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95-GT-49

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MECHANISMS OF COKE FORMATION IN GAS TURBINE COMBUSTION CHAMBERS

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ABSTRACT

New gas turbine combustor designs are developed to reduce pollutant and NO_x -emissions. In these new combustors, the formation of carbonaceous deposits, especially in prevaporizers, affects the reliability and effectiveness of operation. To avoid deposits, a detailed knowledge of the origins and mechanisms of formation is required.

To obtain a deeper insight, the phenomena were studied systematically. The deposits under consideration show differing characteristics suggesting more than one formation mechanism in the combustor. Consequently, the primary goal was to identify the formation mechanisms and, subsequently, to simulate the mechanisms under well-defined conditions in bench tests for determining the relevant parameters of deposit build-up. The mechanisms of formation were identified based on the properties of the deposits in the combustion chamber. In order to characterize the deposits, physical and chemical analysis techniques were utilized.

In summary, tests and numerical predictions identified two major paths of formation: a deposit build-up resulting from flame products such as soot or coked droplets and a deposit build-up resulting from liquid fuel impinging the wall accompanied with chemical reactions at the wall.

The deposits caused by fuel droplet impingement were intensively studied in bench tests. In analyzing the processes, the influence of wall temperature, fuel composition, and the oxygen content in the environment is shown in detail. In addition, the importance of thermal instabilities of the fuel, previously studied under fuel supply system conditions, is demonstrated for a deposit formation inside a combustion chamber.

INTRODUCTION

The development of new gas turbines is focused on improvements in thermal efficiency and pollutant emissions. Higher thermal efficiencies are primarily attained by increasing temperatures and pressures. Both parameters favor pollutant formation during the combustion process, if no special measures are taken. Therefore, primarily for combustors burning liquid fuels, new design concepts and fuel preparation systems have to be developed to meet future laws and regulations for pollutant emissions.

Prevaporizing the liquid fuel and mixing it with primary air before entering the reaction zone of the combustor is a new approach to reduce pollutants. However, in addition to self-ignition of the vaporized fuel and flashback, the formation of carbonaceous deposits affects the reliability and effectiveness of the prevaporizers. Carbonaceous deposits at the wall, for example, reduce the vaporization rates drastically and affect the radiative heat transfer in the combustor. Also, because deposits affect heat transfer from the hot gas flow to the wall, they cause complex wall temperature distributions, thus, leading to thermal stresses and damage to the combustor. To avoid deposit formation, detailed information on the mechanisms and the major influencing parameters is imperative. In contrast to former studies (Starkman et al. (1951), Wear and Hibbard (1955)) in gas turbine combustors which determined different deposits without investigating the formation mechanisms, our new approach considers two different paths of formation: deposits resulting from liquid fuel impinging and reacting at the wall's surface, and from flame products such as soot or coked droplets which agglomerate at the wall. First of all, criteria to distinguish between

Presented at the International Gas Turbine and Aeroengine Congress & Exposition
Houston, Texas - June 5-8, 1995

This paper has been accepted for publication in the Transactions of the ASME
Discussion of it will be accepted at ASME Headquarters until September 30, 1995

both formation mechanisms were derived based on the deposit properties to determine the formation path. After determining the formation mechanisms bench tests were performed for identifying the parameters influencing deposit formation.

Consequently, the following approach had to be taken in the present study: based on observations under real engine conditions accompanied with detailed chemical analyses of the deposits, bench tests under well-defined conditions were required for an understanding of the chemical mechanisms. The methodology of the investigations including the chemical and physical diagnostics as well as the test facilities and the results will be described below.

METHODOLOGY AND ANALYSIS TECHNIQUES

As the experiments under real combustor conditions revealed various deposits suggesting different mechanisms of formation, the primary goal in studying combustor deposits is the definition of the deposit formation path. This can be reached by a systematic analysis of the deposit properties as well as the numerical simulation of the flow field in the combustor under consideration.

For a complete description of the deposits, various coordinated analysis techniques were applied. To get a first indication on the chemical composition, the specimen were tested by elemental analysis. Using this technique, a C-H-N-O determination can be obtained. For chosen experiments a determination of the sulfur content was done, additionally. In particular, the carbon content provides decisive information about the formation mechanism. Furthermore, based on the results of elemental analysis further analysis techniques can be chosen effectively. Infrared spectroscopy was applied to identify chemical functionalities in the deposits to get a deeper insight in chemical reactions. Gas chromatography was performed to investigate the fuels as well as the solid deposits, which were dissolved in advance. Although not all components in the deposits can be dissolved, gas chromatography gives important information about chemical combinations included in the deposits.

To detect whether graphite within the deposits has formed, diffraction patterns of the samples were performed. Furthermore, if graphite does exist, a preferred orientation of the basal planes can be determined using this technique. These patterns distinguish whether deposit formation at the combustor wall is caused by droplet impingement or by intermediate flame particles. Additionally, scanning electron photographs were taken from the samples to characterize the surface morphology of the deposits. Regarding the morphology, a first indication on the path of deposit formation can be obtained (Schirmer (1970)). In applying the different analysis techniques, a systematic and comprehensive characterization of the physical and chemical properties of

the deposits were achieved. Considering the results of all analysis techniques, the deposit formation can be deduced and the major question whether the chemical reactions take place at the wall or in the flow field can be answered. The suggested mechanisms can then be confirmed by numerical studies of the flow field in the combustor and a calculation of the fuel preparation.

In the second step, influencing parameters are derived and quantified through deposit formation studies under well-defined conditions. A quantification of the influencing parameters can only be reached by systematic bench tests as under real engine conditions various parameters are interacting and varying, e. g. wall temperatures oscillate with the thermal load and in the course of starting and stopping.

In laboratory experiments, the formation of deposits caused by droplets impinging on the hot combustor wall is simulated under well-defined conditions. In the investigations, the main parameters influencing deposit formation, such as wall temperature, fuel composition and oxygen content in the environment are tested. By comparing chemical properties of bench test deposits with those derived from a combustion chamber it can be determined whether droplet impingement on the combustor wall is one of the relevant mechanisms leading to deposit formation under gas turbine combustor conditions. In case of identical chemical deposit properties, the same chemical reactions will take place under engine conditions and in laboratory experiments.

In demonstrating that identical mechanisms take place in bench tests and combustors, the results of the present study are of far-reaching consequence for deposit formation in real engines since the results of the bench tests can be applied to real engine conditions. Another advantage of the chosen concept is the fact that the reasons of deposit formation can be determined independently from combustor design and solely on the deposit properties. For example, in considering the elemental analysis, the path of deposit formation under gas turbine combustor conditions can be determined.

EXPERIMENTAL SETUP

For the various laboratory investigations, a bench test rig, shown in Figure 1, was developed. The main objective of the test facility was the simulation of the combustor wall conditions which include the effects of surface temperature, surface material and ambient atmosphere. Thus, deposit formation by droplet impingement on the hot combustor wall was investigated. The monodispersed fuel droplets of 0.022 ml volume, generated by a burette, hit the surface of the test specimen. The test specimen with a diameter of 50 mm was heated by an electrical heater element. Maximum surface temperatures up to 1100 K can be achieved for the investigations such that the whole temperature regime for deposit formation was possible.

The temperature was monitored simultaneously by

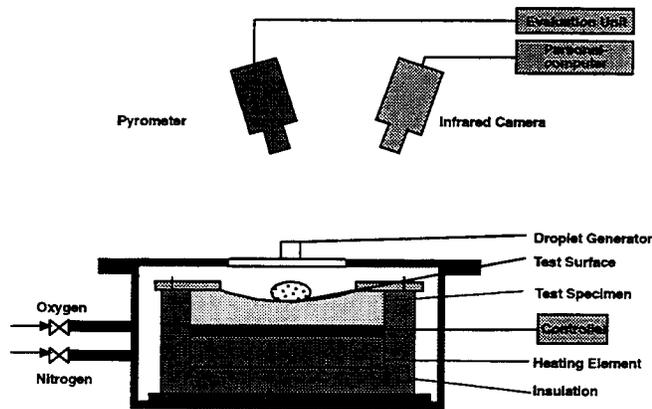


FIG. 1: Schematic of the Bench Test Rig

thermocouples and an infrared system. Surface temperature measurements were performed by pyrometry in the vicinity of deposit build-up which was varnished with a coating exhibiting well-defined emission characteristics. As infrared camera measurements revealed a homogeneous temperature distribution on the specimen surface, temperature measurement by pyrometry beside the deposit formation area provide exact values of the surface temperature at the location of deposit build-up.

Furthermore, the test specimen and the location of deposit build-up are enclosed in order to vary the content of oxygen and nitrogen in the test rig. Due to the design of the test rig, near wall conditions in a combustor including fuel, wall temperature, wall material and gas atmosphere can be adjusted constantly.

After hitting the heated test specimen, the fuel droplets evaporate and chemical reactions are initiated which lead to the build-up of solid carbonaceous deposits. The evaluation of the test is done gravimetrically by weighing the test specimen before and after the test, respectively. The sensitivity of the gravimetric weighing system is 0.1 mg, so that exact values of deposit mass can be given. By the weighing only solid deposits and no gums or deposit precursors are considered. The design of the present bench test rig provides a quantitative relation between the mass of the deposit and the mass of the fuel, which is of fundamental importance for identifying major influences. As the characteristic quantity, a dimensionless deposit mass is introduced, defined as:

$$\omega = m_{dep}/m_{fuel} \cdot 100 [\%]$$

where ω denotes the mass ratio of the deposit to the initial test fuel. This dimensionless quantity is established in order to compare tests with different initial fuel masses. The introduction of this dimensionless number is possible as preliminary tests revealed a linear dependence between deposit mass and fuel mass under constant test conditions. Considering deposit mass in the

quantification of the major influences is an advantage of the test rig, because other bench test rigs (see Maier et al. (1985)) allow only visual evaluation of the deposits. Furthermore a chemical evaluation by the above described techniques is done to get information about reactions leading to deposit build-up.

The combustion chamber tests were performed in the film-evaporation research combustor at the Institute for Thermal Turbomachinery. A sketch of the combustor, which is described in detail by Pfeiffer et al. (1991a) (1991b), is given in Figure 2. The combustor was designed for a thermal load of about 150 kW at a pressure pf 4.5 bar with a overall air/fuel ratio of 5.6. Further information on constructional details is given in the above quoted papers.

The combustor was developed to find new design concepts in reducing NO_x -emissions by fuel vaporization and staged combustion. In the following discussion, only a few comments on fuel preparation in the combustor will be made because it affects briefly the deposit formation.

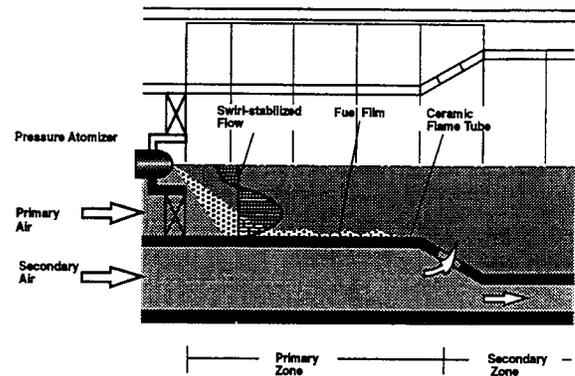


FIG. 2: Schematic of the Film Evaporation Combustor

The fuel (Fuel Oil) is sprayed by a pressure atomizer towards the inner surface of the flame tube. The droplets impinge on the wall and form a liquid film, which is shear driven by the primary air flow. While spreading, the film evaporates through heat transferred from both the wall and the hot gas flow. The primary air enters the combustion chamber by an axial flow swirler, thus forming a recirculation flow regime for flame stabilization. The fuel vapor mixes with the primary air forming a homogeneous fuel/air mixture in the primary zone.

DEPOSIT FORMATION IN THE COMBUSTION CHAMBER

In the research combustion chamber, deposit formation occurred at two locations. Under certain test conditions, deposit build-up was found not only in the flame tube in the region of the fuel film, but also in the vicinity of the fuel nozzle. A sketch of deposit location is

given in Figure 3.

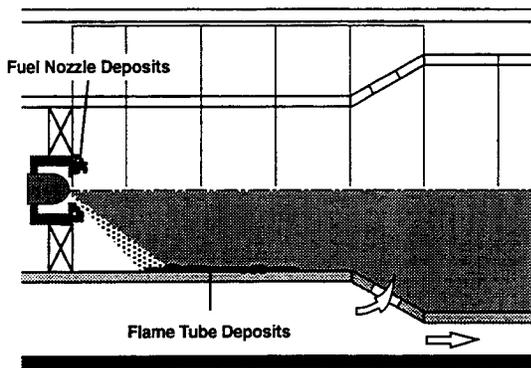


FIG. 3: Sketch of Deposit Location

These deposits were characterized by several analysis techniques to determine their formation mechanisms. To get a first impression of the surface morphology, scanning microscope photographs of both deposits in the flame tube as well as in the vicinity of the fuel nozzle were performed.

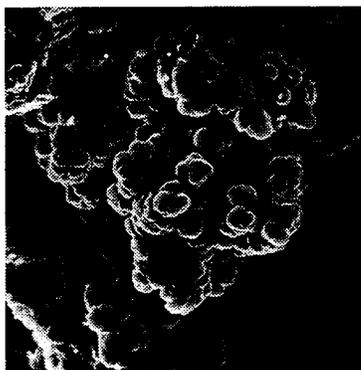


FIG. 4: Scan Microscope Photograph of the Deposits in the Vicinity of the Nozzle ($1\text{cm} \equiv 20\mu\text{m}$)

Figure 4 shows the deposits near the fuel nozzle. An accumulation of spherical particles, which were initiated in the hot flow field and then agglomerated at the wall surface, became apparent. The diameter of these spherical particles was approximately 4-5 microns. The particles are considerably larger than soot particles, as Müller and Wittig (1991) showed in detailed studies. In contrast, the deposits in the flame tube (see Figure 5) exhibited an amorphous structure without any particles falling out from the hot gas flow. The differences in the deposits due to their location in the combustor detected by scan microscope were confirmed by their chemical composition.

While the deposits in the vicinity of the nozzle contained about 98 mass percent carbon and 1-2 % oxy-

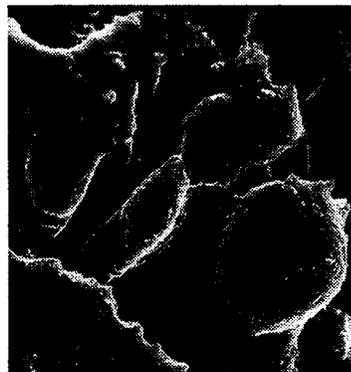


FIG. 5: Scan Microscope Photograph of the Deposits in the Flame Tube ($1\text{cm} \equiv 20\mu\text{m}$)

gen, the deposits in the flame tube consisted of about 74-80 % carbon, 20 % oxygen, 4 % hydrogen and small amounts of nitrogen. Based on the results of the elemental analysis, further analysis techniques were done. Thus, from the deposits in the flame tube, infrared spectra were taken in order to obtain more information about functional groups containing hetero-compounds, especially oxygen. The deposits in the vicinity of the fuel nozzle, which consisted almost entirely of carbon, were described by diffraction patterns to identify whether graphitization has occurred in the material. The pattern shown in Figure 6 demonstrates the presence of some random oriented crystallites in the deposits. Thus, these particles have been exposed to extremely high temperatures in the reaction zone of the combustor.

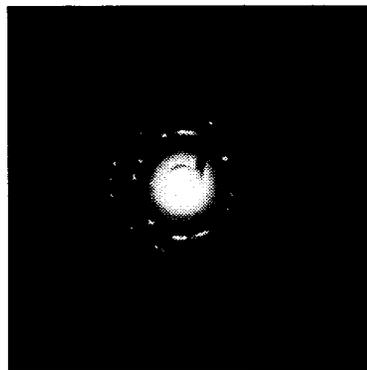


FIG. 6: Diffraction Pattern of the Deposits in the Vicinity of the Fuel Nozzle

For the initiation of graphitization processes, temperatures beyond 1200 K were required (Heidenreich et al. (1968)). However, these temperatures were not reached at the combustor wall where the deposits were found as shown by thermal paints. This hypothesis is supported

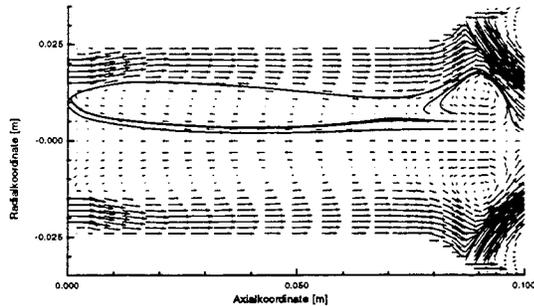


FIG. 7: Computed Flow Field in the Primary Zone of the Combustor

by the fact that no preferred orientation of the intensity in Figure 6 is present, which would indicate an orientation of the basal planes in the deposits parallel to the wall surface. Since no preferred orientation occurred, an indication is given that the deposit forming reactions and graphite formation would take place in a flow field and not at a plain surface (Grisdale (1953)). Therefore, on account on diffraction analysis, deposit formation at the fuel nozzle is primarily caused by particles formed in the hot reaction zone of the combustor.

Summarizing the analyses of the deposits in the vicinity of the fuel nozzle, deposit formation here was caused by flame products as soot or coked droplets, as examined by Urban and Dryer (1990). This conclusion can be supported by analyzing the flow field in the combustion chamber.

To show the impact of the flow field in the combustor on deposit formation, a numerical calculation was performed. The numerical simulation was done by a finite-volume code developed at the Institute for Thermal Turbomachinery (see Noll (1993)) for computing three-dimensional, turbulent combustor flows. The code uses the standard- $k-\epsilon$ -turbulence model with high-order-discretization. The case discussed in the following represents typical test conditions, where deposit build-up was found (see also Pfeiffer et al. (1991a)). In Figure 7 the flow field in the primary zone of the combustor is shown. In the vector plot the spreading of the recirculation regime within the primary zone becomes apparent. For reasons of clarity, some streamlines are plotted. The streamlines show that the recirculation flow regime spreads back to the fuel nozzle so that hot gas and intermediate flame products can fall out there. In considering the flow field, a transport of small particles, formed in the hot reaction zone of the combustor, towards the vicinity of the fuel nozzle can be determined.

Therefore, regarding both the chemical properties of the deposits as well as the flow situation in the combustor, a deposit build-up at the fuel nozzle by small particles such as soot or coked droplets can be formulated. The size of the particles corresponds with those found by Urban and Dryer (1991), who give a correlation between the initial droplet diameter and the diameter of

the coked droplet. Because these authors considered solely heavier fuels, further studies are necessary to validate the assumption that these spherical particles are coked fuel droplets.

The deposits within the flame tube differ distinctly from those in the vicinity of the nozzle. In particular, the high oxygen content, which is identified by elemental analysis as well as by infrared analysis, exhibits similarities with deposits formed through thermal instabilities of hydrocarbons. These mechanisms were studied by Chin and Lefebvre (1992) and Krazinski et al. (1990) under fuel supply system conditions. Both authors investigated deposit formation by heating fuels in tubes outside a combustor. To illustrate that these mechanisms are also relevant inside the combustor for fuel impinging the combustor walls, the present combustor results were further studied in detail. To verify the hypothesis, the deposits of laboratory experiments are contrasted to those in the film section of the combustor and those found in investigating thermal instabilities of fuels.

DEPOSIT FORMATION IN BENCH TESTS

In the bench test rig, a deposit formation caused by droplet impingement on the hot combustor wall was simulated. All the tests described below were performed at a pressure of 1 bar. To compare the deposits in the bench tests and those in the flame tube of the previously described combustor, fuel oil was initially utilized. As Figure 8 shows, both the elemental analyses and the infrared spectra of deposits in the bench test as well as in the film section of the combustor under consideration show good agreement.

In both cases, the deposits consist of approximately 80 % carbon and 20 % oxygen. The oxygen is chemically fixed in -OH, C=O and C-O bonds. Based on the correlation of deposit composition in both test rigs, it can be assumed that identical mechanisms of formation take place and the dependencies found for deposit formation in the bench tests can be applied to combustor conditions. Contrasting the deposit composition to deposits observed in fuel supply systems, it is apparent that the mechanism stemming from thermal instability of the fuel also takes place inside a combustor when liquid fuel impinges the hot wall. The composition of the deposits resulting from thermal instabilities (Hazlett and Hall (1981)) and those occurring in internal engines, such as found by Ebert et al. (1981), corresponds with those detected in the bench test rig as well as in the flame tube of the combustor.

In our first laboratory experiments, the influences of the wall temperature and the fuel composition were investigated for several gas turbine fuels as well as pure hydrocarbons. Before testing the gas turbine fuels, a detailed analysis using gas chromatography and boiling point curves was done to characterize the fuels. As Figure 9 shows, for fuel oil deposit formation takes place

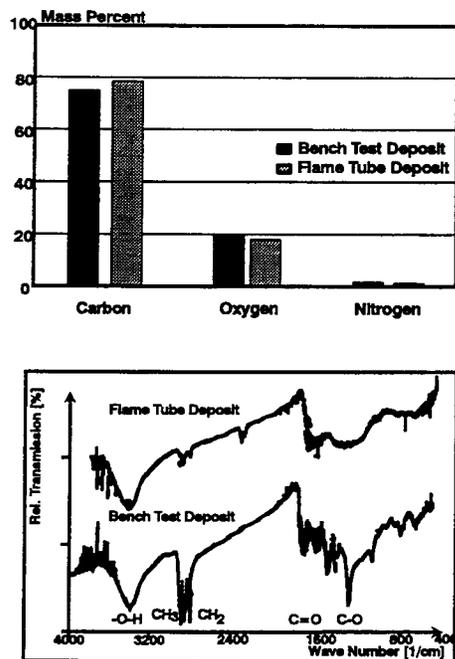


FIG. 8: Chemical Characteristics of the Deposits in the Flame Tube and in the Bench Test Rig

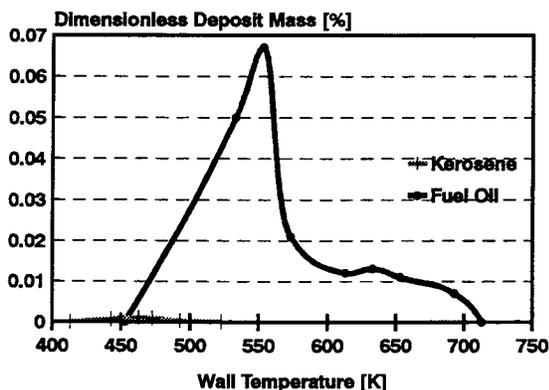


FIG. 9: Deposit Formation of Gas Turbine Fuels

in a temperature regime between 450 K and 730 K.

Using kerosine (Jet 1A) as a test fuel, deposit formation occurs at lower temperatures. This is in correspondence with the boiling characteristics of the fuels because the measured boiling range (DIN 51751) of kerosine lies between 413 K and 518 K while fuel oil boils between 450 K and 650 K. Contrasting the deposit mass of fuel oil and the kerosine under consideration, it is apparent that the high-boiling fuel oil contributes stronger to deposit formation than the kerosine.

To demonstrate that the high-boiling components in the fuel are responsible for deposit formation, the fuel oil was split into two fractions. The components boiling above 613 K were split from the compounds boiling

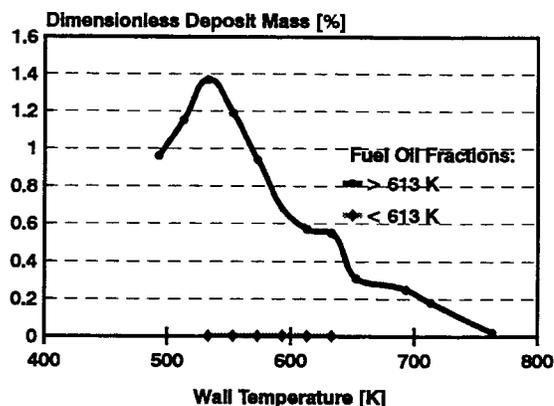


FIG. 10: Deposit Formation of Fractions of Fuel Oil

below 613 K. While the high-boiling components contribute very strongly to deposit formation, the fraction boiling below 613 K formed no deposits (see Figure 10).

From gas chromatography measurements of the high-boiling fuel fraction, no combinations with less than 19 carbon atoms were identified. The tendency of the long-chain components in the fuel to form deposits was confirmed by studies with pure hydrocarbons. In these studies considering alkanes, alkenes and aromatics it was found that with increasing carbon chain length of the hydrocarbon deposit formation increases (see Brandauer et al. (1993)). As a further result of the investigations with pure hydrocarbons, a first approach to describe the reactions leading to deposit formation can be given. In particular, autooxidation of the fuel plays an important role.

To confirm our reaction path, which indicate a strong influence of oxygen on deposit formation, our studies in an air environment were compared to those in a nitrogen atmosphere. As Figure 11 indicates, the oxygen in the ambient atmosphere contributes to a high deposit forming tendency. This is confirmed by studies of Hurley (1982), who was unable to find any deposits from liquid fuels under nitrogen tests when he used fuel without oxygen.

In the presence of oxygen, the deposit formation based on fuel oil takes place in a temperature regime between 450 K and 730 K showing maximum deposit mass at approximately 570 K. At 450 K, the onset of fast autooxidation in the fuel takes place. At temperatures above 570 K both the rate of autooxidation decreases and the droplet vaporization time decreases as shown by our droplet investigations. Both effects lead to a decrease in deposit formation. The time the liquid fuel wets the hot wall affects strongly deposit formation. To determine unequivocally this fact, experiments with various solutions of hexane and hexadecene were performed. Hexane and hexadecene were chosen because hexane forms no deposits and hexadecene contributes strongly to deposit formation. The deposit mass shown in Figure 12 is thus normalized to the initial hexadecene

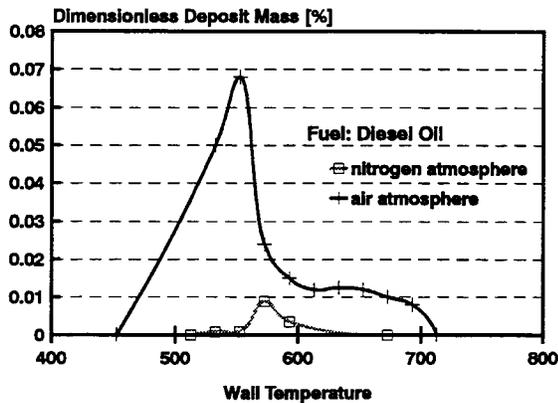


FIG. 11: Influence of Ambient Atmosphere on Deposit Formation

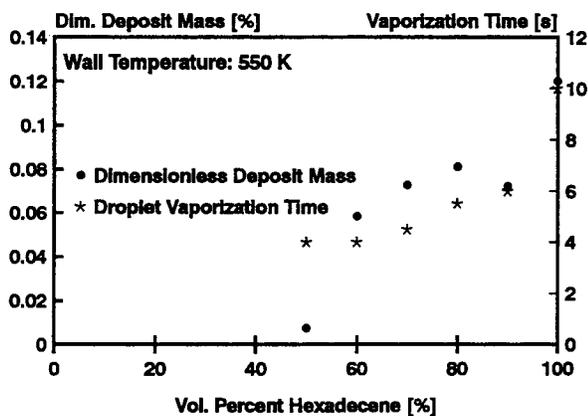


FIG. 12: Influence of Vaporization Time on Deposit Formation

mass. With increasing hexadecene content in the solution the dimensionless deposit mass increases. The vaporization time of the two-component droplets, which is also the time for chemical reactions in the liquid fuel, is also plotted in Figure 12. In this plot, the increase of deposit formation with increasing vaporization time becomes apparent.

The influence of vaporization time also becomes clear when comparing the deposit forming tendency of the high-boiling compounds in the fuel to the fuel oil itself. Our recorded boiling point curves show that above 613 K only 12 % by volume of the fuel oil boils. Because these combinations are responsible for deposit formation solely, about 8 times more dimensionless deposit mass will be expected in the experiments. However, these combinations cause about 20 times more dimensionless deposit mass (compare Figures 9 and 10) because they exhibit longer vaporization times at the same surface temperature than the fuel oil itself.

In summary, long-chain components in the fuel, which allow long residence times at high surface temperatures, contribute strong to deposit formation. The

temperature regime as well as the fact of increasing deposit mass with increasing residence time on the surface is in accordance with investigations of Marteney and Spadaccini (1986) in a heated tube. This confirms that identical processes are taking place in the combustor with fuel impinging on the hot wall and during thermal decomposition in a fuel supply system. The increase in deposit formation with rising oxygen concentration in the ambient atmosphere indicates the presence of reactions with oxygen in the formation path of these deposits. Therefore, autooxidation reactions take place in the fuel, as other authors postulate in investigating thermal instabilities of fuels (Krazinski et al. (1990), Roback et al. (1983), Katta et al. (1993)).

CONCLUSIONS

Deposit formation in gas turbine combustion chambers is found to occur at two locations with different formation mechanisms. In the combustor under consideration, deposit formation originates in the vicinity of the fuel nozzle by accumulation of particles formed in the reaction zone. This is demonstrated by detailed chemical and physical analyses and a numerical simulation of the flow field in the combustor. In contrast, the deposit build-up in the flame tube is caused by the impingement of fuel droplets on the hot combustor wall, accompanied by chemical reactions of the fuel at the wall. Finally, based on the physical and chemical characteristics of the deposits, criteria to distinguish the formation mechanisms inside the combustor were formulated. By simply comparing the elemental analyses, it can be detected whether deposit formation is caused by intermediate flame products or liquid fuel impinging on the combustor flame tube.

Detailed simulation of deposit formation by fuel impingement at the flame tube was accomplished in bench tests. From the apparent chemical similarities between the deposits from bench tests and the deposits in the flame tube, it becomes evident that the study of the relevant parameters for deposit formation in bench tests is permitted. After defining the influence of fuel composition and wall temperature, e. g., a temperature range of approximately 200 K to 300 K depending on fuel composition, it is shown that these deposits are affected by the oxygen concentration in the ambient atmosphere. In further studies the deposit causing combinations in a gas turbine fuel can be identified and the influence of the residence time of the liquid fuel on the hot surface can be determined. Considering the chemical composition and the existing temperature range of deposit formation, it is demonstrated that the thermal instabilities of the fuel, which were investigated in detail under fuel supply system conditions, have a decisive impact on deposit formation in advanced combustors using fuel prevaporization.

ACKNOWLEDGEMENTS

The research program has been supported by the Arbeitsgemeinschaft Hochtemperaturgasturbine, AG TURBO, initiated by the Secretary of Science and Technology of Germany (BMFT), and the Sonderforschungsbereich 167 by the Deutsche Forschungsgemeinschaft (DFG).

Thanks are given to the Director of the Engler-Bunte-Institute at the University of Karlsruhe, Prof. K. Hedden and to Dipl.-Ing. M. Hutfließ, for carrying out the chemical analyses. Further thanks go to the Laboratory of Scanning Microcopy at the University of Karlsruhe for their support.

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