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THERMAL DEGRADATION PROCESS OF SEMI-SYNTHETIC FUELS FOR GAS TURBINE ENGINES IN NON-AERONAUTICAL APPLICATIONS

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ABSTRACT

This article concerns the issue of thermal degradation process of fuels, important from the perspective of the operation of turbine engines, especially in the context of new fuels/bio-fuels and their implementation. The studies of the kerosene-based jet fuel (Jet A-1) and its blends with synthetic components manufactured according to HEFA and ATJ technology, were presented. Both technologies are currently approved by ASTM D7566 to produce components to be added to turbine fuels. Test rig investigations were carried out according to specific methodology which reflects the phenomena taking place in fuel systems of turbine engines. The mechanism of thermal degradation process was assessed on the basis of test results for selected properties, IR spectroscopy and calculation of activation energy. The results show that with the increase of the applied temperature there is an increment of the content of solid contaminants, water and acid for all tested fuels. Thermal degradation process is different for conventional jet fuel when compared to blends, but also semi-synthetic fuels distinguished by different thermal stability depending on a given manufacturing technology.

Keywords: gas turbine engine, thermal stability, bio-fuels, thermal degradation process

INTRODUCTION

Gas turbine engines are currently used in many transport fields. Most commonly they are used to power modern aircraft, i.e. passenger and jet aircraft as well as helicopters. In spite of the broad application in aviation, they can be used as propulsion for power generating sets or stationary peak power plants. Besides, gas turbines become an alternative to conventional engines used in ships. Merchant ships are usually equipped with self-ignition engines and their important drawback is considerable size and mass. Gas turbines are characterized by high power in relation to their mass and have the possibility to operate with different types of fuels. Thus, this kind of propulsion is frequently applied in high-speed military ships (chasers, destroyers, motor torpedo boats) and luxurious cruise ships [1].

Current data show that worldwide there are used approximately 700 ships powered with gas turbines [2], and most of them are military ships. In the Polish Navy, only two ships are provided with such drive units: the patrol ship ORP 'Ślązak' and rocket frigate ORP 'Gen. K.Pułaski' (FFG 7).

The principle of a single - fuel policy (introduced by NATO countries) assumes use of one fuel (F-34, kerosene-based jet fuel) by aircraft, vehicles and military ground equipment during warfare [3]. As the same principle can be applied in the navy (single naval fuel), the studies were performed [4] to define potential problems.

The engines used in land, maritime and air transport from decades are designed to burn conventional fuels, i.e. hydrocarbon. In the last decades however a substantial development of bio-fuels (fuels produced from biomass) can be observed. The reason is necessity to reduce the emission of carbon dioxide (global warming). Previously, bio-fuels were dedicated only to automotive, but constant growth of air transport, observed in the last decade, led to the intensification of works to introduce bio-fuels in aviation. Additionally, the Regulations On Energy Efficiency for Ships in MARPOL Annex VI implemented in 2013 resulted in intensified search for alternative energy sources also in this transport branch [5].

Conventional fuels, including fuels for turbine aircraft engines, are in fact a blend of different (above 1000) hydrocarbons. Physicochemical and operational properties of fuels are not only due to the properties of particular hydrocarbons, but the relations of their concentrations and mutual impacts. It was observed that when small amount of new hydrocarbons is added to conventional fuel (10–50% (V/V)), it significantly disturbs these relations and consequently leads to the change of many operational properties of composed fuel. It was found that the chemical composition of synthetic hydrocarbon components has a significant influence on the properties of fuels, determining the failure-free operation of engine supply systems. Chemical composition and the properties of synthetic components of fuels depend on manufacturing technology. Feedstock from which these components are produced affects chemical composition and consequently also properties of final synthetic blends.

Currently, fuels containing synthetic hydrocarbons are approved for application in turbine aircraft engines. ASTM D7566 [6] allows to add up to 50% synthetic components to conventional fuel to supply turbine engines. The procedure and certification path of new fuels for aviation are described in [7].

The results from tests of different technologies of biofuels and their application in turbine engines, especially to be used in the aviation sector as well as maritime industry, can be found in many publications [8-13].

One of the essential aspects in the context of operation of turbine engines is thermal degradation of fuels. It occurs in the hot section of the fuel supply system and in combustion chamber. Deposits formed in fuel pipelines, injectors and combustion chamber [14] affect stability and kinetics of combustion process, which may cause failures in the engine's hot section, especially the combustor region. To sum up, the forming of deposits negatively affects operation process of turbine engines and thermal conductivity in aircraft systems.

Current large engines maximize the available heat sink capacity of the fuel system. Increased thermal loads, in connection with currently used fuel, cause different operational problems and reduce life and reliability. In order to absorb additional heat produced by future VHBR engine, acceptable temperatures of fuel system have to be increased and future fuel must be thermally optimized to increase its heat sink potential.

The reason of thermal stability importance is that actually used gas turbine engines as well as those planned for production in the future use jet fuel as their primary heat sink. Commercial jet aircraft uses fuel as a heat sink for cooling avionic and hydraulic systems. After absorbing heat, only a fraction of the fuel is burnt, and the rest is recycled back into the fuel tanks to undergo the heat exchange processes. As jet fuel during aircraft operation is subjected to high heat loads (very low temperatures in wing fuel tanks and fuel pipelines, high temperatures in heat exchangers, very high temperatures in fuel nozzles), it undergoes thermal stress and degrades. Actually more and more aircraft systems require intensive cooling, so the need to increase jet fuel "thermal capacity" is a critical factor.

The objective of the studies was to analyze the effects of changes of a hydrocarbon composition of fuel for turbine engines (by adding bio-components) on the mechanism and intensity of thermal degradation of these fuels.

FUEL SAMPLES

For the tests blends of jet fuel and synthetic hydrocarbons approved to be used in aviation turbine engines were used. To prepare those blends, necessary components were selected in compliance with the ASTM D7566. The major component of each blend was a commercial jet fuel (kerosene type Jet A-1) from Merox technology. The other components were synthetic hydrocarbons obtained from hydro-processed esters and fatty acids (HEFA) and alcohol-to-jet (ATJ) technologies. The quantitative selection of components for the preparation of blends involved the usage of maximum allowable contents of synthetic hydrocarbons in jet fuel. The composition of tested fuels is presented in Tab. 1.

Tab.1. Composition of tested fuels

Fuel	Composition [%(V/V)]		
	Jet A-1	HEFA	ATJ
Jet	100	–	–
Jet/HEFA	50	50	–
Jet/ATJ	50	–	50

TEST RIG AND PROCEDURE

To conduct a simulated thermal degradation process of fuels, a specially built test rig was used, which was worked-out as a part of the research project No. DEC-2011/01/D/ST8/06567 funded by the National Science Centre. The project of the non-standard test rig was developed to be a tool for studying thermal degradation of fuels, due to the emerging problem of correlation between the results collected by using the ASTM D3241 method dedicated for this purpose [15], and direct observation in operation.

On this test rig, oxidation and thermal degradation processes occur under certain conditions during the flow of fuel through the system. The fuel sample is re-pumped through subsequent component parts of the set, moving through its fundamental part, a sand bath with precisely controlled working temperature. The intensity of destruction processes depends on the value of the selected temperature in the sand bath and the duration of its influence on fuel. Exposure time (presence of the portion of fuel in hot section) is controlled by changing the fuel flow speed.

The general view of the test rig is shown in Fig. 1, and its more detailed description is provided in [16].

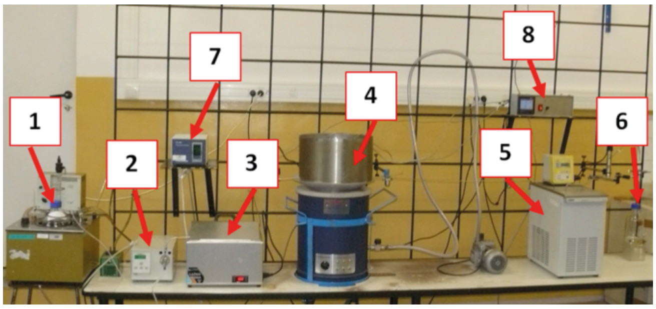

Fig. 1. General view of the test rig: 1. vessel with the tested sample, 2. fuel pump, 3. water bath, 4. sand bath, 5. cooling thermostat, 6. vessel for the tested sample, 7. temperature controller in the sand bath, 8. recorder

To conduct the thermal degradation process of the samples of fuels the following conditions were established:

1. Constant pressure value in test rig : 3.5 MPa – such pressure value occurs in fuel systems of modern aircraft.
2. Temperatures of the sand bath: 300°C and 500°C – the values reflect the thermal degradation of fuel (Fig. 2).
3. Constant value of the fuel flow rate: 3.0 ml/min – it ensures the identical thermal impact on the fuel under investigation.

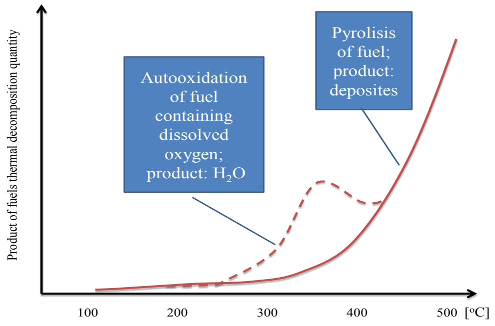

Fig. 2. Thermal degradation process of aviation fuels

As the first step, all fuel samples were prepared by filtering them through 0.8 μm membrane filters to remove contaminants. Then they were thermally stressed on the test rig (Fig. 1), and after that selected properties were checked, according to Tab. 2.

Tab. 2. Selected properties and test methods

Property	Test method
Solid contaminants	ASTM D 5452
Water content	PN-EN ISO 12937
Acid number	ASTM D 3242

The principal sign of thermal degradation of fuel is the formation of solid contaminants (products from the degradation process) and the increase of acid number. As a result, increased water content (a product of oxidation reaction derived from the dehydrogenation of hydrocarbons) can be observed. The analysis of the above mentioned parameters allows to evaluate changes in the fuel, resulting from the thermal stressing.

The IR (infrared) spectrometry was applied to monitor changes of chemical composition caused by fuel thermal stressing. For this purpose FT-IR Nicolet iS10 spectrometer was used. IR spectra were obtained with transmission method by using a variable path-length cell (VPL). The range of the spectrum was 4000-400 cm^{-1}. The spectral research was conducted with samples before and after the thermal stressing process in given experimental conditions.

RESULTS AND DISCUSSION

The influence of thermal stressing conditions on selected properties of investigated fuels is presented in Tab. 3.

Tab. 3. Conditions for bench tests and results of fuel tests

Property	Fuel								
	Jet			Jet/HEFA			Jet/ATJ		
	not stressed	thermally stressed		not stressed	thermally stressed		not stressed	thermally stressed	
Conditions during bench tests									
Test temperature, [°C]	–	300	500	–	300	500	–	300	500
Flow speed, [ml/min]	–	3.0	3.0	–	3.0	3.0	–	3.0	3.0
Pressure, [MPa]	–	3.5	3.5	–	3.5	3.5	–	3.5	3.5
Selected test results									
Solid contaminants, [mg/l]	0.30	5.05	6.98	0.40	3.82	4.39	1.06	5.36	7.48
Water content, [mg/kg]	71.3	126.1	162.1	66.3	95.8	160.5	68.6	88.6	149.0
Acid number, [mg KOH/g]	0.002	0.007	0.020	0.001	0.007	0.009	0.002	0.008	0.018

The analysis of the obtained results clearly shows that there are differences in the thermal degradation process between conventional jet fuel (Jet) and semi-synthetic fuels (Jet/HEFA and Jet/ATJ). For all tested fuels thermal impact causes the same direction of changes in selected properties, i.e. with the increase of the test temperature, there is an increase of the content of solid contaminants, water and acid number. However, the intensity of the changes is different for neat kerosene-based fuel and semi-synthetic fuels. Besides, by analyzing the results only for semi-synthetic fuels, the discrepancies are visible in the course of thermal degradation process depending on the selected technology for manufacturing a synthetic component (different results for different blend).

The above observation allows to formulate a conclusion that though all investigated fuels are approved for use in turbine engines, depending on the technology of bio-component production, thermal impact leads to the degradation of fuel with different intensity. Thermal degradation process is dependent on the chemical composition and properties of a given fuel. This process may be different and have various effects in the current operation of turbine engines depending on the type of applied fuel.

The studies with IR spectroscopy method started with making spectra of neat components, i.e. Jet, HEFA and ATJ. Among the mixture of various hydrocarbons found in Jet, the infrared analysis showed n-paraffins and aromates, HEFA was defined as n-paraffins and ATJ as iso-paraffins. Next, spectra of the tested fuel blends were recorded before and after passing through the test stand. Analyzing the obtained spectra, three bands were identified, where changes indicating the progressing thermal degradation process were visible. Band 1641 cm^{-1} is characteristic for stretching vibrations C=C in alkenes, band 1607 cm^{-1} appears as a result of skeletal vibrations causing the extension of bond C=C inside the ring in aromatic hydrocarbons, and band 909 cm^{-1} originates from wagging vibrations =C–H in alkenes with the double bond at the end of the molecule. For the mentioned bands, qualitative changes were depicted in the form of spectra and quantitative changes, as surface areas below the peak before and after the studies. The surface area was calculated with the integration method, limited by a baseline and absorption curve.

The emergence of bands typical for alkenes indicates that as a result of thermal stressing, hydrocarbon chains are broken, and double bonds are formed. Besides, the accumulated results confirm that in all cases there is an increase of intensity for a band with maximum by 1607 cm^{-1} derived from aromates. For bands derived from alkenes, significant and observable changes in IR spectra occur only at 500°C, where the reactions of thermal decomposition (pyrolysis) took place, but the changes detected for the band derived from aromates, developed equally intensively already in the first stage (Fig. 3 and Fig. 4). The observed changes are not significant, but they show the trend of destruction processes, the intensity of which is significantly dependent on the added components.

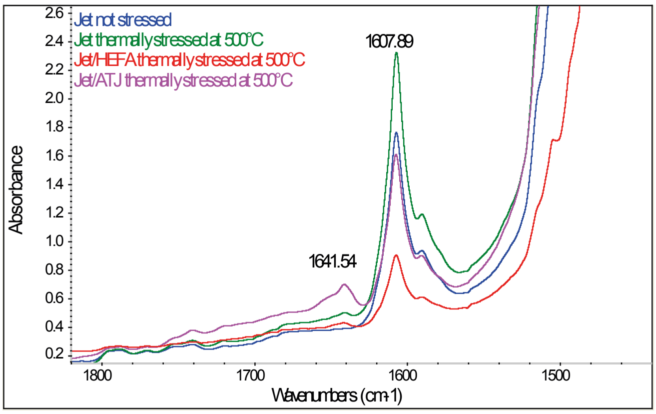

Fig. 3. IR spectra in the range 1800-1450 cm^{-1} for the tested mixtures after the thermal degradation process at 500 ° C and the IR Jet A-1 spectrum before the test

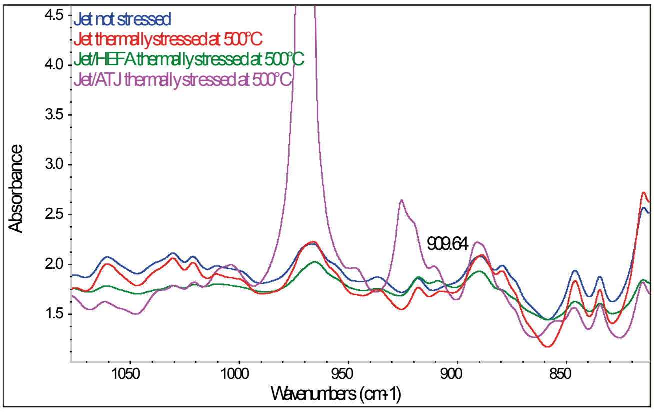

Fig. 4. IR spectra in the range 1100- 800 cm^{-1} for the tested mixtures after the thermal degradation process at 500 ° C and the IR Jet A-1 spectrum before the test

The research outcomes of IR spectra prove that engine fuels are susceptible to degradation to a varying degree, depending on their chemical composition. It is due to the general mechanism of chemical reactions that contribute to the formation of solid contaminants [17, 18, 19, 20, 21].

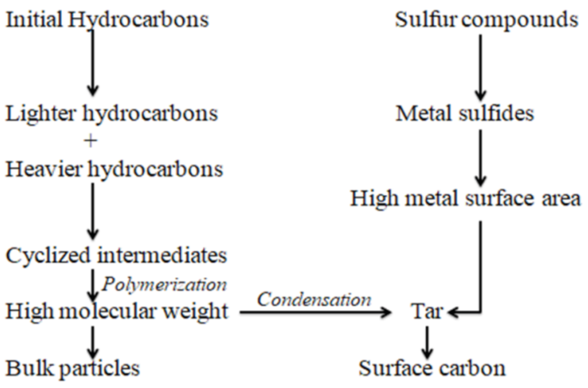

Fig. 5. Diagram of thermal degradation reaction chains, acc.[17]

In high temperature (>350˚C), two processes are dominant: distribution of hydrocarbons which are broken down to carbon and hydrogen or polymerization/condensation of hydrocarbons which are broken down to poly-nuclear aromatic hydrocarbons, from which carbonated deposits are created.

The first of the processes is catalyzed by the surfaces of metals, with which the fuel comes in contact within the hot section of the supply system. The second mechanism, polymerization, is usually a non-catalytic reaction.

The analysis of results compiled in Tab. 4 enables to observe that the changes occurred with different speed depending on the content of the fuel, what was also discovered based on physicochemical studies. The band characteristic for stretching vibrations C=C in alkenes (1641 cm^{-1}) for Jet/ATJ is noticed as post-production impurity already prior to subjecting it to the degradation process. The surface area of this peak for Jet/ATJ after the process in 500°C is ten times bigger comparing to Jet. From the analysis of the spectra, it can be stated that the most stable sample was Jet. For its strongest resistance to thermal degradation are responsible hydrocarbon chains that are n-paraffins and the increased amount of aromates, when compared to the other two blends. Jet/ATJ blend turned out to be the least stable due to presence of iso-paraffins derived from synthetic component.

Tab. 4. Surface area under the peak for bands, where changes were noticed after the thermal degradation processes, for tested fuels.

Fuel	Surface area under the peak		
	1641 cm^{-1} band	1607 cm^{-1} band	909 cm^{-1} band
Jet not stressed	0	24.67	0
Jet thermally stressed at 300 °C	0	28.46	0
Jet thermally stressed at 500 °C	0.26	33.07	0.19
Jet/HEFA not stressed	0	7.20	0
Jet /HEFA thermally stressed at 300 °C	0	11.33	0
Jet/HEFA thermally stressed at 500 °C	0.33	8.40	0.21
Jet/ATJ not stressed	0.16	15.75	0
Jet /ATJ thermally stressed at 300 °C	0.17	15.90	0
Jet /ATJ thermally stressed at 500 °C	2.62	17.79	0.60

On the basis of the research results presented in Tab. 4 it can be stated that:

- water is the oxidation product of hydrogen removed during the formation of olefins from saturated hydrocarbons (n-paraffins and iso-paraffins); these reactions occur at temperature above 300°C and belong to reactions catalyzed by conduits' surfaces, through which the fuel is pumped;
- solid contaminants are products of non-catalytic reactions of condensation of hydrocarbons broken down to poly-nuclear aromatic hydrocarbons.

For all examined samples the dependence was observed between the surface area of the peak 1607 cm^{-1} and the content of solid contaminants. It enables to link the increase of the number of aromatic rings in fuels after thermal degradation with the amount of solid contaminants formed during degradation. This regularity was observed both for the process conducted at the temperature of 300°C as well as at 500°C. It allowed to state that for all examined fuels the mechanism of reaction of thermal degradation is the same at the temperature of 300°C and 500°C. As per the results of the measurement of the amount of deposits, the value of the activation energy of reaction leading to the formation of solid contaminants can be calculated.

Assuming that reactions of thermal destruction of fuel are first-order reactions relative to the final product, the kinetic equation of the thermal destruction process of fuel takes the following form:

$$V_{product} = d\ c_{product}/dt \tag{1}$$

$$V_{product} = k\ c_{product} \tag{2}$$

where: $V_{product}$ – reaction rate, $c_{product}$ – concentration of reactions product, k – reaction rate constant, t – time.

After the integration between c_0 and c_t, and assuming that $t_0 = 0$ and $t = V/v_w$, where:
V – volume of the pumped fuel,
v_w – speed of fuel flow,
$c_{product}$ – concentration of the product.

$$k = [(\ln c_{product}) - (\ln c_{product0})]v_w/V \tag{3}$$

Applying relation (3) and introducing $c_{product}$ as the acid value, the amount of deposits and the water content, the values of the rate constant of thermal degradation reaction was calculated for the investigated fuels.

These values were calculated for both temperatures under which the thermal degradation process was carried out: 500°C and 300°C. Using the Arrhenius equation:

$$k = A\exp(-E_a/RT) \tag{4}$$

where: E_a - activation energy, R – gas constant, T – temperature.

And the values of the rate constant of reactions for the reaction of thermal degradation of fuel at temperature of 500°C and 300°C, the E_a values were calculated for each of the examined fuels, as follows:

$$\ln k = \ln A\text{-}Ea/RT \tag{5}$$

$$\ln k_{500} - \ln k_{300} = E_a/R(-1/773+1/573) \tag{6}$$

$$E_a = R(\ln k_{500} - \ln k_{300})/(-1/773+1/573) \tag{7}$$

The results were shown in Tab. 5. It is worth to highlight that the obtained absolute values of activation energy shall not be analyzed: the content of solid contaminants is not expressed in units of concentration. Thus, the results illustrated in Tab.5 are indicative, enabling to compare the mechanism of thermal degradation of fuel containing different synthetic components.

Tab. 5. Activation energy of thermal degradation reactions for tested fuels based on solid contaminants

Fuel	Activation energy [kJ/mol]
Jet	1.7
Jet/HEFA	1.3
Jet/ATJ	4.1

The presented results indicate that the introduction of synthetic hydrocarbon in the amount of 50 % (V/V) into conventional jet fuel results in the change of mechanism of reactions constituting a thermal degradation. The hydrocarbons obtained by HEFA technology do not substantially alter the mechanism of thermal degradation reaction when compared to the thermal degradation of Jet. ATJ hydrocarbons, however, significantly change the thermal degradation mechanism.

CONCLUSIONS

This work considered the intensity of the thermal degradation process of fuels used in turbine engines, containing synthetic components from HEFA and ATJ technology. The research results are summarized as follows:

1. For all tested fuels the thermal impact causes the same direction of changes of selected properties, associated with the products of the thermal degradation process. With the increase of the applied temperature, there is an increment of the content of solid contaminants, water and acid value. Nonetheless, the intensity of these changes is different for neat kerosene-based fuel and semi-synthetic fuels. There are also differences in the course of the thermal degradation depending on the selected technology of manufacturing a synthetic component.
2. Analysis with IR spectroscopy method confirmed the results for selected properties. The formation of bands characteristic for alkenes reflects the amount of formed water, and bands for aromates correspond to the amount of solid contaminants.
3. Calculated activation energy of reactions as a result of which solid contaminants are formed indicates the change of its mechanism due to the change of hydrocarbon composition of fuel.
4. Thermal degradation process is contingent upon the chemical composition and properties of a given fuel. It can proceed differently and exert various effects in the current operation of turbine engines depending on the used fuel.

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