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Dossier

Second and Third Generation Biofuels: Towards Sustainability and Competitiveness
Seconde et troisième génération de biocarburants : développement durable et compétitivité

BioTfueL Project: Targeting the Development of Second-Generation Biodiesel and Biojet Fuels

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Résumé — Le projet BioTfueL : un projet de développement de biogazole et biokérosène de 2e génération — Les biocarburants de 2e génération ont un rôle important à jouer en ce qui concerne le pool carburant dans le cadre de la transition énergétique. Utilisant comme matière première de la biomasse non comestible, ces carburants éviteront toute compétition directe avec un usage alimentaire de cette ressource. Parmi les biocarburants de 2e génération, la voie BTL consiste en la production de distillats moyens (gazole moteur et carburéacteur) *via* gazéification et synthèse Fischer-Tropsch (FT). Ces carburants sont communément appelés des « *drop in fuels* », ce qui signifie que techniquement ils ne nécessitent aucune modification du véhicule quelque soit leur taux d'incorporation dans les carburants traditionnels. Cette voie entre actuellement en phase préindustrielle pour laquelle une démonstration est nécessaire. Cet article présente le projet BioTfueL qui est porté par Axens, le CEA, IFP Energies nouvelles, Sofiprotéol, ThyssenKrupp Uhde et Total. Ce projet propose le concept original de co-traitement (la biomasse peut être gazéifiée avec des charges fossiles) et propose de développer et démontrer une chaîne complète de procédés qui sera commercialisée mondialement au moyen de ventes de licences.

Abstract* — *BioTfueL Project: Targeting the Development of Second-Generation Biodiesel and Biojet Fuels — *2nd generation biofuels will have an important part to take in the energy transition as far as fuels are concerned. Using non edible biomass, they will avoid any direct competition with food usage. Within 2nd generation biofuels, the BTL route consists in the production of middle distillates (Diesel and jet fuel)* via *gasification and Fischer-Tropsch (FT) synthesis. These fuels are called "drop in" fuels; this means that to be used they technically do not request any modification in the vehicle whatever the blending rate with conventional fuels. This route is currently at the pre-industrial phase where demonstration is required. This article presents the BioTfueL project which has been created by Axens, CEA, IFP Energies nouvelles, Sofiprotéol, ThyssenKrupp Uhde and Total. This project is focused on the original concept of co-processing (biomass can be gasified together with fossil feedstock) and proposes to develop and demonstrate a full process chain to be commercialized worldwide via licensing.*

ABBREVIATIONS

AGR	Acid Gas Removal
BFW	Boiler Feed Water
BTL	Biomass To Liquid
FT	Fischer-Tropsch
kbdoe	Kilo barrels of oil equivalent per day
LCA	Life Cycle Analysis
Mbdoe	Million barrels of oil equivalent per day
MW_{el}	Mega Watt of electrical power
MW_{th}	Mega Watt of thermal power
PSG	PRENFLO® with Steam Generation
PDQ	PRENFLO® with Direct Quench
LHV	Lower Heating Value
WGS	Water Gas Shift

INTRODUCTION

Second-Generation Biofuels

Nowadays, the transport sector is facing an unprecedented challenge. Oil currently accounts for more than 92% of the energy used in transport [1] and global demand for fuel continues to grow. Oil resources, exhaustible by nature, can not indefinitely meet those needs.

Moreover, due to global warming considerations, the control of GHG (GreenHouse Gases) and in particular CO_2 emissions is increasingly present. To protect the environment and conserve natural resources, a more diverse energy mix is essential, particularly in the transportation industry. As the only liquid fuels that can be used to supplement fossil-fuel-based transportation fuels, biofuels play a major role in the diversification process.

First-generation biofuels, blended with conventional gasoline or Diesel are available at the gas pump. They can be divided into two main categories — biodiesel, which is produced from a variety of oils, including rapeseed, sunflower and soybean, and blended with conventional Diesel and ethanol, which is produced by fermenting sugars or starch and blended with gasoline.

Researchers and producers are now working to develop second-generation biofuels that can be made from non-edible, lignocellulosic materials derived from wood, straw, forest waste and dedicated crops.

By using the non-edible parts of plants, second-generation biofuels enable to meet growing biofuel needs without directly competing with food production. In addition, they use raw materials that are in abundant supply and deliver an interesting environmental performance.

Second-generation biofuels can be used alone or blended with conventional gasoline, Diesel or jet fuel as drop-in fuels. Two main pathways are being explored — biochemical conversion and thermochemical conversion as presented in Figure 1. The thermochemical approach can be divided in two main routes. A direct route using a primary liquefaction step of the solid biomass to a bio-liquid (based on thermochemical processes), usually called bio-oil or bio-crude, followed by a dedicated upgrading of the bio-liquid into biofuels. This route includes liquefaction processes without hydrogen addition nor catalyst (fast pyrolysis) or with catalyst (catalytic pyrolysis) or with catalyst and hydrogen addition (hydropyrolysis, hydroliquefaction). Most of these solutions are at research and development stage and will not be addressed in the present paper. The second route is an indirect one based on preliminary gasification of the biomass into CO and H_2 called syngas, followed by a purification step and a final conversion of the purified gas into biofuels *via* Fisher Tropsch synthesis. This route is usually called Biomass To Liquid (BTL). It is currently at a pre-industrial stage.

The aim of the BioTfueL project presented in the following pages is to develop a process chain capable to produce second-generation biofuels (mainly middle distillates) using thermochemical conversion through the gasification of biomass. This type of process chains is described in the literature [2], but most of them are dealing with a single feedstock type [3, 4] and the concept of co-processing with fossil feedstock proposed by BioTfueL is seldom discussed [5].

The BioTfueL Project

BioTfueL is a project to develop and market an end-to-end chain of technologies to produce second-generation biodiesel and biojet fuel that will use the thermochemical conversion.

The very high quality biodiesel and biojet fuel produced using this process chain will be free of sulphur and aromatic compounds and technically suitable for use in all types of Diesel and jet engines, either pure or in blends (according to regulations) as drop in fuels.

The BioTfueL project will further develop technologies for converting lignocellulosic biomass — such as straw, agriculture wastes and forest residues — into synthetic fuel, by validating their technical and economic feasibility in demonstration units and optimizing their energy efficiency and environmental impact.

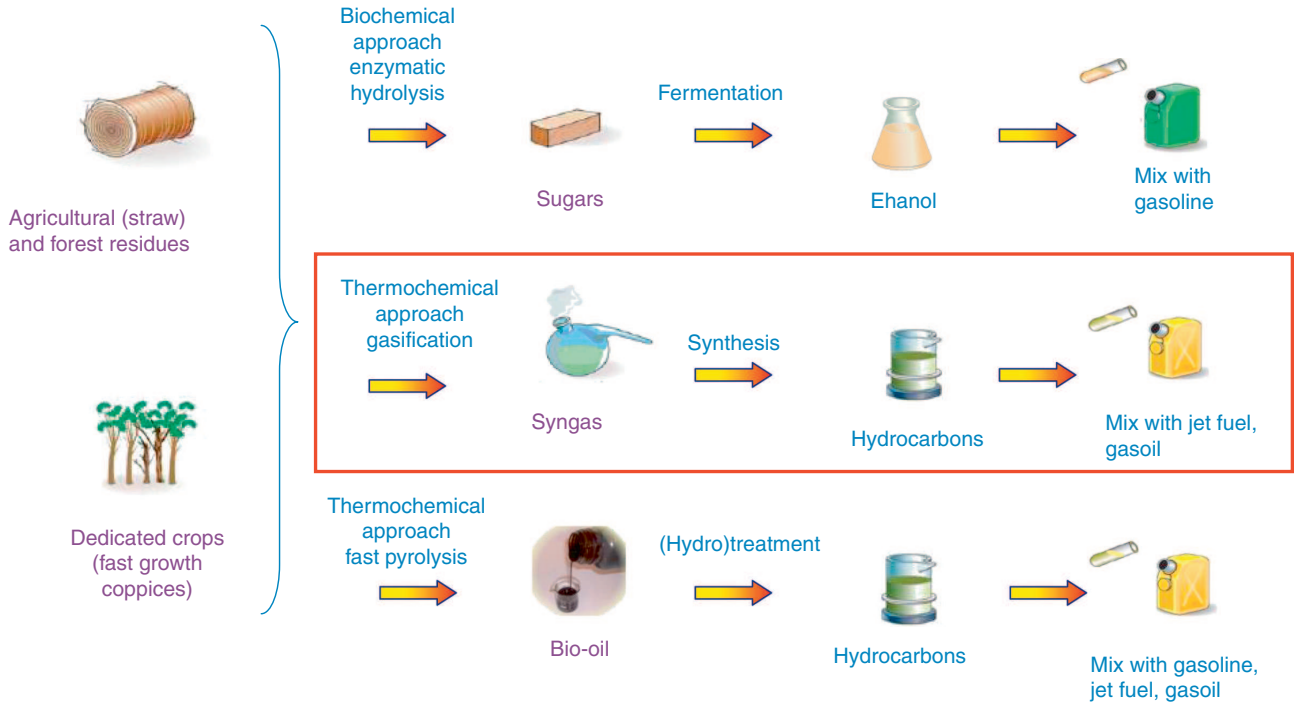


Figure 1

Main routes for second-generation biofuels.

On completion of the BioTfueL project, the process chain will be ready for the transition to industrial-scale production.

In cooperation with the biomass supply industry, Life Cycle Assessments (LCA) include the environmental impact of biomass production, the transportation to the plant, the transformation into biofuel, the combustion of the biofuel, as well as the impact of land use change. The BioTfueL project also examines the possibility of locating pretreatment units in regions that have an abundant supply of biomass, so as to densify biomass material early on in the biofuel production process.

The high quality of the fuel produced, its total compatibility with current fuels and BioTfueL's co-processing-based approach (co-processing biomass with fossil fuel feedstocks ensures a continuous supply to the plant despite seasonal variations in biomass availability), should ensure a global market for the BioTfueL process chain. This process chain will be licensed by *Axens*.

Key Figures

Partners: *Axens, CEA, IFP Energies nouvelles, Sofiprotéol, ThyssenKrupp Uhde and Total*.

Budget: €112.7 million, of which €33.3 million in public funding.

Date for market launch of the process chain: by 2020.

Location of the biomass pretreatment demonstration unit: *Sofiprotéol*'s site in Venette, France.

Location of the gasification, purification and synthesis demonstration unit: *Total*'s site in Dunkirk, France (*Établissement des Flandres*).

1 MARKET AND POLICY CONTEXT

1.1 Prospective of Global Energy Mix and Market Share for the Second-Generation of Biofuels

The overall energy mix is changing with a tendency to reduce the share of fossil energies in the next 20 years (*Fig. 2*). In this change, the fastest growing fuels are renewable (including biofuels) since the rate of growth is expected to be 8.2% p.a. in the period 2010-2030 [6], many nations are seeking to reduce petroleum imports, boost rural economies, and improve air quality through increased use of biomass. Within this frame, the share of second-generation biodiesel could raise up to 8% which means an equivalent of 300 kbdoe (*Fig. 3*). Considering BTL industrial plants able to handle at least 1 Mt/yr of dry biomass with a minimum 17 wt% mass yield [7], it paves the way for an important number of industrial plants.

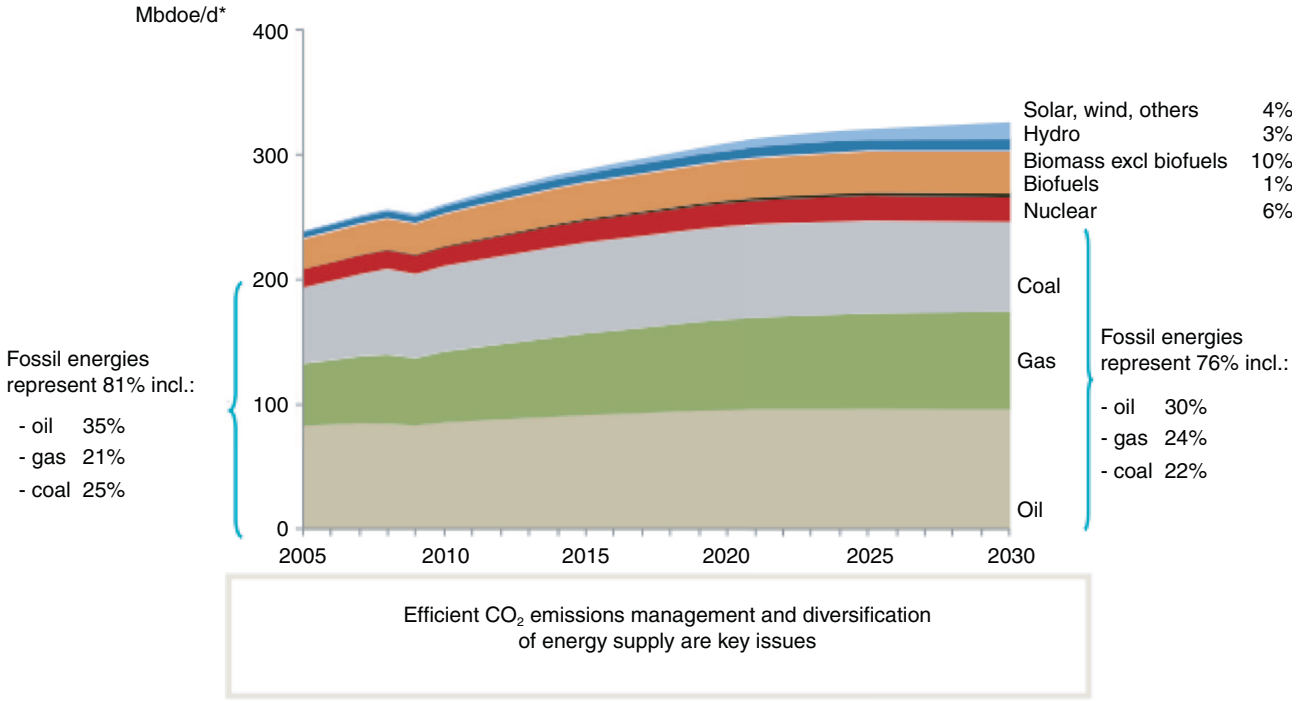


Figure 2

2030 a new energy mix.

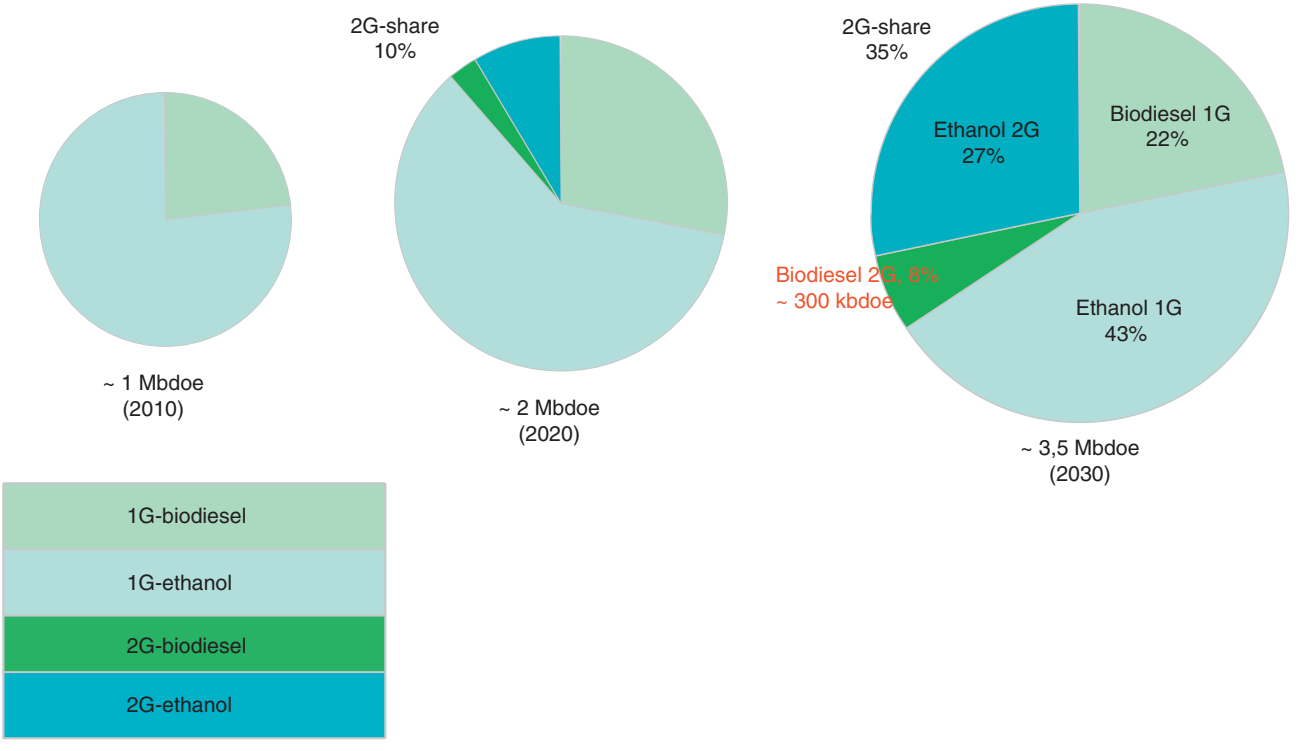


Figure 3

Shares of biofuels in the future (internal source).

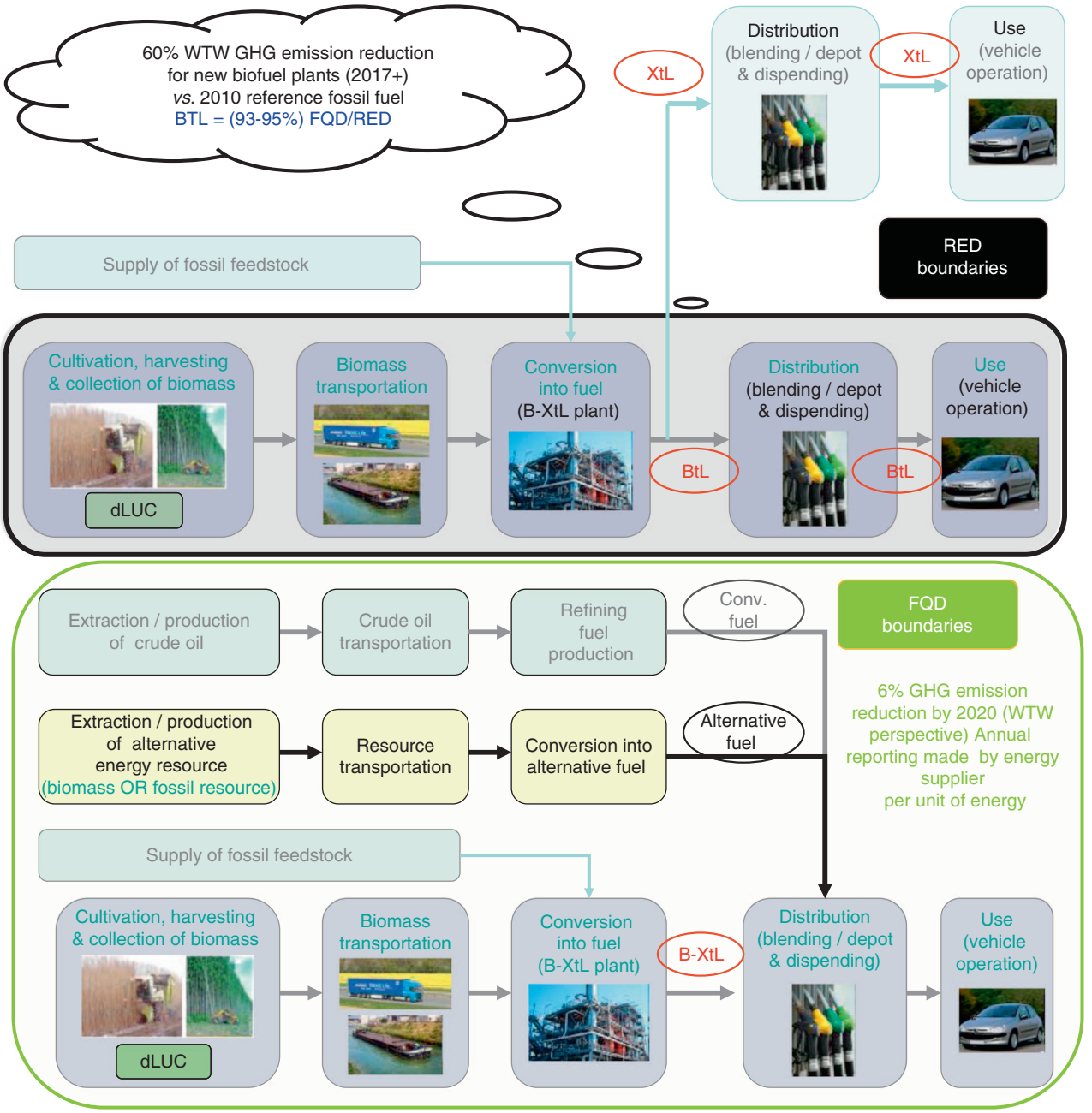


Figure 4

RED and FQD battery limits.

1.2 Policy Context: the Example of European Union

European Union standards limit the biocontent of fuels sold at the pump. For Diesel, the limit is currently 7%(v/v). Within this limit, the actual percentage of biofuels in gasoline and Diesel blends varies from one EU country to the next.

Two main directives are currently driving Biofuels regulation in Europe, the Renewable Energy Directive [8] (RED) and the Fuel Quality Directives [9] (FQD).

The RED provides for the percentage of renewable energies used in the transportation industry to increase to 10% by 2020. The 10% target will be achieved by

gradually supplementing first-generation biofuels with second-generation biofuels. The RED also states that biofuels must meet sustainability criteria focusing on the production process itself from the field to the wheel (*Fig. 4*). Any new biofuel plant to go in operation after end 2016 should deliver fuels with a green house gases emission reduction higher than 60% when compared with reference fossil Diesel oil (83.8 gCO_2 eq/MJ WTW). Currently, the RED proposes a default value of green house gases emissions reduction in the range (93-95%) for second-generation biofuels *via* thermo-chemical route (Fischer-Tropsch Diesel).

The FQD is focusing on the whole fuel pool (*Fig. 4*). It is mandatory for fuel suppliers to be in the position to reduce by 6% from 2011 to 2020 the Green House Gases (GHG) emissions of their pool.

On October the 17th 2012, a draft of EU Directive [10] was released to amend Directive 98/70/CE Directive 2009/28/CE. Member States should amend and vote such a draft which is a long process, so the final version of the Directive is not expected before end 2013. The draft Directive objectives are to start the transition to biofuels that deliver substantial GHG savings when also estimated indirect land-use change emissions are reported, while existing investments should be protected. So, the aims of the current proposal are to:

- limit the contribution that conventional biofuels (with a risk of *ILUC* emissions) make towards attainment of the targets in the Renewable Energy Directive;
- improve the GHG performance of biofuel production processes (reducing associated emissions) by raising the GHG saving threshold for new installations (60% for plants that will start after July 1st 2014 – previously 35% for plants starting up to end of year 2016 and 50% after January the 1st 2017) subject to protecting installations already in operation on 1st July 2014 (35% GHG emissions reduction is active till December 31st 2017 – previously December 31st 2016 – and 50% GHG emissions reduction is mandatory from January 1st 2018);
- encourage a greater market penetration of advanced (low-ILUC) biofuels by allowing such fuels to contribute more to the targets in the Renewable Energy Directive than conventional biofuels;
- improve the reporting of GreenHouse Gas emissions by obliging Member States and fuel suppliers to report the estimated indirect land-use change emissions biofuels.

This type of regulation is providing a favorable framework for the development of second-generation biofuels.

It is worth to note that in the US, since July 2010, the Renewable Fuel Standard (RFS) program 2 [11] replaced the RFS1. The RFS2 specifies the volumes of cellulosic biofuel, biomass-based Diesel, advanced biofuel, and total renewable fuel that must be used in transportation fuel up to 2022. These volumes are revised every year.

2 AN AMBITIOUS R&D PROGRAM TO OPTIMISE SECOND-GENERATION BIODIESEL AND BIOJET FUEL

This overall frame paves the way for a successful development of the BTL technology if developed paying specific attention on *key technological issues* and using a development methodology that *guaranties a safe scale-up*. This is the way BioTfueL project has been conceived:

- identification and critical analysis of the issues for scale-up;
- studies of the different technical options in order to reach the best way to solve those issues using modeling, pilot plant, and cold mock up;
- from the results of those preliminary studies, design of the demo plant as the best suited tool within the framework of a complete scale-up strategy program.

2.1 BioTfueL Process Chain: Main Hurdles

BioTfueL will focus on a four-stage industrial process presented in Figure 5. In order to be in the position to provide to the market a flexible and reliable process chain, it is compulsory to tackle any issue during the project.

Biomass pretreatment: 1st step

The biomass must be pretreated, dried and pulverized so that it can be injected under pressure into a gasifier *via* fluidization and pneumatic conveying. The BioTfueL project partners have decided to use a low-temperature pretreatment process known as torrefaction. Torrefaction is a mild thermal treatment of the biomass under inert atmosphere conducted in the range of temperature of (250-300°C), at atmospheric pressure with a residence time in the range of 20 to 60 minutes. A typical mass yield for solid material (dry basis) is in the range of ≈70% corresponding to an energy yield for woody biomass (dry basis) ≈90%. The torrefaction gas, which contains only 10% of the energy of the biomass can be used internally for the torrefaction process [12].

According to the severity of the thermal treatment, it is possible to decrease the energy for grinding significantly [13]. The torrefied material obtained is more brittle, its fluidization properties — particle size and morphology — are enhanced and the Lower Heating Value (LHV) is also increased from 18-19 up to 22-23 MJ/kg [14].

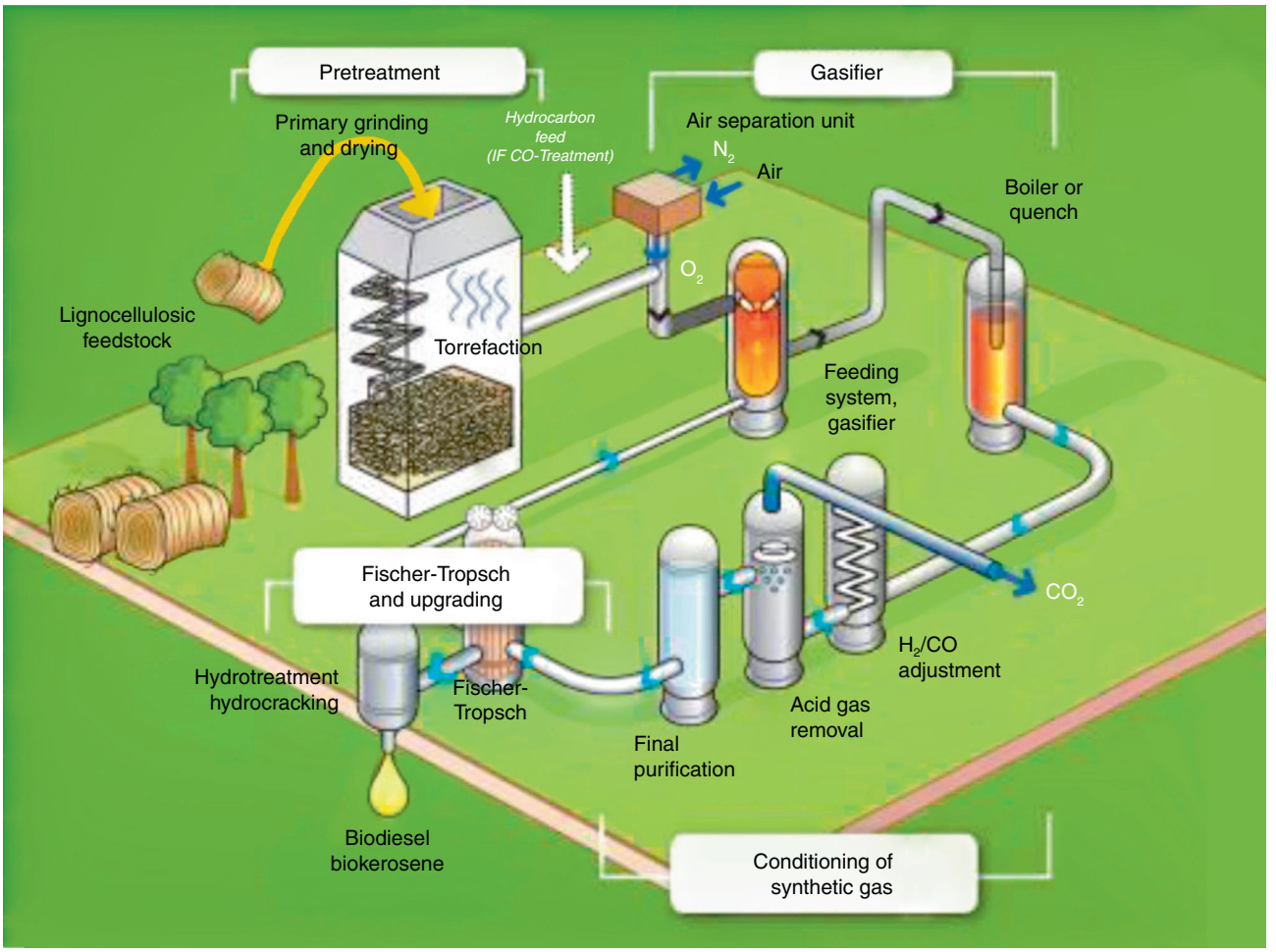


Figure 5

Second-generation biodiesel and biojet fuel production chain.

As far as the biomass torrefaction is concerned, it is mandatory to be in the position to obtain a product easy to grind without facing considerable mass losses. Moreover the final product must be homogeneous. Currently, there is no industrial plant capable to deliver large flow rates (typically 10 to 30 t/h) while following the previous requirements.

The pretreatment and torrefaction takes places at *Sofiproteol*'s site in Venette, while the subsequent process steps are performed at *Total*'s site in Dunkirk.

Torrefied biomass gasification: 2nd step

Once the torrefied biomass has been ground into a powder (50-200 μm), it is injected into a gasifier where it is exposed to very high temperatures of between 1 200 and 1 600°C in the presence of oxygen to convert it into syngas. The conversion takes less than two seconds, for a conversion efficiency of more than 99%. A pressurized, oxygen blown, entrained-flow gasifier has been selected by BioTfueL's partners, because it offers high levels of treatment capacity and the greatest flexibility for treating a wide variety of biomass feedstock, and because it generates the purest syngas. Moreover a pressurised gasification step enables to avoid any compression of the syngas before the Fischer-Tropsch unit operated at 20-25 bar. The partial oxidation conducted under pure oxygen avoid the presence of nitrogen in the down stream sections hence a diminution of the size of the equipment and increase catalytic activity (no dilution effect). Another advantage of this type of gasification technology is the use of membrane wall as heat protection. As shown in Figure 6, all tubes are cooled by Boiler Feed Water (BFW) producing steam. The membrane wall not only has the advantage that it is designed to last

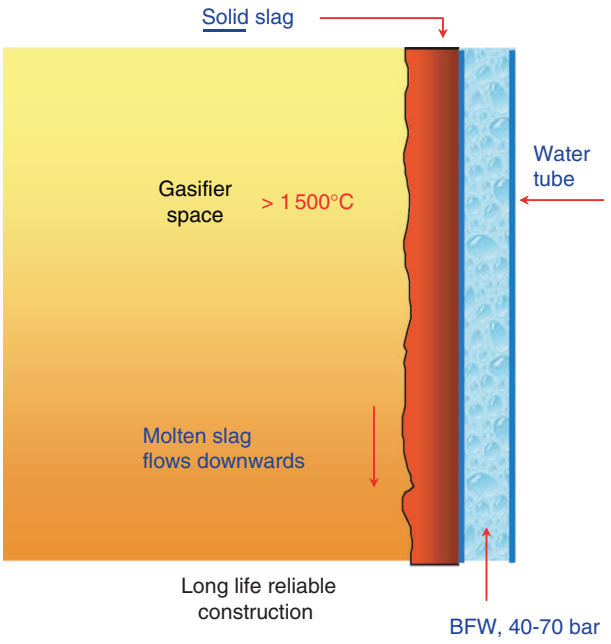


Figure 6

PRENFLO membrane wall protected by a slag layer.

the lifetime of the project with virtually no maintenance, it also has the advantage that gasification at high temperature is possible connected with a long life reliable construction. In the PRENFLO® with Direct Quench (PDQ) gasifier the feedstock is pneumatically transported to the gasifier in pulverized form. The gasification reactions take place under a pressure of 30 to 42 bar. The produced synthesis gas is leaving the reactor at the bottom is cooled down by a water quench to about 220°C. Thus, the syngas is fully saturated and ready for the Water Gas Shift (WGS) conversion reaction to enhance the amount of hydrogen in the syngas by reaction of CO with water to form hydrogen and carbon dioxyde. This type of reactor is preferred if a shift reaction is following the gasification to enhance the hydrogen to carbon monoxide ratio for the Fischer-Tropsch process.

The PRENFLO® with Steam Generation (PSG) technology is already used commercially to treat fossil fuel feedstock (*Fig. 7*). However, significant changes are required to enable the injection of biomass into the gasifier, either alone or mixed with fossil fuel feedstock in varying proportions. These changes represent an important technological challenge. The PRENFLO® PSG was first commercialized in the Fürstenhausen,

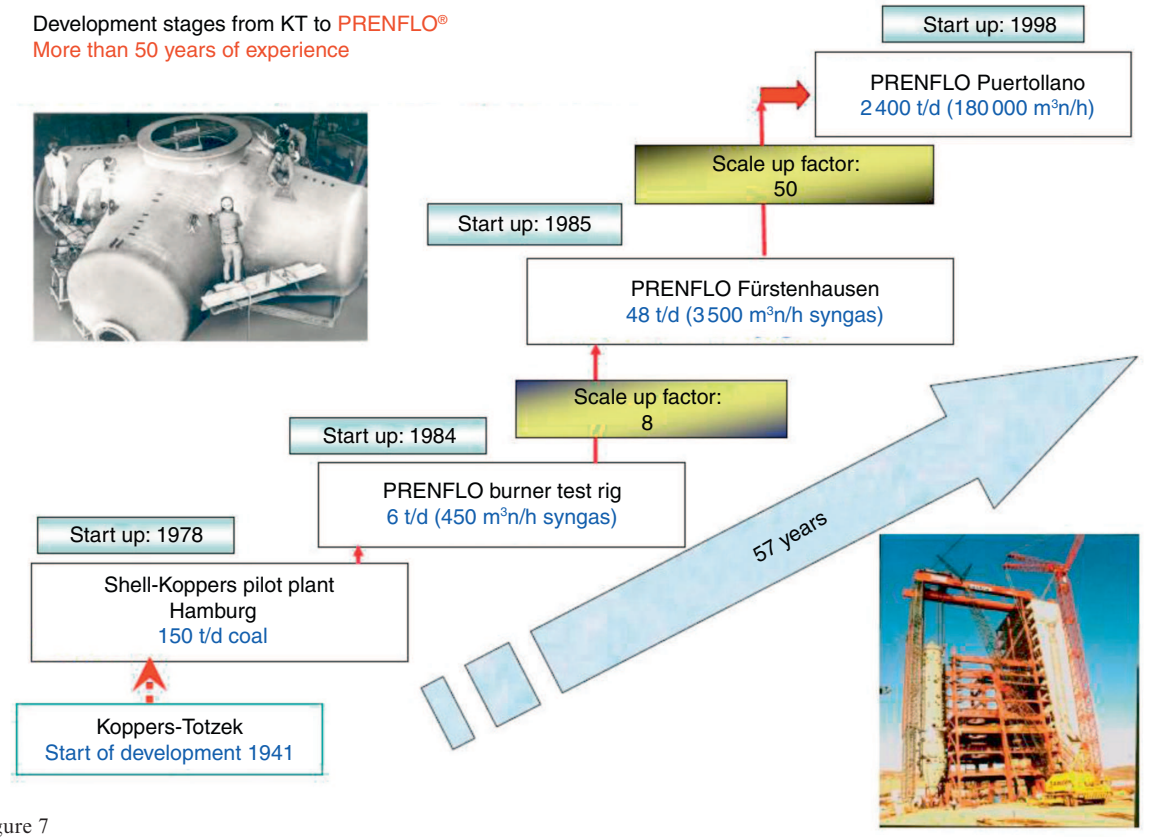


Figure 7

Development stages for the PRENFLO gasification technology.

Germany, demonstration plant, and thereafter selected by *Elcogas S.A.*, Spain for the Puertollano IGCC plant. It should be noted that the Puertollano plant, which utilizes the PRENFLO® technology and was designed and built by *ThyssenKrupp Uhde*, is still the world's largest solids based IGCC (Integrated Gasification Combined Cycle) with an output of some 300 MW_{el} from a single train. The PRENFLO® technology is the result of more than 50 years of development [15].

Syngas clean up and conditioning: 3rd step

To be converted into liquid fuel, the syngas must be very pure and have the correct chemical composition. Indeed, the syngas produced in the PRENFLO gasifier has a low H_2/CO (0.5-0.7) ratio when compared with the requirement of the FT reactor (almost 2), hence WGS reaction is performed on a part of the syngas, and the CO_2 formed is removed with H_2S and other impurities using an Acid Gas Removal (AGR) unit. An AGR plant is quite a common process for natural gas treatment as well as for refinery gas treatment. An AGR plant is basically formed of two sections: one section dedicated to the absorption of impurities into the solvent and one section dedicated to solvent regeneration. According to the pollutants to be removed and the licensors, the solvent can be a physical solvent (such as methanol) or a chemical solvent (such as amines).

Since the FT catalyst requires very low levels of impurities [16], a final purification step is performed with guard beds (chemisorption). Involving several processes, the purification of biomass-derived syngas is highly complex and has not yet been implemented on an industrial scale. It begins with relatively mature cleaning processes, which are followed by a final purification process that uses special high-performance catalysts. These need to be further developed to enable continuous use in industrial-scale applications.

The aim of the BioTfueL project is to select the most energy and cost-efficient chain of processes possible. The technological developments will focus mainly on the final stages of the cleanup and conditioning process, when the gas is prepared for Fischer-Tropsch synthesis.

Final conversion into biofuel* via *Fischer-Tropsch synthesis and upgrading: 4th step

The Fischer-Tropsch process converts purified syngas into a mixture of hydrocarbons, this step is followed by hydrocracking and hydroisomerization stages in order to get final products: mainly middle distillates (Diesel, jet fuel), and naphtha.

Unlike the first three stages described above, no R&D work will be carried out on the FT process during the BioTfueL project, because this process is already available commercially. The Fischer-Tropsch technology for the BioTfueL project is the *Axens Gasel*^{TM} technology (technology developed by *IFP Energies nouvelles* and *ENI*) [17]. This technology involves a cobalt catalyst, maximizing the production of middle distillates, into a slurry bubble column reactor. This technology allows an isothermal profile within the reactor as well as a low pressure drop and is well suited for catalyst make-up and withdrawal. *Axens* will also provide the hydro-isomerisation processes (upgrading). *Axens* can take care of the full process route by:

- licensing a complete FT plus upgrading technology chain for the conversion of syngas to middle distillates (as jet fuel and Diesel);
- manufacturing associated FT and upgrading catalyst;
- providing a single point guarantee from syngas to final products.

BioTfueL Project Partnership

In order to overcome all those challenges, the BioTfueL project is being carried out by a consortium of six partners whose multiple areas of expertise together cover the entire biofuel production value chain.

CEA and *IFP Energies nouvelles* are the two major French research and development actors in the energy sector. *Axens* is the 2nd largest licensor in the world of refining and petrochemicals processes, *Axens* will be the licensor of the BTL process chain developed by the BioTfueL consortium. *ThyssenKruppUhde* is a first class engineering and contracting company, licensor of gas technologies processes, leader in the field of gasification. *Sofiprotéol* is the leading European biodiesel producer. *Total* is the 5th largest integrated international oil group. Those partners created a dedicated company called Bionext, to coordinate the project as well as the construction and operation of the demonstration units.

Throughout the project, research teams will focus on the following topics: resource screening and biomass pretreatment, gasification technology, syngas purification technologies, construction and operation of the biomass pretreatment demonstration plant, construction and operation of the gasification demonstration plant, syngas purification and Fischer-Tropsch process demonstration unit, financial and environmental performance (Life Cycle Analyses, LCA), technology survey, analytical and materials issues, and risk assessment.

Presentation of the Demonstration Plants

The BioTfueL demonstration plants will be realized as a multiscale demonstration plant to get the scale up data

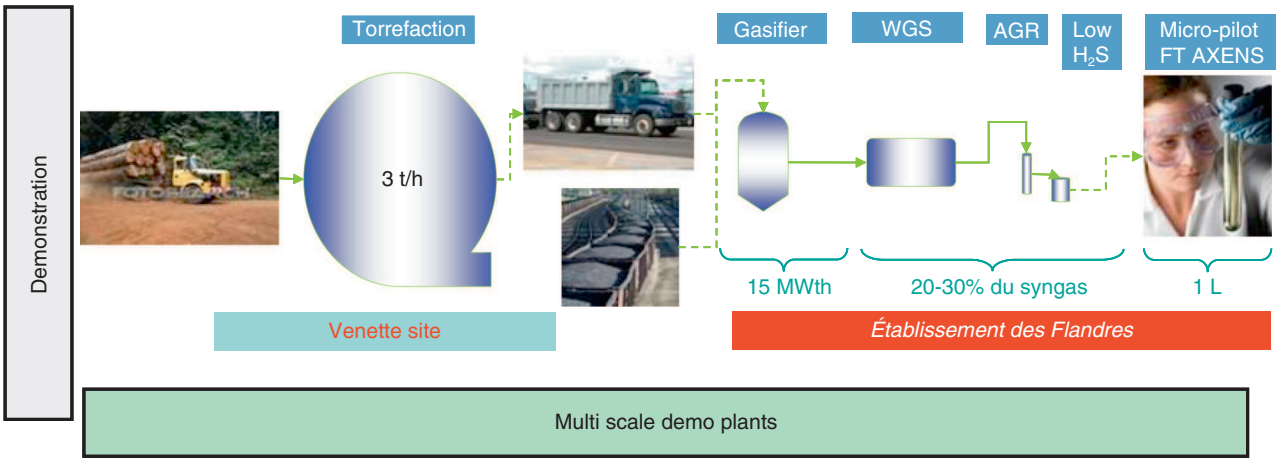


Figure 8

Presentation of the demo plant that will be constructed in the BioTfueL project.

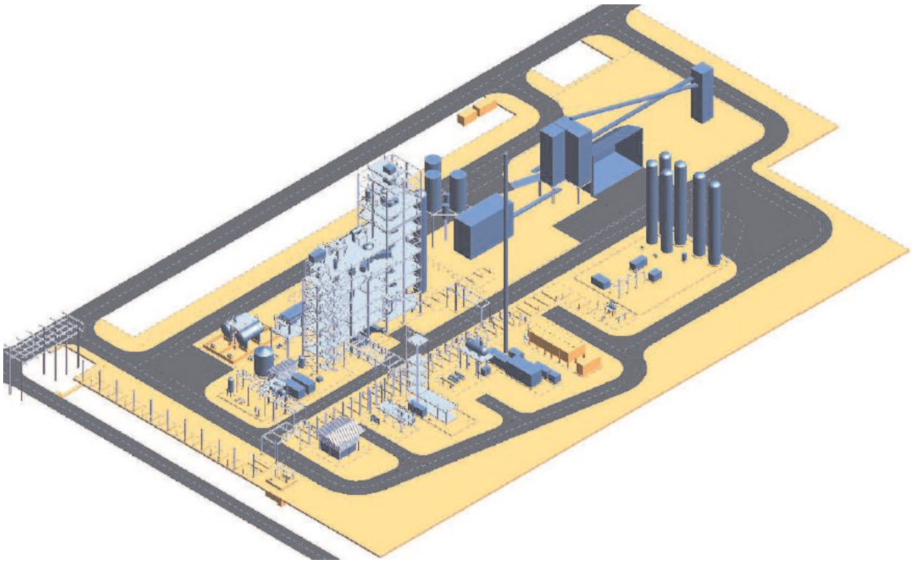


Figure 9

A 3D view of Dunkirk site demo plant.

needed and to validate the various configurations (*Fig. 8*).

The torrefaction and the gasification sections will use up to 5 tons per hour of fresh biomass, about 15 MWth for the gasifier. The Water Gas Shift (WGS), the Acid Gas Removal (AGR) and the final purification step will use only 5 to 10% of the syngas and the FT unit is designed as a catalyst poisoning small scale (1 L) test unit. The FT section does not require any technological validation, it will be used as a complementary analytical tool to qualify the syngas purification step.

In the BioTfueL project, the consortium will study the centralized and also the decentralized production scheme. The two different sites for the demonstration plants (Venette and Dunkirk) offer the opportunity to test the torrefied biomass conditioning, storage and transport as well as the optimisation of the grinding steps and the thermochemical transformation.

Biomasses which will be tested in Venette will be delivered in different forms due to the diversity of their nature and depending on their conditioning after harvesting for transport.

In parallel, torrefied biomass will be delivered to the gasification plant located in Dunkirk site (*Fig. 9*) according to quantities required for the testing programme. The road transport will be used for the demonstration project.

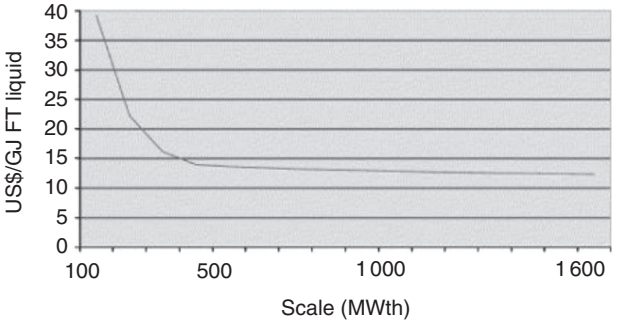


Figure 10

Illustration of plant scale impact (figure from reference [18]).

CONCLUSIONS: BIOTFUEL PROJECT ASSETS

Ensuring Higher Profitability Ratios through Feedstock Flexibility

To be profitable, second-generation biofuel production requires large industrial units [18] (*Fig. 10*) and a continuous supply of feedstock. The biomass used in the BioTfueL project is made up of forest and plant wastes, straw and other lignocellulosic materials sourced from different locations. The availability of these materials is subject to seasonal variations.

To secure continuity of supply and ensure the financial viability of production units, the BioTfueL project is targeting full flexibility through the innovative concept of co-processing. This means developing a process chain that works with a wide variety of biomass feedstock, as well as with liquid and solid fossil fuel feedstock. The target is to propose a process chain capable to handle pure biomass as well as pure fossil feedstock. This type of concept is highly favorable to be in the position to build king size plants while guarantying those plants will run at 100% capacity along the year since petroleum coke or similar fossil feedstock will easily be available to complement biomass. If the process chain uses only biomass, the fuels will be fully considered as a biofuel. If a co-gasification is performed, the fuels will be described partly as fossils fuels, partly as biofuels on the basis of energy content, in conformity with European directive ENR 2009/28 CE.

Thanks to this flexibility in the choice of raw materials, the feedstock can be adapted to offset the seasonal variations in biomass availability, thereby increasing the industry's energy efficiency and reducing production costs per ton of fuel.

Integrating Technologies across the Process Chain to Optimize Economic and Environmental Performance

The various technologies developed by the BioTfueL project partners will be brought together and validated in two demonstration plants. The objective is to identify the solutions that deliver the best technical, economic and environmental performance, by looking at such issues as return on investment, mass yield, flexibility, energy yield and CO_2 emissions. Life Cycle Assessments are carried out throughout the project to help selecting the most efficient technologies.

Financed by the ADEME's *via* the Research Demonstrators for €30 million, and by the Picardy region *via* the European Union's European Regional Development Fund, for €3.2 million, the BioTfueL R&D project is the start of a new second-generation biodiesel and biojet fuel production industry in France.

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