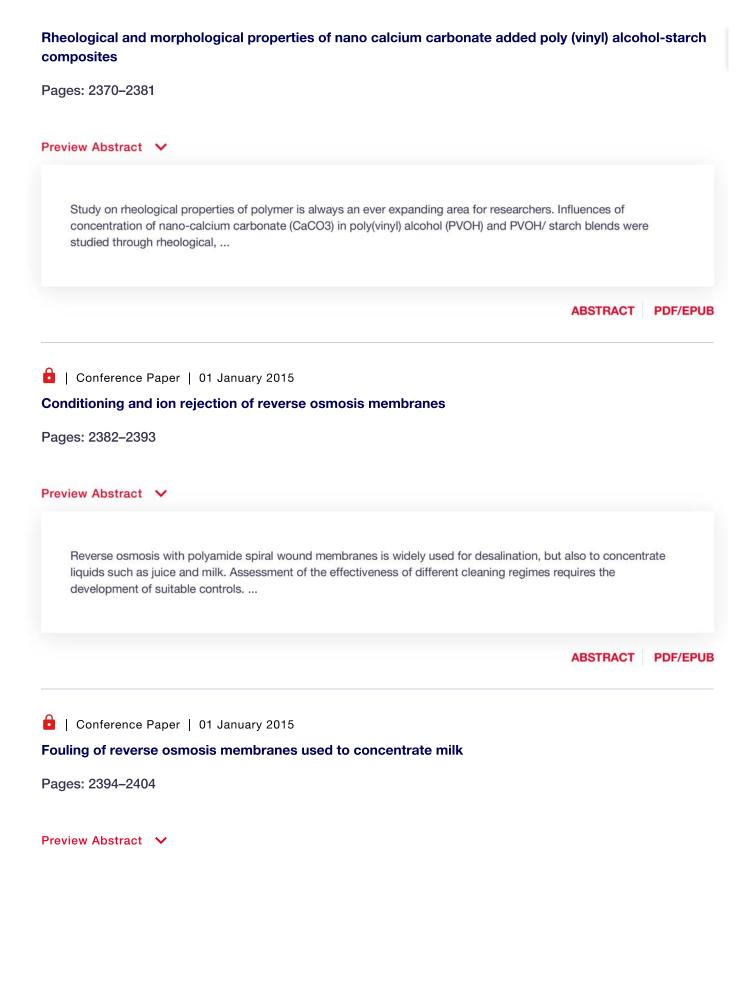


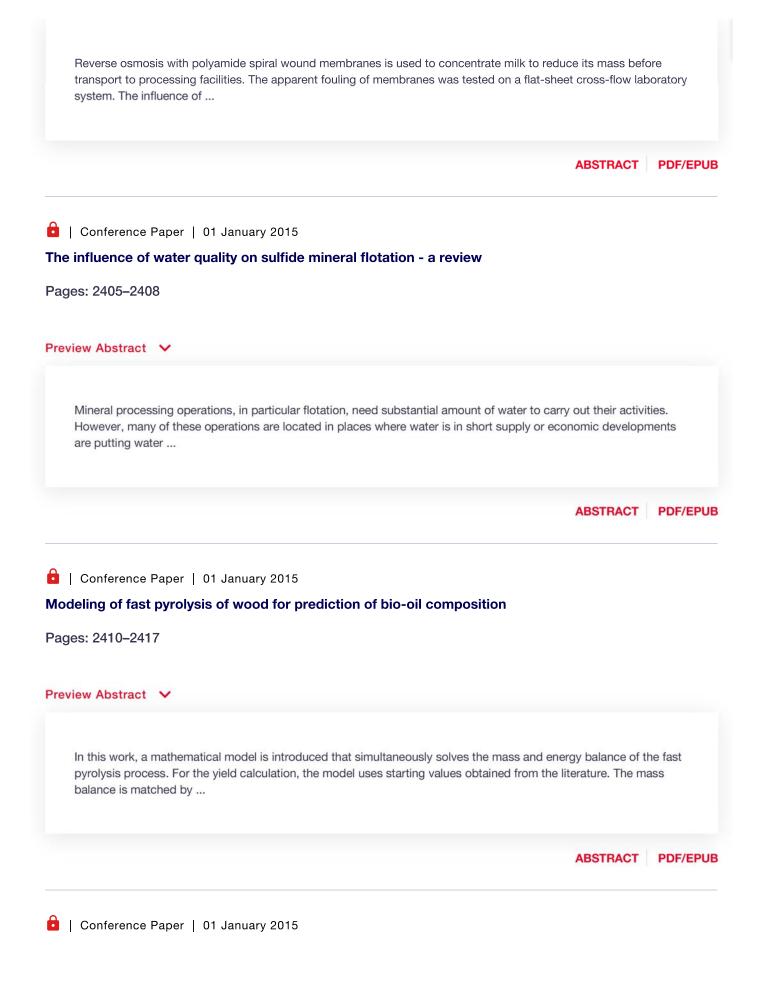
🔓 Conference Paper 01 January 2015	
Mixed matrix membranes embedding modified	hydroxyapatite for protein adsorption
Pages: 2284–2288	
Preview Abstract 🗸	
	ultrafiltration and microfiltration membranes because of its nertness. Hydroxyapatite (HAP) is a nano inorganic material and
	ABSTRACT PDF/EPUI
	on and NOx emission characteristics in a 660 MWe
tangential firing ultra-supercritical boiler Pages: 2289–2296	on and NOx emission characteristics in a 660 MWe
tangential firing ultra-supercritical boiler Pages: 2289–2296 Preview Abstract A three-dimensional numerical simulation was carrie	ed out to study the pulverized coal combustion process on a ble k-ε model for gas coupled with discrete phase model
tangential firing ultra-supercritical boiler Pages: 2289–2296 Preview Abstract ↓ A three-dimensional numerical simulation was carrie tangential firing ultra-supercritical boiler. The realizal	ed out to study the pulverized coal combustion process on a
tangential firing ultra-supercritical boiler Pages: 2289–2296 Preview Abstract ↓ A three-dimensional numerical simulation was carrie tangential firing ultra-supercritical boiler. The realizal	ed out to study the pulverized coal combustion process on a ble k-ε model for gas coupled with discrete phase model
tangential firing ultra-supercritical boiler Pages: 2289–2296 Preview Abstract A three-dimensional numerical simulation was carrie tangential firing ultra-supercritical boiler. The realizat (DPM) for coal particles Image: Conference Paper 01 January 2015 Konjac glucomannan-based microspheres for	ed out to study the pulverized coal combustion process on a ble k-ε model for gas coupled with discrete phase model
tangential firing ultra-supercritical boiler Pages: 2289–2296 Preview Abstract ✓ A three-dimensional numerical simulation was carrie tangential firing ultra-supercritical boiler. The realizat (DPM) for coal particles Image: Conference Paper 01 January 2015	ed out to study the pulverized coal combustion process on a ble k-ε model for gas coupled with discrete phase model ABSTRACT PDF/EPUI

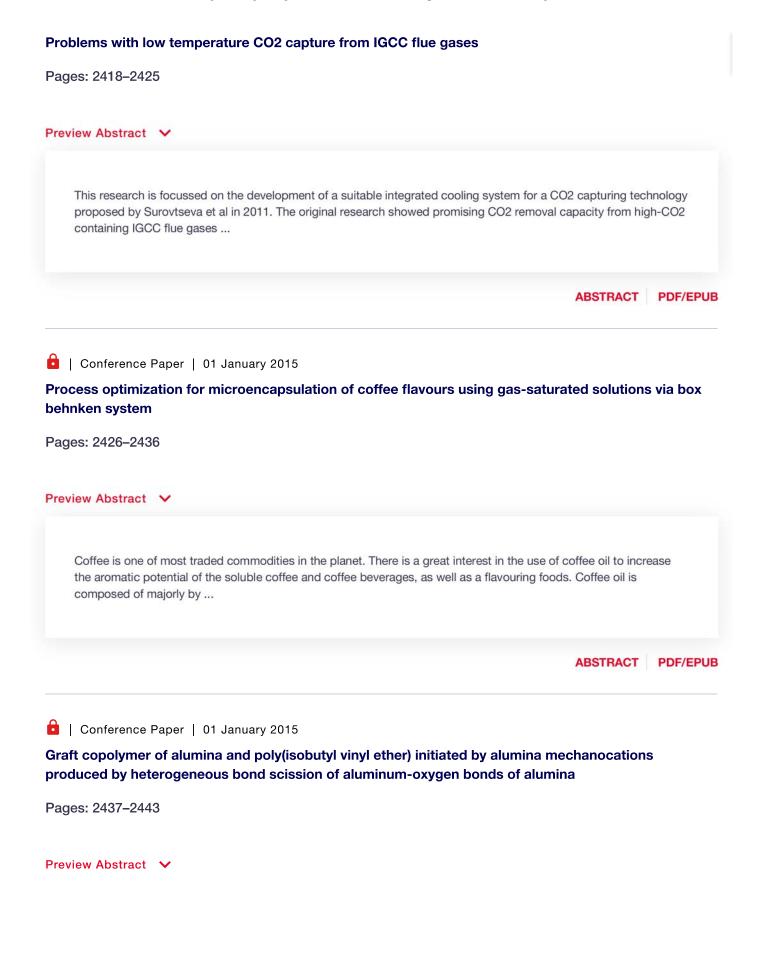


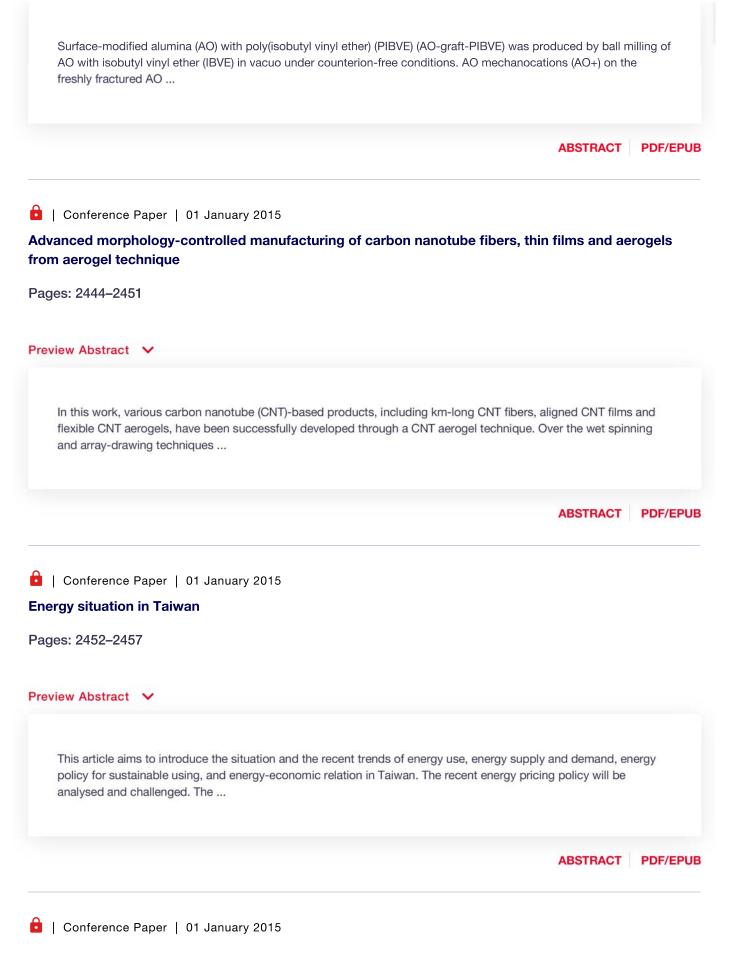


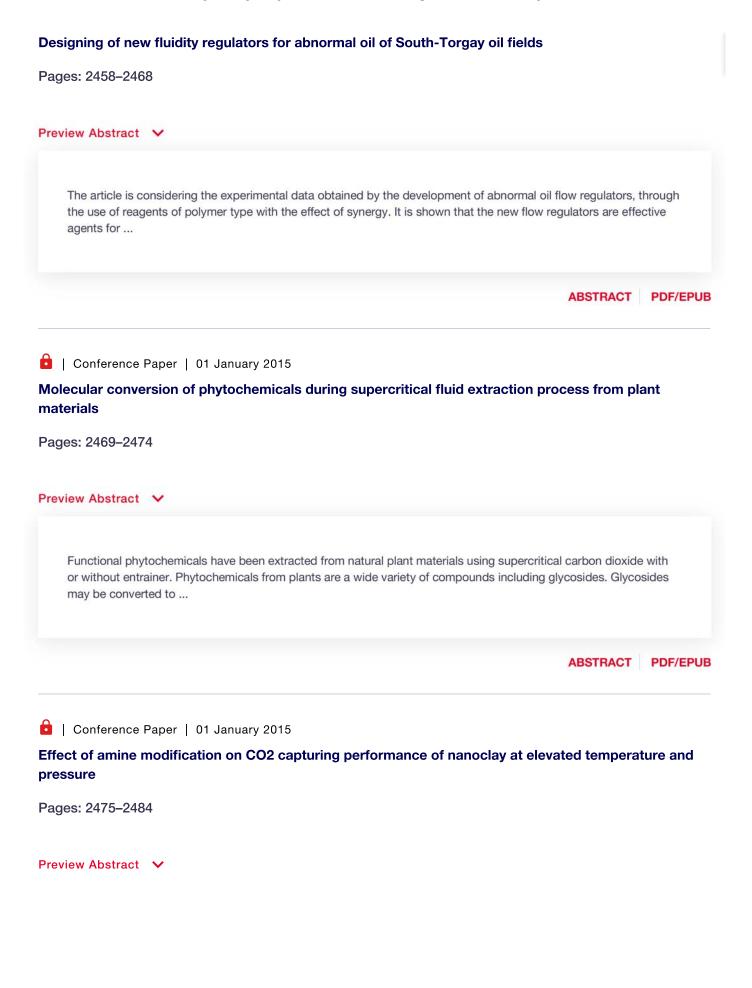


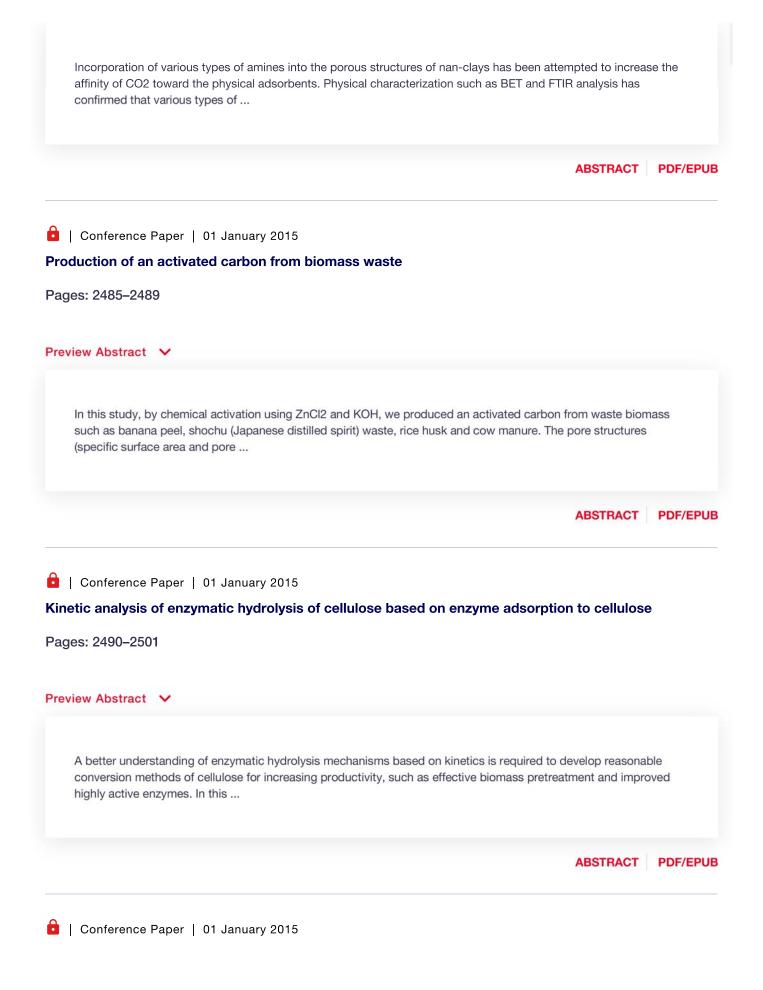


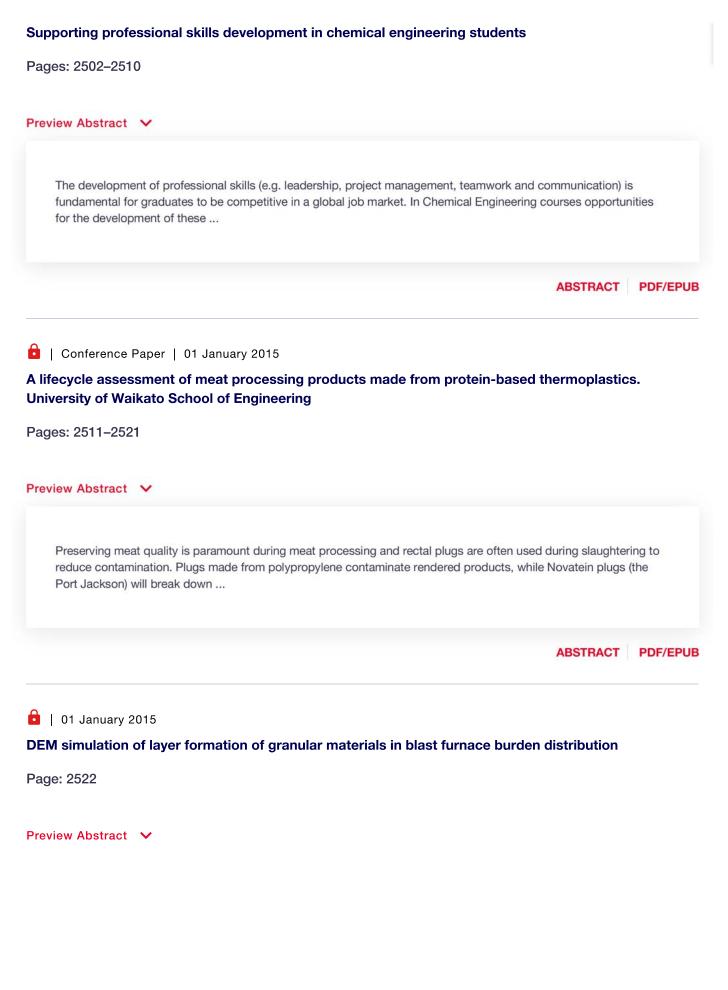


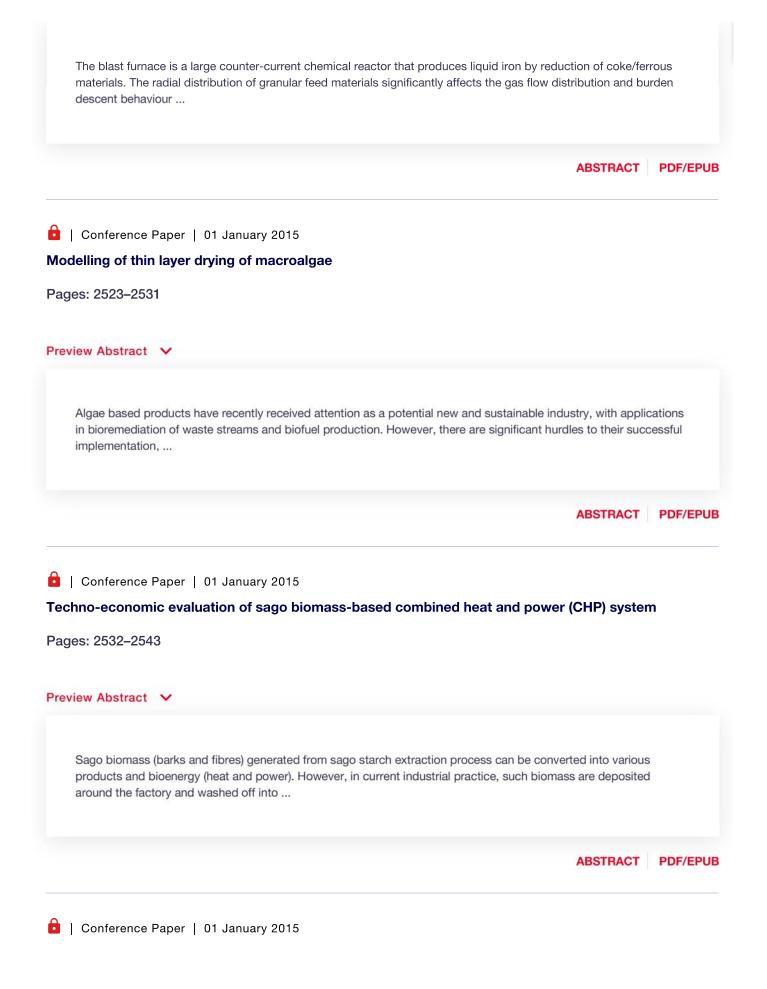


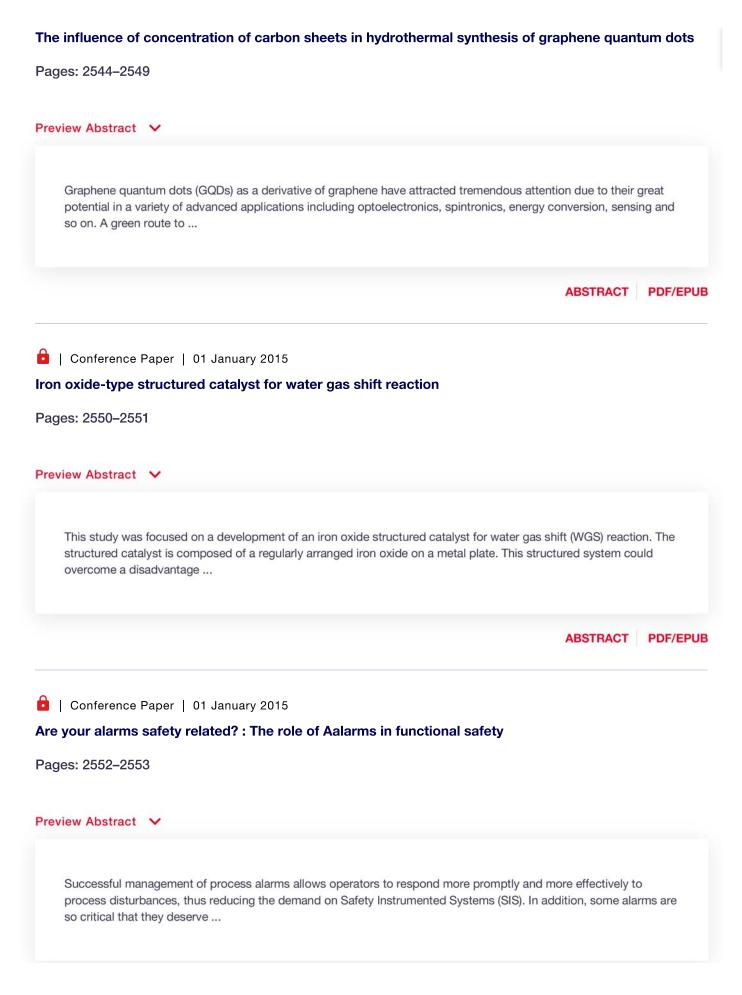


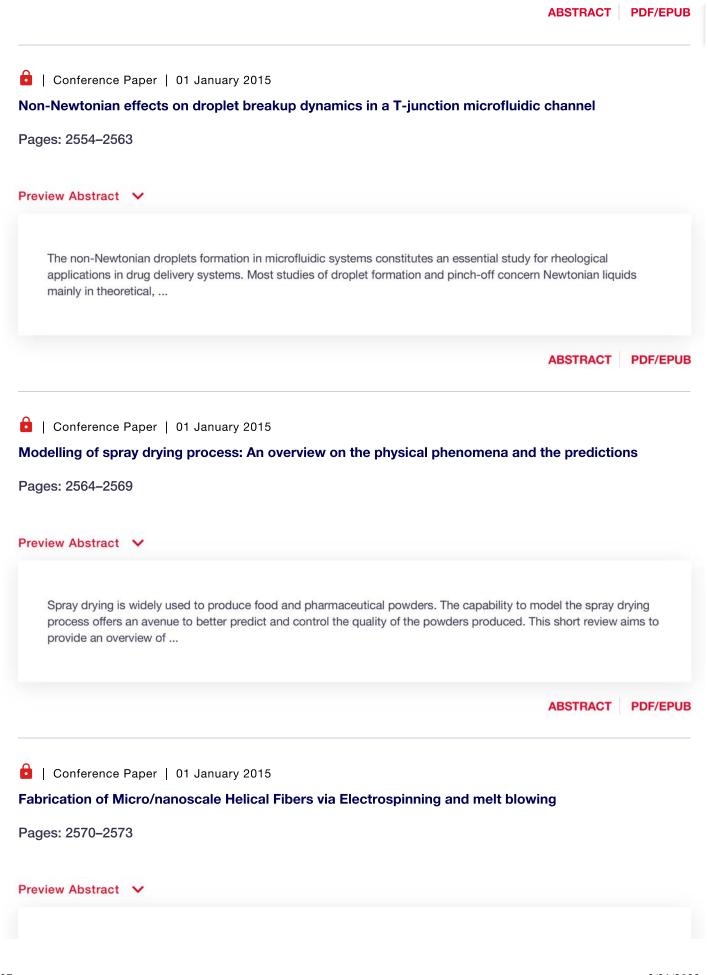


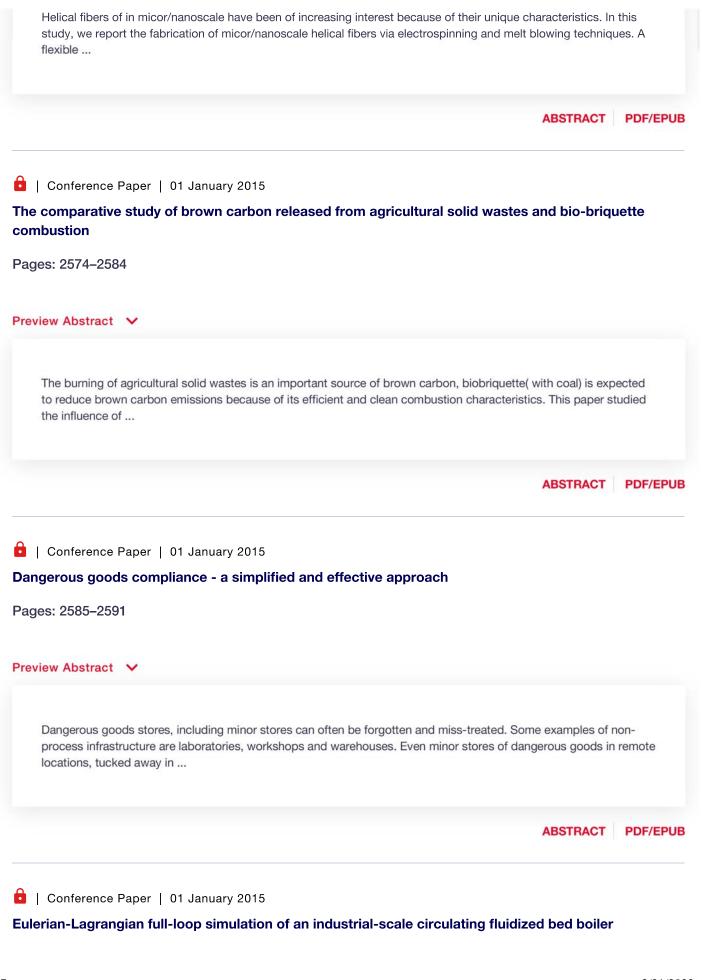




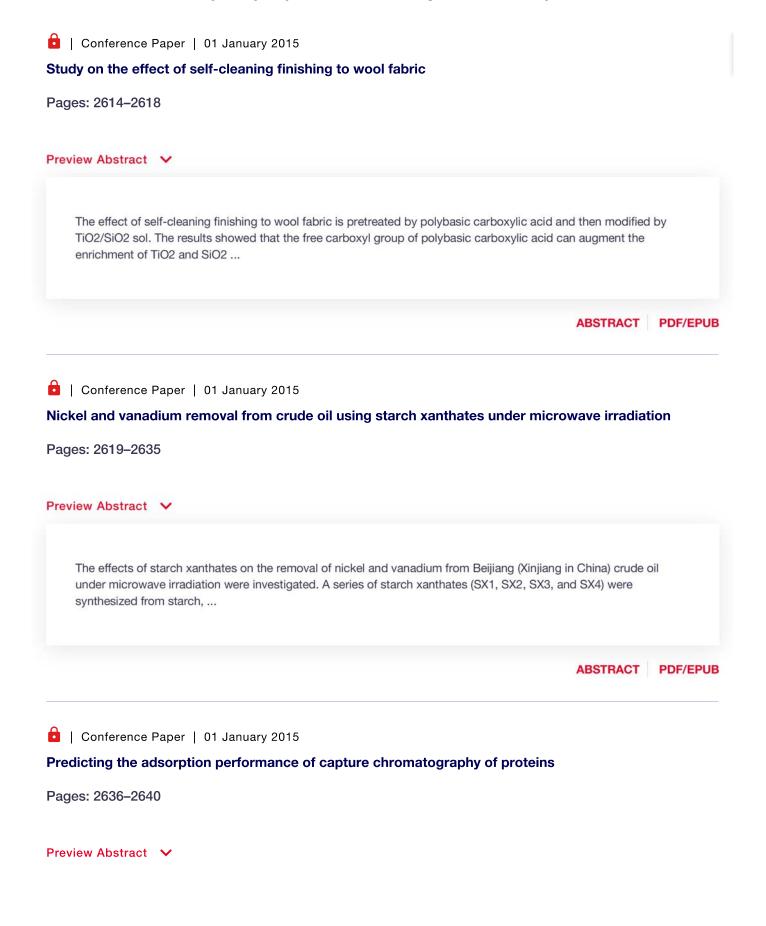


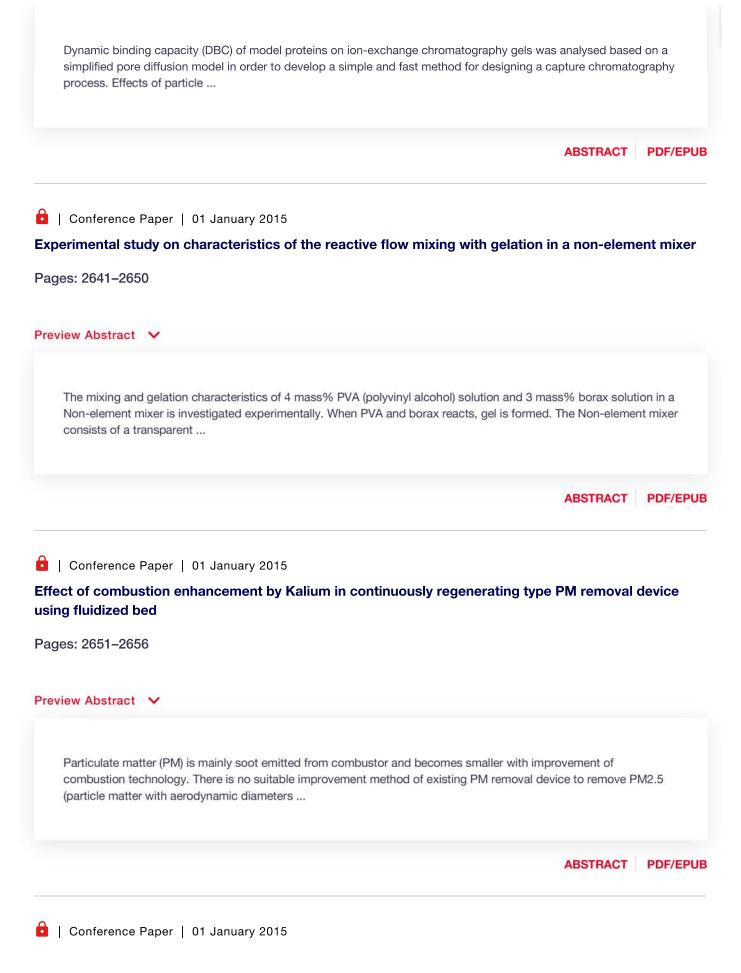


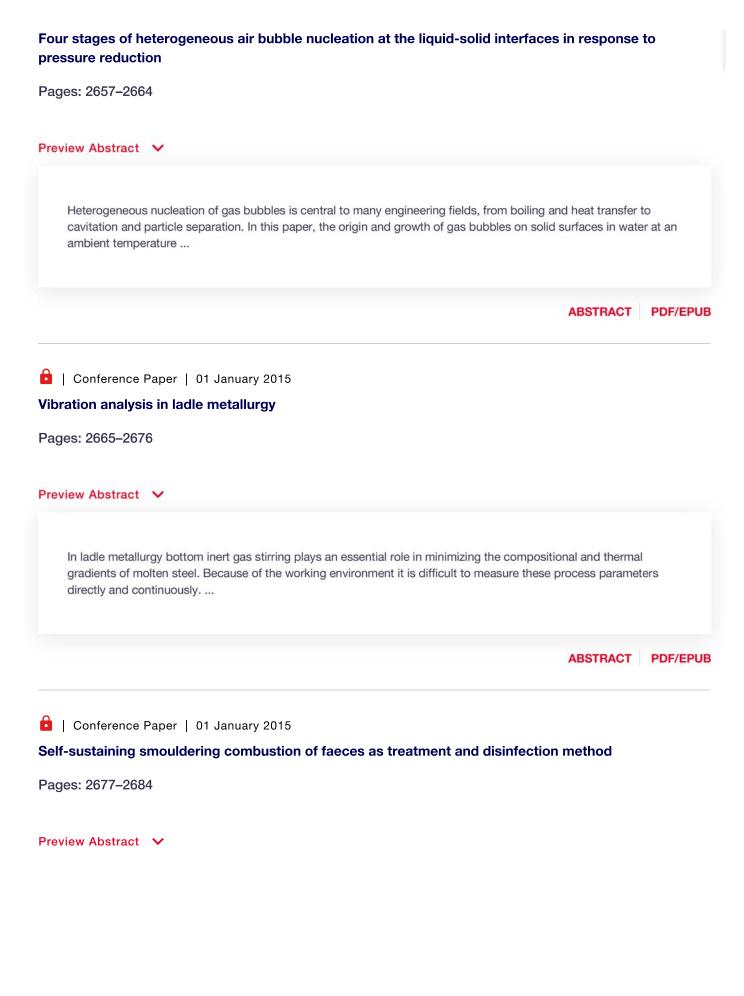




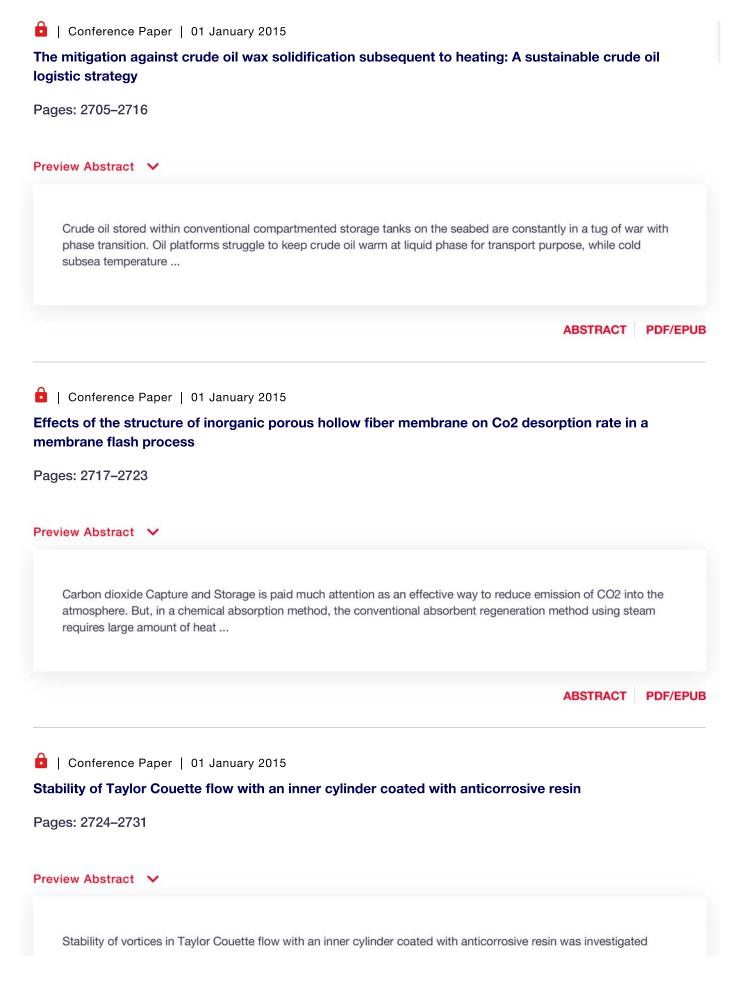
	view Abstract 🗸
	A three-dimensional (3D) full-loop simulation is adopted to predict the gas-solid flow and reaction characteristics during the combustion of mixture of municipal solid waste (MSW) and coal in an industrial-scale circulating fluidized bed (CFB) boiler
	ABSTRACT PDF/EPUB
•	Conference Paper 01 January 2015
Enz	zyme catalysis in ionic liquid based microemulsion
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Pre	view Abstract 🗸
	Room temperature ionic liquids (ILs) are molten salts at or below room temperature. They are entirely composed of

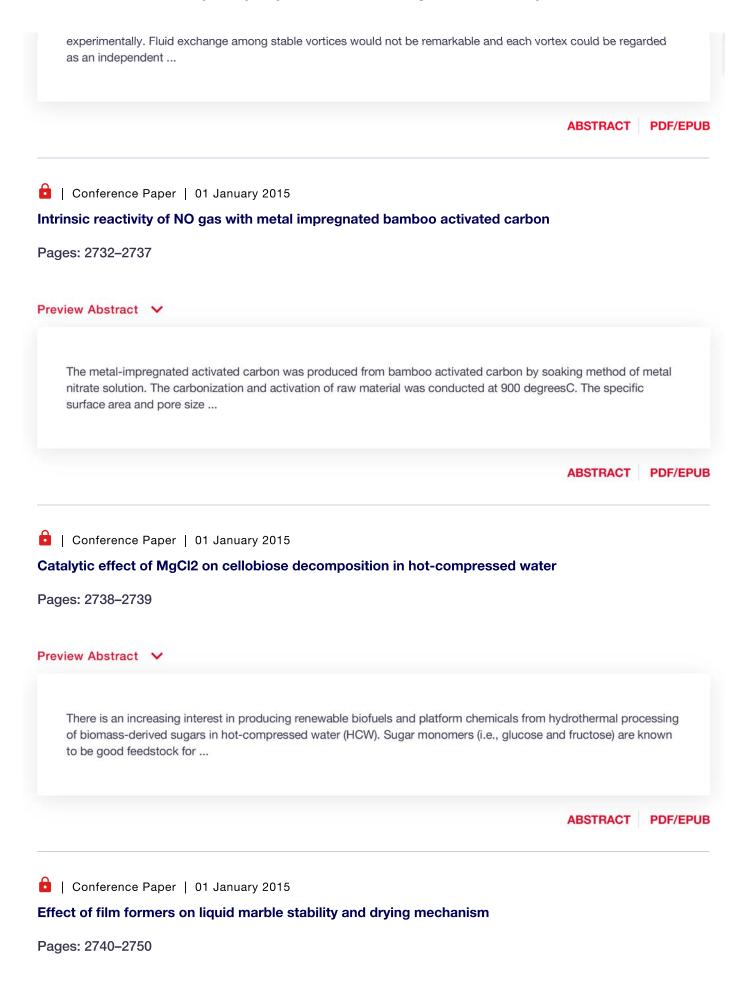


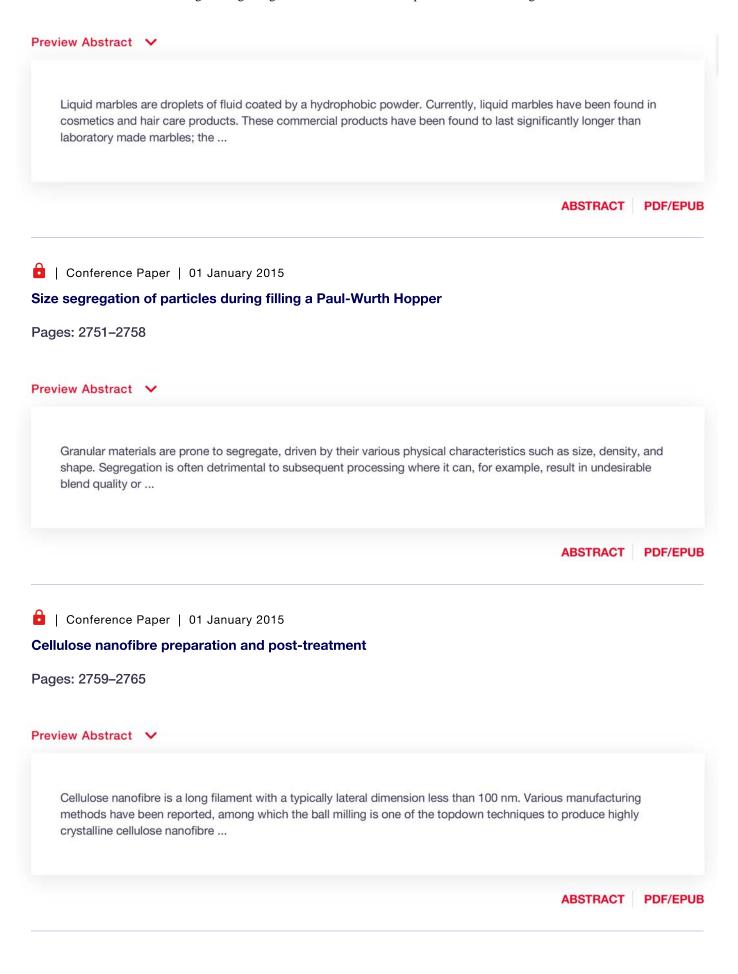


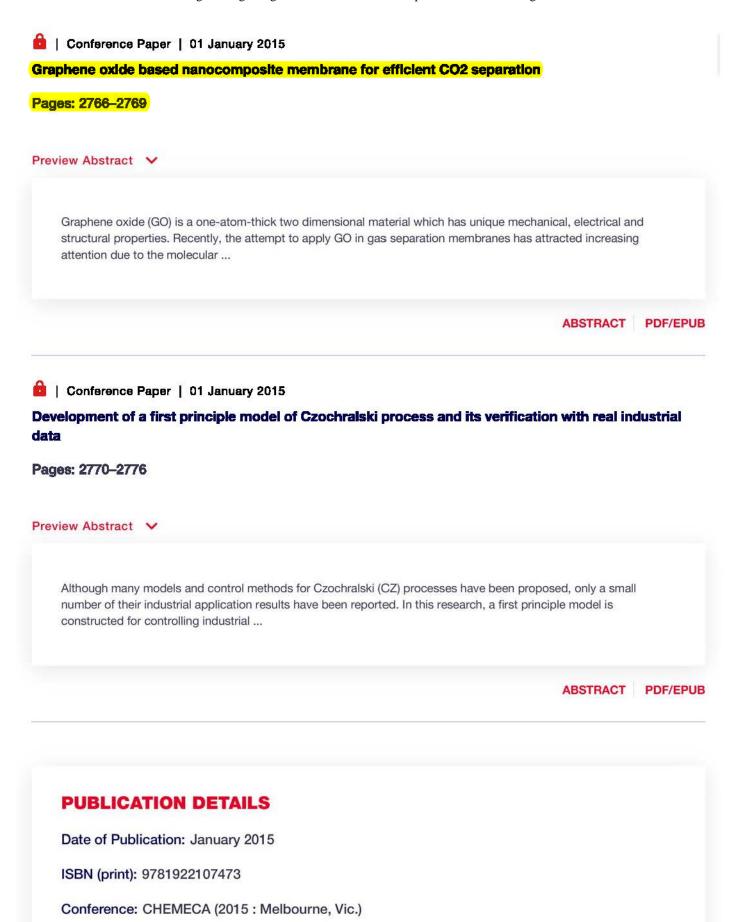


application scale,	bgy, that has been recently applied for soil remediation. Due to the high energy efficiency and the
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nthesis and applic drogenation	cation of core-shell zeolitic imidazole framework-8 (ZIF-8) catalyst in alkene
ages: 2685–2695	
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	eworks (MOF) are considered to have strong potentials as the catalyst support and host for oble-metal catalysts. Many works have been reported on the preparations and applications of F composites
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nino acid profile, a	antihypertensive and antimicrobial properties of brown seaweed ('Sargassum es obtained from pressurized hydrothermal extraction
nino acid profile, a prneri') hydrolysate	antihypertensive and antimicrobial properties of brown seaweed ('Sargassum
nino acid profile, a orneri') hydrolysate ages: 2696–2704 eview Abstract 🖍	example antimicrobial properties of brown seaweed ('Sargassum es obtained from pressurized hydrothermal extraction









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