

ICSO 2016

International Conference on Space Optics

Biarritz, France

18–21 October 2016

Edited by Bruno Cugny, Nikos Karafolas and Zoran Sodnik



Caveats and pitfalls of high energy UV laser operation on ground

L. Mondin

D. Wernham

F. F. Era

H. Kheyrandish

et al.



International Conference on Space Optics — ICSO 2016, edited by Bruno Cugny, Nikos Karafolas, Zoran Sodnik, Proc. of SPIE Vol. 10562, 105621U · © 2016 ESA and CNES
CCC code: 0277-786X/17/\$18 · doi: 10.1117/12.2296209

Caveats and pitfalls of high energy UV laser operation on ground

L. Mondin¹, D. Wernham¹, F.F. Era³, H. Kheyrandish⁵, A. Bolkhovitinov¹, A. Baselga Mateo¹, A. Ciapponi¹, C. Heese¹, T. Ivanov¹, M. Hippler², D. Thibault⁴, G. Van Papendrecht¹, W. Riede², Y. Butenko¹, H. Schroeder²

1 ESA ESTEC Netherlands, 2 DLR Institut für Technische Physik Germany, 3 Leonardo Company Florence Italy, 4 Airbus DS Toulouse France, 5 Aystorm Scientific Ltd UK.

Abstract—During the Aeolus laser and instrument transmitter development it was shown that atmosphere quality was one major limiting factor for high energy UV laser operation at ambient pressure. As already proven in literature operation can only be safely obtained in the presence of oxygen ([1] to [6]).

Furthermore, air quality has to be adequately controlled and monitored to ensure that no catastrophic event occurs. On-line qualitative (no/no-go discriminator) can be obtained using fluorescence monitoring of the laser beam, while GC-MS remains the technique of choice to quantitatively evaluate suitability of air supply a priori. LIC testing is in the process of being adapted to trace contaminant evaluation.

Finally, the formation of laser generated absorbing features is described, during air irradiation. The potential root causes of these absorbing dot-like features are explored, using optical measurements, microscopic inspections and chemical analysis. Current results indicate organic deposits associated with metallic particles and a presence of phosphates.

Index Terms—High Energy UV Laser, LIC/LIDT, GC-MS, trace gas analysis, absorbing features.

I. INTRODUCTION

The Aeolus mission is part of ESA's Earth Explorer program. The goal of the mission is to determine the first global wind data set in near real time. For this mission the high spectral resolution lidar instrument Aladin was developed. In order to retain an acceptable level of backscattered signal the laser shall emit 80mJ, which translates to laser fluences of the order of 1 J/cm² at 355nm over a high number of shots (5.5Gshots for the mission) for the optical elements.

One major challenge has been the development of Ultra Violet (UV) resistant optical systems, such as the laser transmitter, and the Transmit-Receive Optics. An equally challenging concern has been Laser Induced Contamination (LIC) in vacuum, and at ambient pressure, where the interaction of the outgassing species from organic materials within the different sub-systems volumes, and the UV laser beam irradiating the surface of the optics led to the formation of highly absorbing deposits. Despite taking all of the normal preventative measures, such as reducing the organic materials in the vicinity of the high fluence optics, employing high temperature vacuum bake-outs at component, sub-system, and instrument level, the LIC formation was reduced but not eliminated. It was found that low pressures of oxygen prevented the formation of these deposits, and could contribute

to cleaning pre-existing ones. This conclusion led to the development of an in-situ cleaning system keeping a low pressure oxygen environment. The high purity of the technical oxygen to be used in orbit and during storage / testing had to be checked and verified by test at component and system level.

It was observed that while LIC is a phenomenon mostly observed in vacuum, operation in technical gas (even in the presence of oxygen) can also lead to contamination. The presence of organics and metallic particles has to be monitored at all stages of integration and, even more importantly, testing when the number of cumulated shots will be significant (0.2-0.4Gshots). In particular, the potential presence of compounds which are not removed by oxidation (such as silicones) has to be identified and avoided before any safe operation of the laser system can be accomplished.

In addition to standard GC-MS (Gas-Chromatography – Mass-Spectroscopy) techniques, to detect the presence of trace contaminants so called LIC single material contamination testing has been implemented. This is an additional analysis tool for screening air supplies or isolated components for LIC formation hazard. The combination of both techniques lead high confidence in the purity of the gas sources under test, excluding LIC formation during AIT (Assembly Integration and Testing).

Finally, the formation of small black dot like features on the optics exposed to laser light is reported. This phenomenon appears surprisingly also in standard clean room air conditions (up to class 100). These small absorbing features cannot be completely correlated spatially with the laser beam high fluence areas meaning that they are also activated with relatively low fluences. Despite their having low impact on the overall transmission of affected optics (few %), they block the transmission locally in the order of several tenths of percent.

No direct evidence is present to implicate the absorbing features in damage creation. So far long term irradiation of these structures, up to a few Gshots, has not resulted in catastrophic laser damage. Nonetheless, the presence of metallic contamination at their core (Time-of-Flight Secondary Ion Mass Spectroscopy measurements after etching of the uppermost layers on the surface) does not permit to exonerate them as precursors to LID (Laser Induced Damage). Results of analysis shall be presented in the following text.

II. LASER INDUCED CONTAMINATION RISK REDUCTION AND OXYGEN

In the course of the Aladin development it was shown that for 355nm even the slightest trace of organic contaminants (as might still be present after typical curing and bakeout of organic materials such as paints and adhesives) could produce Laser Induced Contamination deposits which could then lead to damage. The creation of said deposits was rapidly evidenced under vacuum conditions [1].

As shown in [2], [3], and most comprehensively in [4] one possible cure for the LIC conundrum, in vacuum, is to inject an oxygen flow in the optical system keeping the residual pressure to at least a few tens of Pa.

LIC is not, however, an effect purely limited to hard vacuum conditions. LIC deposits can and will form at ambient pressure. The presence of O₂ is a deterrent to the LIC formation in these conditions too. As such operation with normal dry air is to be preferred to operation in nitrogen (or any other mixture not containing reactive oxidants), as evidenced in *Figure II-1*.

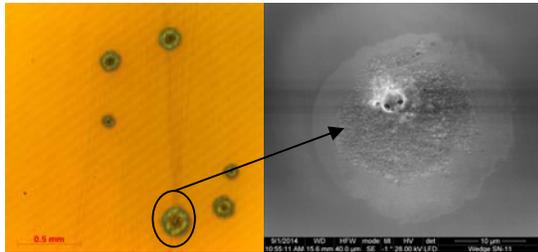


Figure II-1 LIC induced damage event after irradiation in N₂ ambient (microscope image (left) and detail with Atomic Force Microscope (right))

Oxygen is therefore as already demonstrated in literature a necessary ingredient in the safe operation of high energy laser, especially in the UV domain.

III. IN AIR OPERATION: GAS QUALITY VERIFICATION

If the oxygen content is a necessary ingredient to the survival of a laser, it doesn't guarantee safe operation on its own. The residual contaminants in the air supply itself have to be screened and monitored in order to ensure that their concentration is not high enough to induce LIC.

A. Laser Induced Contamination testing

The first logical step would be to determine the threshold concentrations for the formation of an LIC deposit. This is not a trivial matter as it will be chemical composition dependent.

Several different techniques, described hereunder, can be used to identify a potential LIC risk before and during laser operation.

1) Fluorescence

One standard indicator which is used in most tests pertaining to LIC is fluorescence. Fluorescence is a qualitative indicator, which allows to establish when gross contamination is occurring (preferentially in vacuum). It is however "blind" to some contaminants which do not show a large degree of

luminescence. Even more importantly, this parameter is not adapted to any quantitative verification as it is dependent on the fluorescence quantum yield of the deposits and the quenching potential of the surrounding environment both of which can vary considerably.

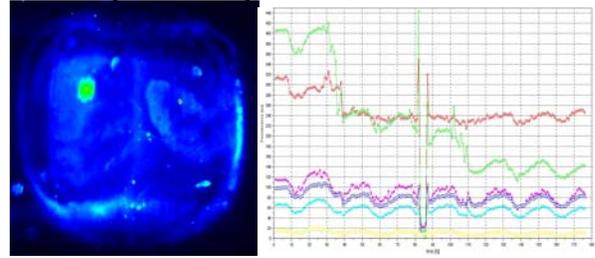


Figure III-1 Fluorescence image FM-A TV test campaign and different areas of interest evolution over test campaign.

On the other hand, as shown in *Figure III-1*, fluorescence based setup can be used as a good indicator of the air quality and alerts raised as needed. Fluorescence verification remains an efficient check during testing but cannot be used to demarcate the usable gas supplies a priori.

2) LIC testing: deposit observation

When testing the suitability of a material in terms of LIC the standard procedure includes the exposure of an optical sample to the contaminant during laser irradiation. The sample surface is then analysed using spatially resolved transmission and white light interferometry. In situ fluorescence is also present during irradiation.

The resolution on the white light interferometry is of the order of magnitude of 1nm, provided that the surface is reflective enough with the deposit in place. Subsequent chemical (ToF-SIMS) analysis can, additionally, determine the nature of the deposit.

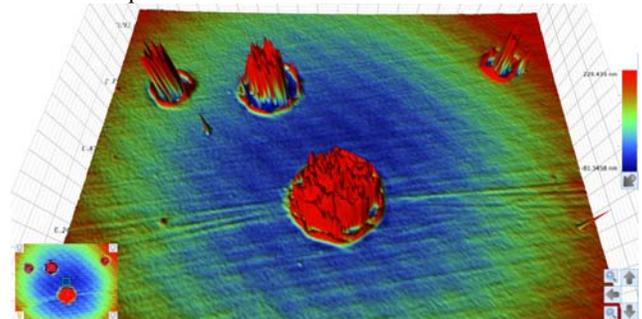


Figure III-2 white light interferometry measurement for LIC induced event presented in *Figure II-1*.

Application of LIC testing in representative fluence conditions, prior to test start, would yield the most cogent proof of quality for a given air supply.

The difficulty in this case comes with adapting the set-up for the verification of an air supply.

A vacuum chamber of reduced size can be easily transported and used. The air under test can be circulated into the chamber, thus essentially using the air supply as the

contaminant in the LIC test. Additionally the clean room air could also be tested as the external surface of the chamber window would be exposed to this environment.

A high energy laser (with its ancillary equipment such as cooling system) is on the other hand difficult to transport. In-house analysis has shown that it is possible to obtain more significant LIC deposit in a continuous wave laser regime, than in the pulsed one, for comparable test duration, with the same contaminant. A small transportable CW laser could thus be substituted in the LIC verification set-up in order to make it transportable and allow for in situ verifications.

The following points have to be taken into account, however:

- Only few contaminants have been tested in CW and pulsed regime both.
- In order to obtain required power/fluences the beam spot for a conventional CW UV laser (tens of mW output) has to be focalized to a size of a few 100µm. Finding a laser induced feature of this size is difficult and time-consuming.
- The optical quality (roughness) of the window has to be sub nm in order to permit identification of eventual deposit. This is not trivial for a brazed window.
- The difficulty in establishing a priori which is/are the irradiated area/s can leave some uncertainty, especially when the deposit is not easily visible. ("Huffing" and checking the evaporation signature of the surface with potential contamination of the same is often the only solution)

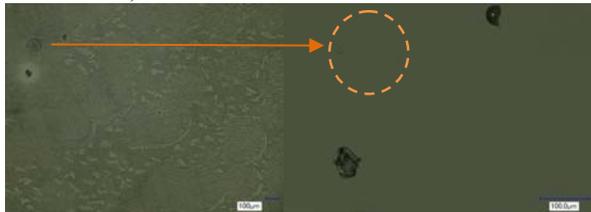


Figure III-3 Beam footprint visualized using huffing (left). Visible droplets on surface in beam footprint after huffing (right). Successful test no WLIM detectable growth.

This type of test, while promising, has not currently sufficient maturity to be the pass/fail criteria for laser switch on.

3) GC-MS analysis

The traditional GC-MS technique for gas analysis remains by default the deciding criterion to stipulate, a priori, if laser operation in presence of air, coming from a clean room interior or from bottles, is acceptable.

B. Air supply filtering against silicones

As previously mentioned, and shown in [5], silicones represent a real danger during UV laser irradiation. In order to reduce the silicone input in an air supply system a trial run has been performed to estimate which attenuation (over short time intervals of a few days maximum) is available after passage

through a sorbent tube for silicone based contaminants. The sorbent tubes used for GC-MS are in fact effective at trapping silicones as they have to allow for their capture prior to release for detection.

In series usage of two such traps allows to reach much lower silicones concentration than those seen on the given purge line as can be shown in Figure III-4.

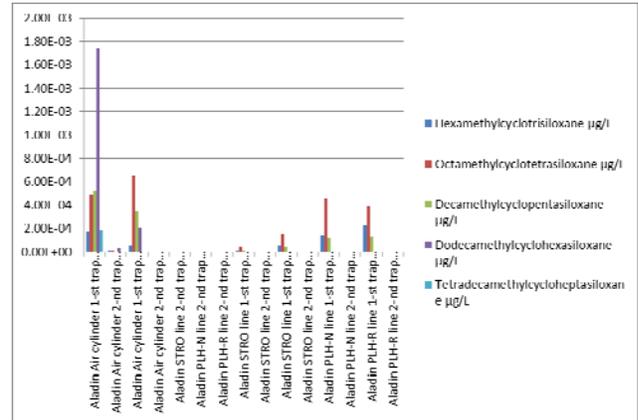


Figure III-4 Reduced concentration [µg/L] of silicone contaminants after two in series traps.

IV. ABSORBING FEATURES

It has been observed that after irradiation in air both in clean room and enclosed (air bottle supplied) environment, small dot-like features, slightly absorbing on transmission optics, are present on the surface of the optics. First observation of this phenomenon occurred during testing of the Aladin instrument in Airbus Defense and Space, Toulouse.

This type of features has also been ascertained in other facilities, including (but not limited to) ESA ESTEC, DLR, Leonardo Finmeccanica Florence... So far, features seem to be ubiquitous, but none have been observed after irradiation in near vacuum conditions with a residual oxygen pressure, yet.

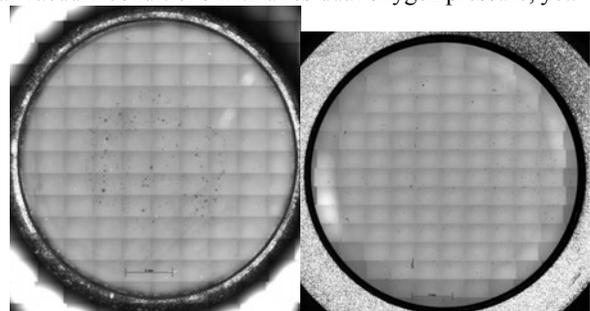


Figure IV-1 Left: input side of laser window, Right: output side. Same environment on both sides.

The formation of these features is not limited to the UV domain as they were observed on IR optics, as well as dichroic (IR/green) mirrors. The coating type and supplier are not influencing significantly the appearance of this phenomenon.

A correlation seems to be present between presence of absorbing features and air environment (more features present on standard clean-room side, compared to enclosed environment) suggesting that a large volume of air passing over the optic is required in order to observe the phenomenon

Analogously features are more abundant on the laser input surface of a transmissive optic. This is well evidenced by the instrument input window sample in *Figure IV-1*. The left-hand (laser input side) shows a high concentration of features in the beam foot-print, the concentration is random and not fluence dependent. The output surface shows dust and a few isolated absorbing features in the central laser irradiated area.

On the laser input side, at higher magnification, the features have a dot-like appearance, and are commingled with rare, small compared to beam size ($\approx 100\mu\text{m}$ vs. $5\times 6\text{mm}$) laser damages, *Figure IV-2*. Same morphology is evidenced in *Figure IV-4* for output surface.

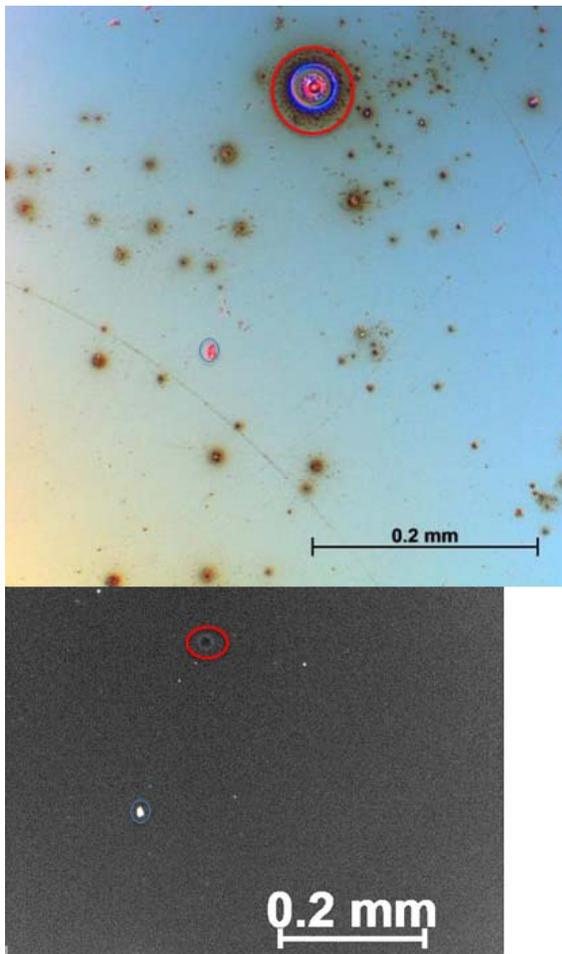


Figure IV-2 Input surface: absorbing features, organic particle and small damage. Bright field, fluorescence.

The features are located on the surface of the optic; the damage structure position with respect to the same is less easily monitored, due to lack of reflectivity in the central area (*Figure IV-3*). Atomic Force Microscopy confirms the position of the absorbing features (*Figure IV-5*).

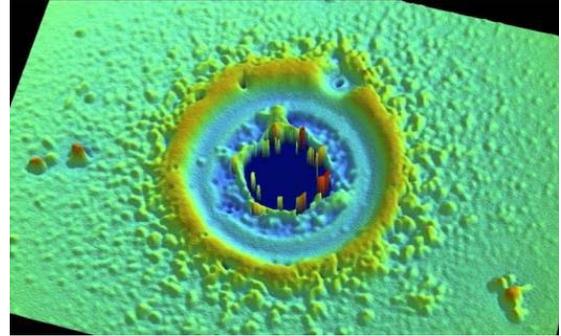


Figure IV-3 WRLM of damage area presenting a few of the absorbing features.

The absorbing features are very faintly emissive at best as it pertains to fluorescence. Not fluorescing, but for the annulus of the damage area as can be seen on the input surface, *Figure IV-2* or only faintly fluorescing, as can be seen on the output side, *Figure IV-4*.

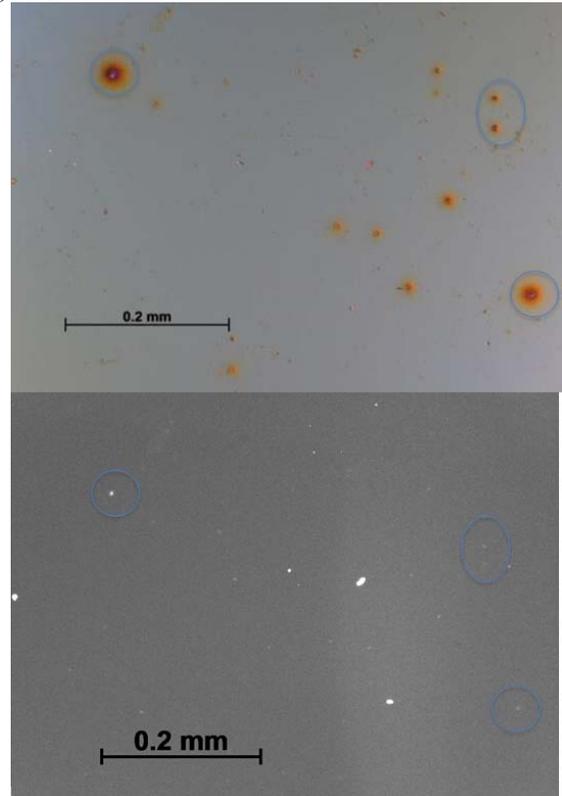


Figure IV-4 Output surface detail bright field and fluorescence.

One of the possible explanations for the lack of observable fluorescence is a silicon based deposit; the presence of weak fluorescence signals for the dot like features on the output surface, on the other hand, points to a potentially different

contaminant as a root cause. In the case of silicone based contamination a higher local concentration of Si and O (SiO_2) would be expected. The shape of a silicone deposit is shown in *Figure IV-6* and is not unequivocally germane to the one observed for the dot like features, the features being much less sparse.

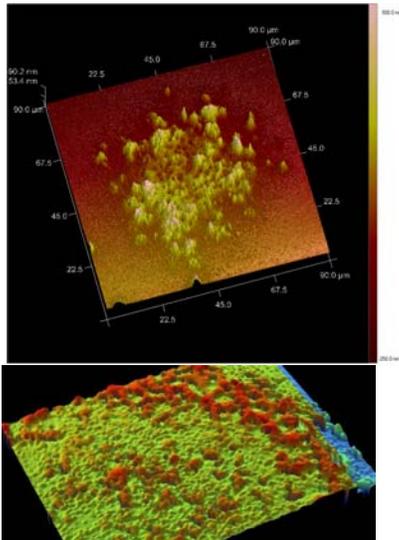


Figure IV-5 AFM (top) and WLIM (bottom) micrographs of instrument output window.

An argument could also be made for increased carbonization of an organic deposit having occurred, thus explaining the reduced fluorescence of the input side of the sample. The fluence would be expected to be slightly higher on the output surface, conversely. On other samples fluorescence signature of features is also variable, and uncorrelated with laser directionality. Higher concentration of C would be expected, from chemical analysis, in this scenario.

A commingled origin (silicone based and not) cannot be excluded. More detailed chemical analysis on available samples is required to further understanding of this phenomenon.

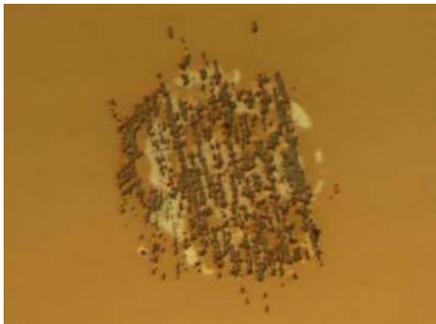


Figure IV-6 Power Laser Head samples 01 from DLR showing silicone based contamination covering whole beam area.

A. Chemical analysis of absorbing features

Several different techniques can be used to determine the constituents of a deposit present on an optical surface. Given the likely complexity of the molecules involved, techniques

such as Energy Dispersive X-ray Spectroscopy (EDS) are not adapted as they will give concentrations of constituent elements (such as carbon) only.

Chosen techniques were ToF-SIMS and X-ray Photoelectron Spectroscopy.

The former will the lateral distribution of the the various molecules or elements on the surface with very good sensitivity, affording a cartography of the different ions present on the surface. The latter gives a quantitative measure of the concentration of base elements with additional information on their chemical state (thus permitting to distinguish between possible different source molecules.)

ToF-SIMS can also be used in conjunction with Argon cluster sputter cleaning to remove surface organics and thus examine the underlying layers of a deposit.

The SIMS chemical maps obtained on several different samples are presented in the following sections.

1) Time-of-Flight Secondary Ion Mass Spectroscopy results

Using ToF-SIMS, a cartography of a beam footprint or a smaller area around a damage or several dot like features is possible. In the following we shall present the details of several different samples.

a) Beam foot-print

Consistently the presence of Phosphates (PO_2^- and PO_3^-) on the optical surface in conjunction with the laser foot-print is evidenced, *Figure IV-7*.

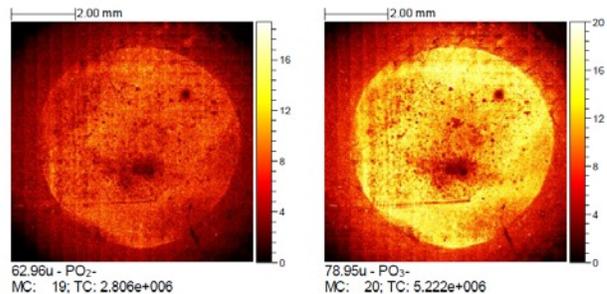


Figure IV-7 Aladin Laser FM-A output window: SIMS map for negative phosphate ions. Beam footprint evident, depletion co-located with damage structures.

In analysis so far the phosphate deposit is accompanied by not fully co-located deposits of Na, K, amines (CN^- , CNO^-), F and Cl.

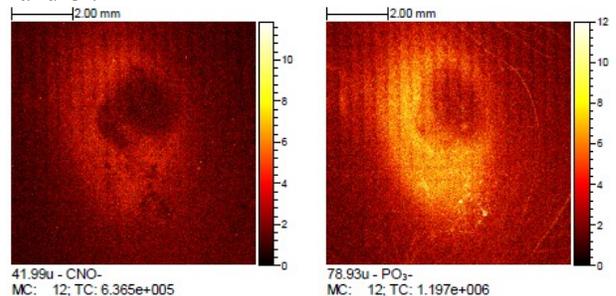


Figure IV-8 Long Term Sample A, halo at extrema of beam.

The phosphate/ amine deposit is not always covering the whole beam footprint as seen in *Figure IV-8*, but is occasionally present as a halo surrounding the beam.

The corresponding depletion of Si in the SIMS map of the positive ions corresponding to the laser footprint shows that a deposit is covering the optical surface (*Figure IV-9*). This is also a generic observation which holds for all samples investigated so far.

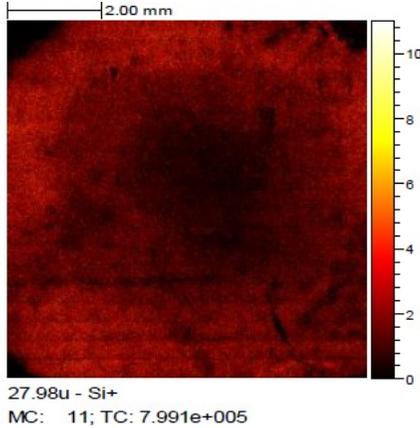


Figure IV-9 Aladin Laser FM-A output window SIMS map of positive ions. Si depletion is evident in the beam footprint.

On two samples, the ones shown above higher levels of metals, specifically Al, Fe, Cr, Cu, Ni, Ti and higher levels of polydimethylsiloxane (PDMS, i.e. SiC_3H_9^+ and $\text{Si}_2\text{C}_5\text{H}_{15}\text{O}^+$) and aliphatic organics (e.g. C_2H_3^+ , C_2H_5^+ , C_4H_7^+) are also associated with the laser area.

PDMS, Cl and F are also present in control areas outside the beam, so are the phosphates but, as evidenced in *Figure IV-7*, with lower levels.

b) Damage area

If we now concentrate on an area around a damage feature such as the one observed on the Aladin FM-A output window the optical image (*Figure IV-10*) shows a clear correlation with the chemical maps.

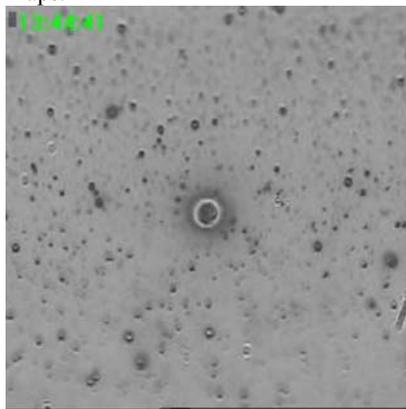


Figure IV-10 Aladin Laser FM-A output window: optical microscope image of detailed area of analysis taken from ToF-SIMS set-up.

Correlation is obvious with the phosphates which show a significant depletion in the center of the damage structure, a bright ring and a second darker annulus.

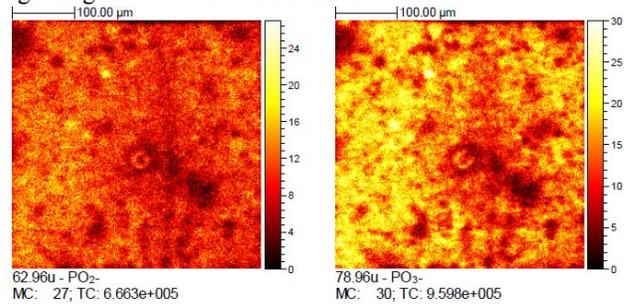


Figure IV-11 Aladin Laser FM-A output window: SIMS map of negative ions before Ar sputter cleaning.

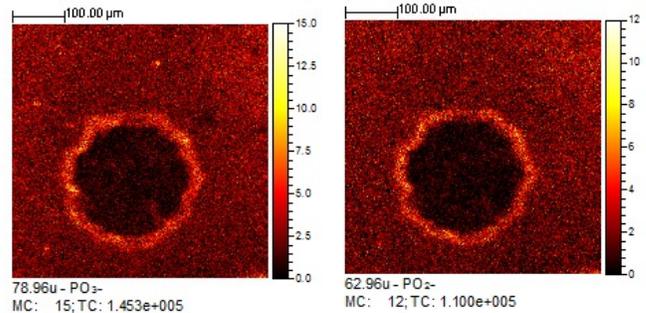


Figure IV-12 LIC induced damage after laser irradiation in N₂ ambient ToF-SIMS detail of circled area.

After sputter cleaning the majority of the remaining organics, PDMS, Cl and F have been removed. In the positive SIMS maps the presence of metals (iron) at the centre of the damage structure becomes evident. Also notable is the corresponding depletion of silicon which correspond to the presence of a metallic particle (i.e Fe) in or on the coating as a potential root-cause for the damage creation (*Figure IV-13*).

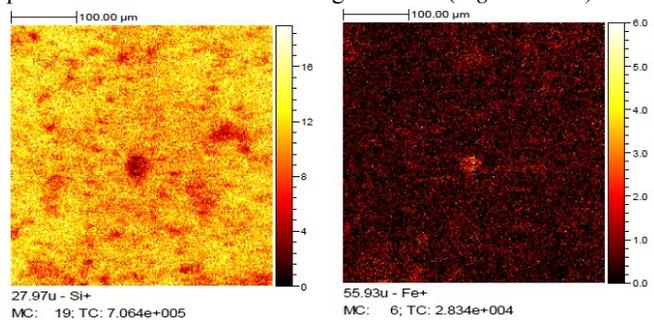


Figure IV-13 Aladin Laser FM-A output window: SIMS maps of Si+ and Fe+ after sputter cleaning.

Analysis of negative ions maps shows (*Figure IV-14*) significant residual of the phosphate deposits still present after Ar cluster sputter cleaning.

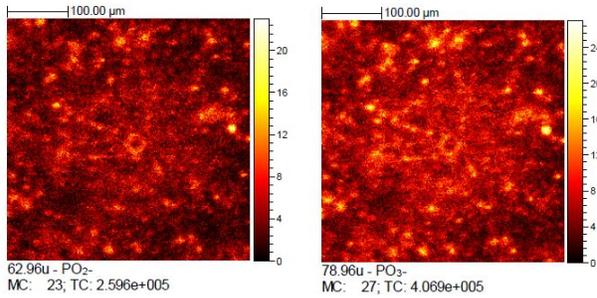


Figure IV-14 Aladin Laser FM-A output window: SIMS maps of PO_2^- and PO_3^- after sputter cleaning.

c) Black dot like absorbing features

The absorbing features themselves show a consistent chemical make-up with the damage structures. The best sample to visualize the dot like features without any damages (which have a potential to relocate contaminants) is a flight hard-ware grade Half Wave Plate coming from a test set-up (Figure IV-15).

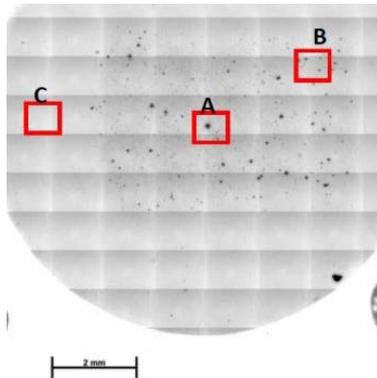


Figure IV-15 HWP from Airbus Toulouse laser test set-up.

Analysis of the samples has shown the customary slew of contaminants associated with the laser footprint on the surface (PDMS, aliphatic organics, the ubiquitous phosphates etc.).

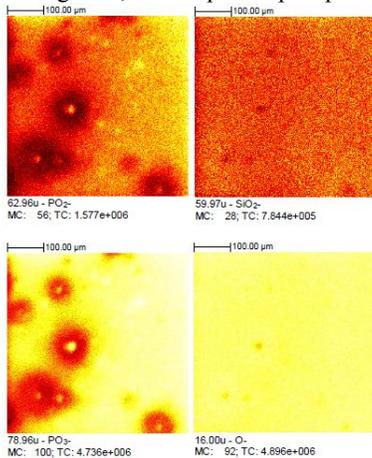


Figure IV-16 Area B negative maps: PO_2^- , SiO_2^- , PO_3^- , O^- .

In area B where only dot like features are present, the negative ion maps show the usual good correlation between phosphates and surface structures. The centers of the dot like features show higher phosphate content than the borders. This may be due to view factors, and may also hold for the damage features.

Significantly no accretion (in fact a faint depletion can be surmised) of SiO_2 , Si or O is visible in correspondence with the dot like features. This tentatively rejects the hypothesis of an origin linked with silicones (PDMS are present on the surface but their localization is also not germane).

As far as the positive ion maps are concerned it is important to recognize the presence of metals associated with some (but not all) of the features.

Chemically the signature of the dot like features is similar to that of the damages.

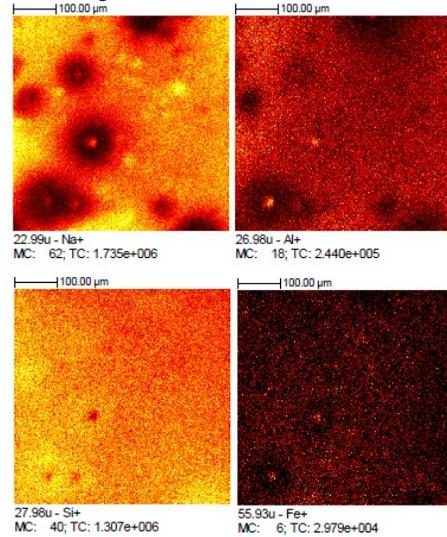


Figure IV-17 Area B positive maps: Na^+ , Al^+ , Si^+ , Fe^+ .

2) X-ray Photoelectron Spectroscopy

The presence of a phosphorous content in irradiated area, but not outside, is confirmed by XPS.

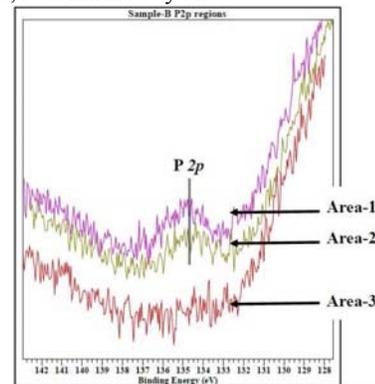


Figure IV-18 Phosphorous $2p_{1/2+3/2}$ peak, coming from phosphates. Concentration in Area-1 beam footprint, centre, Area-2 beam footprint periphery, Area-3 control area outside of beam.

B. Evolution of the absorbing features

In order to determine whether the features are damage precursors, likely to evolve in catastrophic damage, further irradiation on affected samples has been undertaken at 355nm.

At least a commercial sample, from a test set-up has been exposed to over 2Gshots irradiation. Progressively the concentration of small damage feature and dot like absorbing features has increased. No catastrophic damage has been observed.

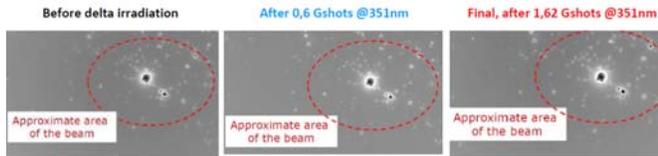


Figure IV-19 Evolution of damage precursor and absorbing features during irradiation up to 1.6Gshots.

Analogously on a flight batch sample from long term irradiation test (Figure IV-19) has been submitted to further irradiation (additional 1.6Gshots) showing few cosmetic changes and marginal transmission loss.

Several more samples on the test set-ups in the different laboratories are now under scrutiny and their evolution with time is being monitored. So far evolution is resulting in progressive attenuation (a few percent) and creation of both black dot like features and small non catastrophic damages.

V. CONCLUSIONS

Air quality is the most likely long term limiting factor for a high-energy laser operating under pressure. Air quality monitoring is a necessity prior and during laser irradiation to ensure Gshot operation of high energy lasers.

Operation in air of a high energy laser results in creation of small dot like absorbing features for which evolution with additional irradiation can be discounted, so far. Progressive attenuation for transmissive elements will be obtained. The feature aspect is remarkably similar, visually, on all the different elements and coatings. The only driving criterion for the appearance of the “black dots” seems to be the presence of a more/less stringently controlled atmosphere. Usage of high purity (above 99.999%) gas bottles is not sufficient to prevent their formation.

Chemical analysis suggests that catastrophic damage root cause/s, at least in air operation, may be the same which lead to the formation of the damages interspersed with the black dot

absorbing features. No obvious discriminant between catastrophic event and benign features is evident so far, but may be dependent on size/type/location of metallic damage core, and level of associated LIC coming from ancillary organic contamination. Similarly chemical signature for dot-like features is analogous to that found for the small damages.

The lack of correspondence between PDMS/SiO₂/Si/O₂ maps and absorbing feature maps rebuts the presence of PDMS (silicones) as the root cause for the creation of the dot like structures.

The origin of the phosphate contamination is not known so far but it has to be observed that a potential candidate may reside in flame retardants, which would be present (but not in a significantly volatile form) in a standard clean room environment. Screening for phosphate contamination is suggested. Unfortunately GC-MS is not well adapted to phosphate detection, adaptation of LIC detection techniques to air contamination analysis may be beneficiary.

ACKNOWLEDGMENT

The authors thank Airbus Defense and Space, Leonardo Finmeccanica, DLR ITP and ESA OOEL and Aeolus teams for their insights on on-ground laser operation.

REFERENCES

- [1] W. Riede et al. “*Laser-Induced hydro-carbon contamination in vacuum*”, SPIE Vol 2714, 2005.
- [2] D. Wernham, et al. “*Risk Mitigation for Laser-Induced Contamination on the ADM-Aeolus Satellite*”, SPIE Laser Damage 2010 Symposium proceedings.
- [3] D. Morancais, “*The Atmospheric Wind Lidar Instrument (ALADIN): Development Status*,” in Advanced Solid-State Lasers Congress, P. Mc Manamon, . Watson, and O. Steinvall, eds., OSA Technical Digest (online) (Optical Society of America, 2013), paper LTh1B.2.
- [4] M. Hippler Diploma Thesis, University Dresden, “*Influence of Oxygen on laser induced Contamination*”, 2015
- [5] H. Schröder et al, “*Investigation of UV laser induced depositions on optics under space conditions in presence of outgassing materials*”, ICSO 2006.
- [6] Hovis, Floyd et al. “*Mechanisms of Contamination Induced Damage in Lasers*”, SPIE Proceedings Vol. 2428, pp72-831995.