

Determining the Role of Oxygen in Obtaining Long-Term Stable Superhydrophilic Surfaces on Metals Treated with a Femtosecond Laser

Vadim Ialyshev and Ali S. Alnaser*



Cite This: *ACS Omega* 2023, 8, 33904–33911



Read Online

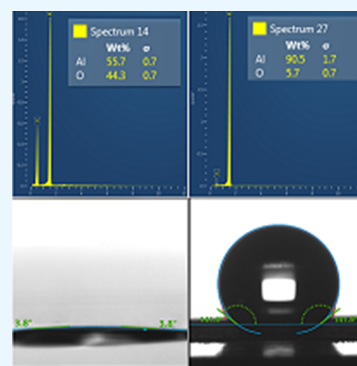
ACCESS |

Metrics & More

Article Recommendations

Supporting Information

ABSTRACT: Laser processing is a simple way to obtain hydrophobic or even superhydrophobic properties of metal surfaces. However, preparation of superhydrophilic surfaces by this method, the properties of which do not change under the influence of various factors, remains a difficult task. In this work, we show that with increasing laser power, the degree of oxidation of the treated metal surface also increases. As a result, highly oxidized samples showed highly stable superhydrophilic properties. A Janus membrane fabricated from a stainless steel mesh with asymmetric hydrophilic-hydrophobic wettability demonstrated stable water diode properties. In addition, it was found that during the examination of sample surfaces by Raman spectroscopy, organic compounds adsorbed on the hydrophobic surface were decomposed by the laser of the spectrometer, which imposes limitations on the laser power when using this method in characterizing hydrophobic surfaces of metals fabricated by laser processing.



1. INTRODUCTION

The interaction of liquids with solid surfaces is one of the essential properties of a material, depending on which this material can be used in specific applications. The ability of a liquid to maintain contact with a solid medium is determined by the intermolecular interaction and depends on both the degree of surface roughness and its chemical composition.^{1–3} Therefore, the properties of both the solid and liquid surfaces are critical at the interface to determine the final wetting behavior. It is important to understand how to fabricate the desired topography and impose favorable chemistry on engineering metal substrates for desired wettability. It has been widely demonstrated that the wettability state of a solid surface can be changed by micro/nanostructuring of the surface.^{4–6} Fabrication techniques such as thermal oxidation, chemical vapor deposition, laser structuring, and plasma spray were widely used to create wetting properties based on two primary factors, and those are surface chemistry and surface micro-/nanostructures.^{7–11} Laser processing of metal surfaces is a reliable technique to produce superhydrophilic or superhydrophobic materials, depending on the surface energy of the material.^{12–15} Furthermore, a femtosecond laser with its good consistency, high precision, and negligible heat-affected zone is a one-step solution for creating multifunctional surfaces. Modification of the surface morphology under ultrafast laser irradiation is mainly carried out by ablation of a localized area with a focused beam, which makes it possible to change both the chemical composition of the material and the surface microstructure.^{16–19}

Various metallic materials, such as aluminum, stainless steel, copper, titanium, and others, have been used as substrates to prepare superhydrophobic or superhydrophilic surfaces. Superhydrophobic surfaces of metals attracted lot of attention and research efforts in recent years due to potential applications in many fields, such as self-cleaning, anti-icing, anticorrosion, or enhanced heat transfer.^{20–23} On the other hand, hydrophilic metallic materials with good adhesion surfaces are considered important applications for both scientific findings and practical applications, such as cell-based biosensors, cell–cell communication, coating fabrication, and water-assisted flow generation.^{24–26} In addition, Janus membranes with asymmetric hydrophobic/hydrophilic sides make them promising in many applications such as liquid manipulation, highly efficient separation of oil and water, water harvesting, and switchable ion transport.^{27–31}

Freshly laser-processed samples are usually superhydrophilic due to the enhanced surface roughness and the formation of metal oxide sites created during treatment.^{32–34} When oxygen molecules interact with the treated metal surfaces, polar compounds are formed, as a result of which the surface energy increases, allowing attracting the water droplets. However, it

Received: June 28, 2023

Accepted: August 21, 2023

Published: September 5, 2023



was found that the laser-textured metals transfer from a hydrophilic to hydrophobic state when samples are left in ambient air. This process occurs due to the chemisorption of nonpolar hydrocarbons contained in the air and does not require any additional effort or chemicals.³⁵ The transition, in which the laser-treated metal surface acquires hydrophobic or superhydrophobic properties, could last from several days to several weeks. On the other hand, storing laser-processed metals in a vacuum significantly reduced this transition time (up to several hours).³² One of the reasons for this phenomenon is the very low water vapor content inside the vacuum chamber, which avoids passivation of OH centers by hydrogen-bound water molecules. Another is the presence of organic contaminants in the vacuum chamber, which may come from the lubrication system of the vacuum pumps. Consequently, the chemisorption of hydrocarbons occurs much more efficiently in the chamber than in atmospheric conditions.

Thus, the superhydrophobic surfaces of metals can be fabricated by laser processing and subsequent storage of samples in a vacuum chamber. However, obtaining long-term stable superhydrophilic properties of laser-treated metals without additional treatment or coating remains a challenge. The stable hydrophilicity of metals within 30 days after their laser treatment has been shown in several studies.^{36,37} However, these results are not entirely correct, since the samples were stored in air, where the time of transition from a hydrophilic to a hydrophobic state is largely determined by the concentration of hydrocarbons in the air.

In the present work, laser-treated surfaces of aluminum plates were tested for their ability to retain their hydrophilic properties during long-term storage in the environment or in a vacuum chamber. It was found that the main factor determining the stability of the hydrophilic state was the oxidation of the structured surface created by the laser. An increase in the degree of oxidation of the samples, depending on the power of the laser used, improved the ability of aluminum to retain its obtained hydrophilicity from the transition to a hydrophobic state. To confirm the role of oxygen, the properties of samples prepared in air and in an argon medium were compared. Based on the results obtained, a Janus membrane with asymmetric hydrophobic–hydrophilic wettability of the sides was fabricated from a stainless steel mesh. This membrane demonstrated stable water diode properties, in which water could only pass through the mesh in one direction. In addition, it was found that, under certain measurement conditions, the XPS method commonly used to analyze the composition of the surface of samples could be replaced by the simpler method of Raman spectroscopy. It was found that the intensity of the peaks in the Raman spectra, attributed to organic compounds adsorbed on the hydrophobic surface, decreases with time due to the degradation of these compounds upon the interaction with the laser used with the Raman spectrometer. Thus, a necessary condition to properly analyze the chemical composition of hydrophobic–hydrophilic surfaces of laser-treated metals by Raman spectroscopy is to decrease the power of the spectrometer's laser.

2. MATERIALS AND METHODS

Commercially pure aluminum plates (Al 99.6%, ALDRICH) with 2 mm thickness were used as the main investigated material. Stainless steel meshes with a pore size of 350 μm and a wire diameter of 180 μm were additionally used as a tested

material. For the treatment of surfaces, an ultrafast Yb-doped fiber laser (UFFL_300_2000_1030_300; Active Fiber System) with a central wavelength of 1030 nm, a repetition rate of 50 kHz, and a pulse duration of 40 fs was used. The laser beam with linear polarization and different powers was directed to a computer-driven galvanometric scan head (FARO tech., Xtreme-20), with which the laser could process the metal surface with the specified parameters, as shown in Figure 1.

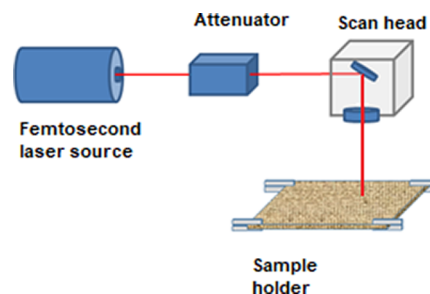


Figure 1. Schematic diagram of the laser texturing system. The direction and speed of the beam scanning from the femtosecond laser source were controlled by the scanning head and focused on the sample surface.

The scanning speed and interval between lines were 15 mm/s and 20 μm , respectively. The surface of the samples was patterned by a crosshatch configuration. To control the power of laser pulses, we used a half-wave plate and a thin film polarizer. The polarization of the laser beam was perpendicular to the scan direction. F-Theta lens of the scan head with $f = 160$ mm provided a spot size (~ 70 μm) across the entire image plane.

The surface morphology of the laser-treated samples was studied using a scanning electron microscope (SEM, TESCAN, Czech Republic) equipped with an X-ray detector (energy-dispersive X-ray spectroscopy (EDS, Oxford). The water contact angle (WCA) was measured by the sessile drop technique using a video-based static contact angle computing device (Drop Shape Analyzer, KRUSS, Germany) with a distilled water droplet volume of 5 μL . The X-ray diffraction (XRD) pattern was recorded on a Panalytical X'Pert diffractometer with a monochromatic Cu-K α radiation source at θ -2 θ configuration. The samples were analyzed by a Raman microscope (Witec alpha 300) equipped with a spectrograph (Witec UHTS 300).

3. RESULTS AND DISCUSSION

Laser ablation makes changes in the surface morphology and chemical composition, which strongly depend on the laser power. Figure 2 shows the SEM images and EDX spectra of the surfaces structured by femtosecond pulses at 2, 4, and 8 W laser power in air. An increase in the laser radiation power leads to a significant change in the morphology of the metal surface both in area and in depth, reaching ~ 100 μm in thickness at 8 W. At the same time, the atomic weight fraction of O increased from $\sim 17\%$ for the sample treated at 2 W to $\sim 45\%$ for samples treated at 8 W, indicating oxidation of the Al surface after the femtosecond laser treatment.

Just after laser treatment, the original wettability of samples (WCA $\sim 80^\circ$) acquired pronounced superhydrophilic properties with the WCA less than 5° . The sample treated with a laser power of 8 W showed superhydrophilic behavior, in which a drop of water, after contact with the treated surface, quickly

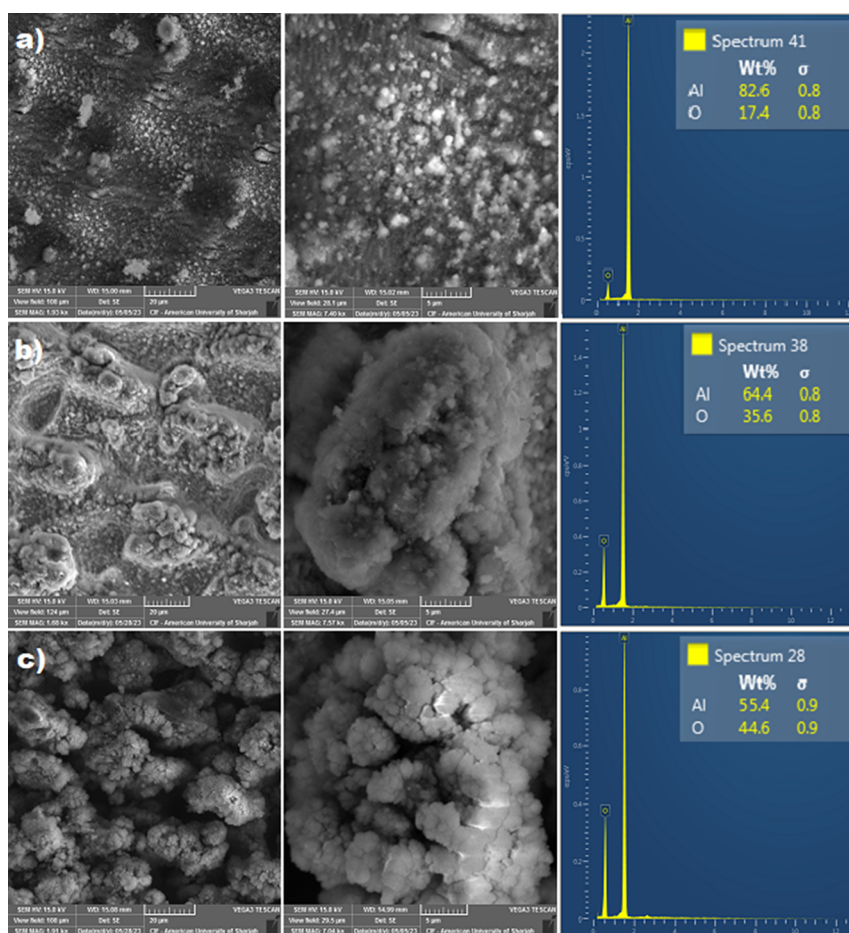


Figure 2. SEM images (resolution scales 20 and 5 μm) and EDX spectra of aluminum samples processed at a laser power of 2 W (a), 4 W (b), and 8 W (c) in air.

spread out in a horizontal direction (see [Movie S1](#) in the Supporting Information). This superhydrophilicity of the surfaces was attributed to the enhanced surface roughness and accumulation of a large number of polar oxide sites created on the treated surface, which have a strong affinity for hydroxylation.^{32–34}

It is well known that laser-treated hydrophilic metallic surfaces become hydrophobic or super-hydrophobic when exposed to air for a long period of time.³⁵ The primary reason for this wetting transition is related to absorption of organic compounds from the air. This is a very slow process, and in ambient conditions, it can take from a few days to several weeks. However, it has been found that vacuum storage of laser-treated metal samples greatly accelerates the wettability transformation from hydrophilic to hydrophobic/superhydrophobic, taking only a few hours.^{17,23,32} The adsorption of hydrocarbons under vacuum takes place in a more efficient way than in air due to a lower amount of water molecules, which passivate OH sites on the surface. Thus, the accelerated wettability transition of samples kept under vacuum was basically due to the adsorption of hydrocarbon molecules inside the vacuum chamber. The main source of organic contaminants is primarily the vacuum pumps, which usually use mineral oils and additives that can flow through the pumping line into the chamber and get adsorbed onto metal surfaces.

The typical values of the WCA obtained from samples stored in the vacuum chamber for 12 h are shown in [Figure 3](#).

As can be seen, the samples processed at a laser power of 2 and 4 W were successfully converted to hydrophobic states with a WCA of about 130°. The samples processed at 8 W almost did not change their superhydrophilicity, only slightly increasing the WCA to 4°. It should be noted that these samples remained superhydrophilic even after 3 months.

The main differences between the samples processed at low (2 and 4 W) and high (8 W) laser powers were the morphology and degree of oxidation of their surfaces. Therefore, in order to find out the influence of these two factors on the wettability properties of the samples, one sample was prepared using 8 W laser power in an inert argon gas medium. As can be seen from [Figure 4](#), the samples prepared in air and Ar environments have a similar nanostructural morphology, indicating that the growth of structures does not depend on the processing medium. However, due to the lack of oxygen during laser treatment, the surface remains practically nonoxidized. An insignificant amount of oxygen observed in the EDX spectrum can be caused by both natural oxidation and the presence of hydroxyl groups on the surface when the sample comes into contact with air. At the same time, the sample treated in an argon atmosphere became highly hydrophobic after storage in a vacuum, which clearly indicates the main role of oxidation in the transformation of wettability from a hydrophilic state to a hydrophobic one.

Laser processing of metals in air leads to the formation nano/microstructures of metal oxides on the treated surface. Metal oxides have a higher surface energy and tend to be

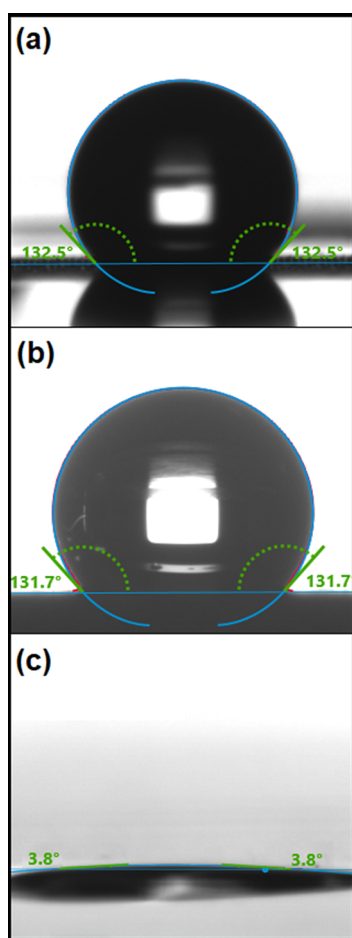


Figure 3. Typical photographs of WCA measurements of aluminum samples processed at a laser power of 2 W (a), 4 W (b), and 8 W (c) after storing in a vacuum chamber.

hydrophilic because their electronic structure favors the formation of hydrogen bonds.¹⁵ Therefore, immediately after laser processing, the surface usually demonstrates superhydrophilic behavior. For example, the process of laser ablation of aluminum leads to the formation of Al^{3+} and O^{-2} , which subsequently create a passivating Al_2O_3 layer on the surface. Since aluminum atoms on the surface are electron-deficient, this leads to the formation of a hydrogen bond with interfacial water molecules, promoting the heterolytic dissociative adsorption of water molecules.³² Therefore, surfaces after laser treatment behave as superhydrophilic. However, hydroxyl groups on the surface can act as effective adsorption or reaction sites. It has been established that the presence of hydroxyl groups on the surface of oxides plays a significant role in the chemisorption of organic molecules, for example, airborne hydrocarbons, which can include short nonpolar or hydrophobic molecules and thus decrease the surface energy and transform the wetting property from hydrophilic to hydrophobic.³²

Increasing the laser power leads to both the creation of a more developed nanostructured surface and its greater oxidation. The adsorption of nondissociated water molecules on the surface of highly oxidized metals can significantly slow down the adsorption of hydrocarbons due to the formation of a passive layer on reactive sites. We attribute this to be the main reason for the stable hydrophilicity of the samples.

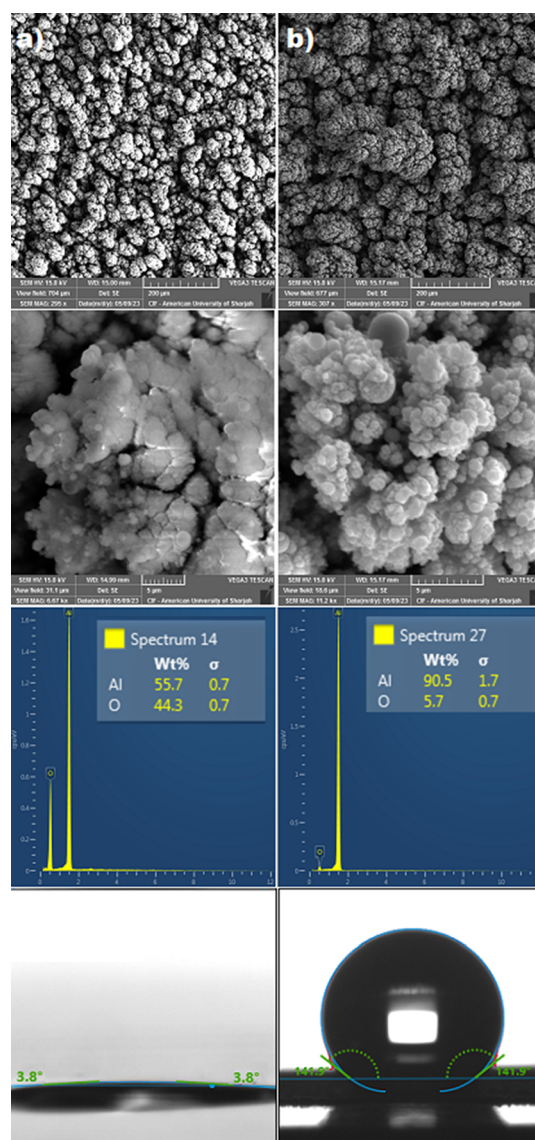


Figure 4. SEM images (resolution scales 200 and 5 μm), EDX spectra, and photographs for WCA measurements of the aluminum samples processed at a laser power of 8 W in air (a) and Ar (b) medium. The WCA measurements were carried out after the samples were stored in a vacuum for 12 h.

The XRD patterns of the samples treated in Ar and air environments are shown in Figure 5. For comparison, the spectrum of untreated Al was also added. The four strong diffraction peaks were observed in the spectra at 38, 44, 65, and 78 degrees, which correspond to (111), (200), (220), and (311) crystallographic orientation of aluminum (JCPDS No. 89-4037). As can be seen, after laser-processing the samples, the intensity of the diffraction peak in the plane (111) increased significantly (for greater clarity, this peak is shown separately in the inset of the figure). This increase in intensity may mean that the microstructure obtained on the sample surface is mainly formed from crystals with the (111) orientation. A decrease in the intensity of this peak observed in samples treated in air may be associated with the oxidation of these crystals. On the other hand, the absence of peaks corresponding to aluminum oxide in the spectra suggests their amorphous nature.

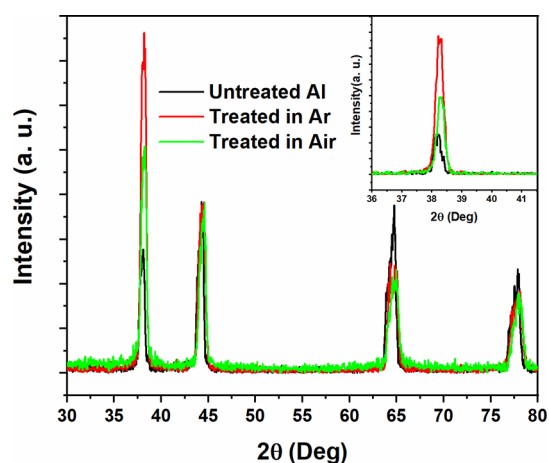


Figure 5. XRD patterns collected from untreated and laser-treated in air and Ar environments at a laser power of 8 W.

Since XPS is a popular and powerful method for studying the surface composition, it has usually been used to analyze hydrophobic and hydrophilic surfaces of laser-treated samples. At the same time, only a few studies were found in the literature where another method of surface analysis, such as Raman spectroscopy, was used for these purposes. Perhaps, as it was found in this work, this was due to the degradation of organic components on the surface during their interaction with the laser used in the spectrometer. When using a laser with energies above 10 mJ, the difference in the signals from hydrophilic and hydrophobic samples was negligible. However, the difference in spectra was clearly observed at a laser energy of 2 mJ, as shown in Figure 6. The broad band at ~ 800 and ~ 1800 cm^{-1} could be assigned to the asymmetric COO⁻ deformation of the carboxylate groups of organic molecules adsorbed on the surface after storage in a vacuum chamber.³⁸

These peaks degraded under laser irradiation (Figure 6b). After the first measurement, the sample remained under the laser and was measured again. After 10 and 20 min of irradiation, the initial intensity of the 800 cm^{-1} band was decreased by 1.7 and 2.9 times, respectively. This degradation of the signal could be caused by desorption of organic

molecules from the surface due to the heating effect. As was shown, the hydrophobicity of samples obtained as a result of vacuum storage could be converted back to hydrophilicity by annealing them in air at a temperature of 250 °C, as a result of which organic components were removed from the surface of the samples.³⁹ Thus, the absence of this signal at a laser power of more than 10 mJ could be caused by rapid heating of the samples.

Based on the above results on the important role of the oxidation state in obtaining a permanent hydrophilicity of the metal surface, we applied this method to fabricate a Janus membrane using a stainless steel mesh. To produce a hydrophobic/hydrophilic asymmetric structure, one side of the mesh was processed by the laser with a power of 8 W. Figure 7 shows SEM images, EDX spectrum, and WCA of the laser-treated side of the mesh. As in the case of an aluminum plate, the formation of laser-induced structures on the surface of the mesh wires was accompanied by their oxidation. As expected, after laser treatment, the wettability of the sample changed to hydrophilic with a WCA of $\sim 58^\circ$ (versus 120° for the untreated mesh). Due to the porous mesh geometry, a drop of water cannot completely spread over its surface.

The asymmetric wettability of the Janus membrane surfaces (hydrophilic on the treated side and hydrophobic on the untreated side) provides the behavior of the water diode in this system. As shown in Figure 8a (see also Movie S2 in the Supporting Information), the passage of water droplets through the mesh was blocked by the bottom hydrophobic side, as a result of which they accumulated on the hydrophilic side. In contrast, when water droplets from the upper hydrophobic side were in contact with the hydrophobic part of the mesh, they could easily pass through the mesh, thereby demonstrating the properties of a water diode (Figure 8b and Movie S3 in the Supporting Information). This anisotropic water transport property arose from a cross-sectional wettability gradient created in the mesh by laser treatment.⁴⁰ As can be seen from Figure 8c,d, almost the entire surface of the untreated side of the mesh wires was covered with deposited micro/nano oxide droplets. These droplets create a wettability gradient along the perimeter of the wire, starting with superhydrophilic on the treated side and gradually turning

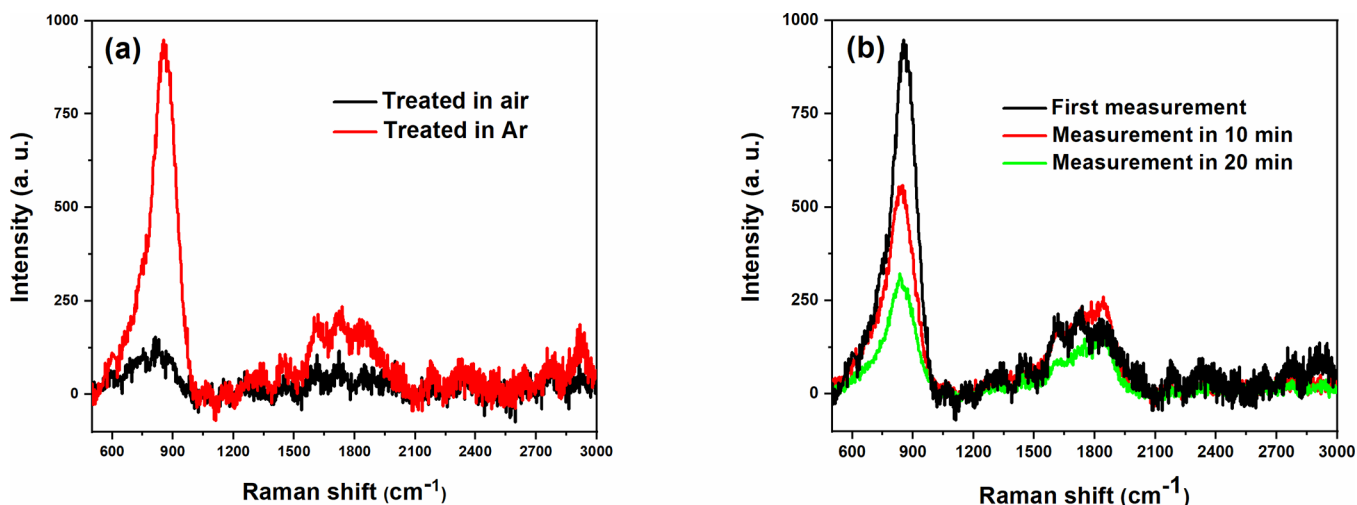


Figure 6. (a) Raman spectra taken from Al samples treated in air (superhydrophilic) and Ar (highly hydrophobic) after storage in a vacuum for 12 h. (b) Dependence of the spectrum of the sample processed in the Ar medium on the time of laser irradiation of the spectrometer.

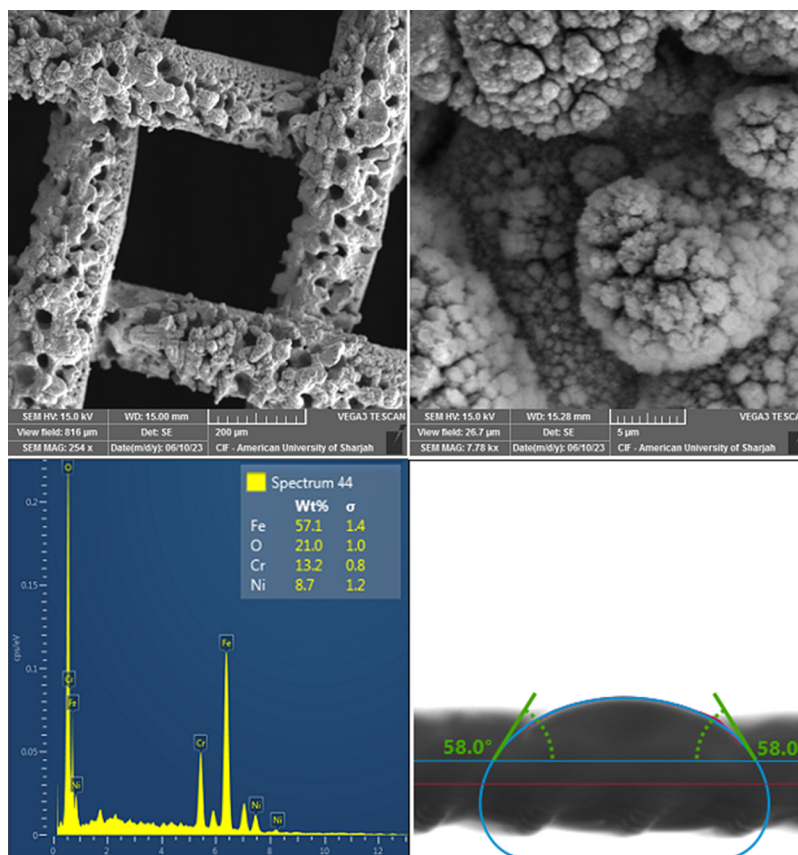


Figure 7. SEM images (resolution scales 200 and 5 μm), EDX spectra, and photographs for WCA measurements of the stainless steel mesh processed at a laser power of 8 W in air.

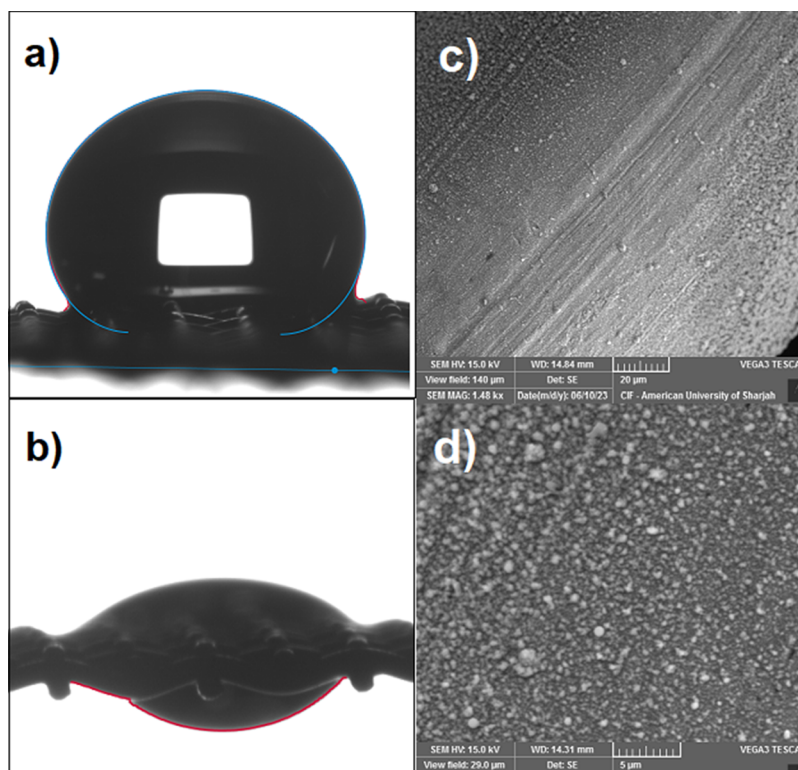


Figure 8. Asymmetric water transport of the mesh with different wettability surfaces. (a) Accumulation of drops on the hydrophilic side. (b) Passing of drops through the mesh from the hydrophobic side. SEM images of wires from the untreated side of the mesh (resolution scales (c) 20 and (d) 5 μm).

into hydrophobic on the untreated side. Depending on the direction of the wettability gradient along the wires, there is a difference in the critical pressure of the water breakthrough, which is decisive when water passes through the mesh.⁴¹

Finally, the mesh was placed in a vacuum chamber for 24 h to check the consistency of its properties. As expected, the wettability of the treated side remained hydrophilic and the Janus membrane continued to demonstrate its asymmetric water transport properties.

4. CONCLUSIONS

This study demonstrated one way to obtain long-lasting hydrophilic metal surfaces prepared by laser treatment without additional chemical coating. It was shown that an increase in laser power contributed to the creation of a highly developed nanostructured surface with its simultaneous strong oxidation, as a result of which a passivating oxide layer was formed on the surface. As a result of this amorphous oxide layer, the acquired high hydrophilicity of metal samples remained unchanged for a long time, without showing any symptom of transition to a hydrophobic state, which was usually observed in metals after their laser treatment. This method has been tested on both aluminum plates and stainless steel meshes. The Janus membrane with asymmetric wettability, fabricated from the mesh, continued to demonstrate the properties of a water diode even after 3 months of monitoring. In addition, organic molecules adsorbed on surfaces could be detected using Raman spectroscopy (instead of the commonly used XPS method). However, this method required a reduction in the laser power of the spectrometer, since during the measurement, these hydrocarbons degraded under laser irradiation.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.3c04618>.

Movie S1 – laser-treated superhydrophilic surface of the Al plate (MP4)

Movie S2 – untreated hydrophobic side of the mesh is bottom (MP4)

Movie S3 – untreated hydrophobic side of the mesh is top (MP4)

■ AUTHOR INFORMATION

Corresponding Author

Ali S. Alnaser – Department of Physics and Materials Science and Engineering, College of Arts and Sciences, American University of Sharjah, Sharjah 26666, United Arab Emirates; orcid.org/0000-0003-4822-9747; Email: aalnaser@aus.edu

Author

Vadim Ialyshev – Department of Physics and Materials Science and Engineering, College of Arts and Sciences, American University of Sharjah, Sharjah 26666, United Arab Emirates

Complete contact information is available at: <https://pubs.acs.org/doi/10.1021/acsomega.3c04618>

Notes

The authors declare no competing financial interest.

Single water droplet and multiple water droplet penetration processes on the Al plate and stainless steel meshes

■ ACKNOWLEDGMENTS

This study was supported by FRG grant # FRG19-L-S61 from the American University of Sharjah, United Arab Emirates. We thank Mazhar Iqbal and Piotr Piatkowski for their help with the measurements.

■ REFERENCES

- (1) Zhang, C.; McAdams, D.-A., II; Grunlan, J.-C. Nano/Micro-Manufacturing of Bioinspired Materials: a Review of Methods to Mimic Natural Structures. *Adv. Mater.* **2016**, *28*, 6292–6321.
- (2) Yu, Z.; Yun, F.-F.; Wang, Y.; Yao, L.; Dou, S.; Liu, K.; Jiang, L.; Wang, X. Desert Beetle-Inspired Superwetable Patterned Surfaces for Water Harvesting. *Small* **2017**, *13*, No. 1701403.
- (3) Huang, J.; Lai, Y.; Wang, L.; Li, S.; Ge, M.; Zhang, K.; Fuchs, H.; Chi, L. Controllable wettability and adhesion on bioinspired multifunctional TiO₂ nanostructure surfaces for liquid manipulation. *J. Mater. Chem. A* **2014**, *2*, 18531–18538.
- (4) Bico, J.; Tordeux, C.; Quere, D. Rough wetting. *Europhys. Lett.* **2001**, *55*, 214–220.
- (5) McHale, G.; Shirtcliffe, N.-J.; Aqil, S.; Perry, C.-C.; Newton, M.-I. Topography driven spreading. *Phys. Rev. Lett.* **2004**, *93*, No. 036102.
- (6) Hay, K.-M.; Dragila, M.-I.; Liburdy, J. Theoretical model for the wetting of a rough Surface. *J. Colloid Interface Sci.* **2008**, *325*, 472–477.
- (7) Yan, B.; Tao, J.; Pang, C.; Zheng, Z.; Shen, Z.; Huan, C.-H.; Yu, T. Reversible UV-Light-Induced Ultrahydrophobic-to-Ultrahydrophilic Transition in an α -Fe₂O₃ Nanoflakes Film. *Langmuir* **2008**, *24*, 10569–10571.
- (8) Liu, H.; Feng, L.; Zhai, J.; Jiang, L.; Zhu, D. Reversible Wettability of a Chemical Vapor Deposition Prepared ZnO Film between Superhydrophobicity and Superhydrophilicity. *Langmuir* **2004**, *20*, 5659–5661.
- (9) Kietzig, A.-M.; Hatzikiriakos, S.-J.; Englezos, P. Patterned Superhydrophobic Metallic Surfaces. *Langmuir* **2009**, *25*, 4821–4827.
- (10) Li, Z.; Zheng, Y.; Zhao, J.; Cui, L. Wettability of Atmospheric Plasma Sprayed Fe, Ni, Cr and Their Mixture Coating. *J. Therm. Spray Technol.* **2012**, *21*, 255–262.
- (11) Roach, P.; Shirtcliffe, N.-J.; Newton, M.-I. Progress in superhydrophobic surface Development. *Soft Matter* **2008**, *4*, 224.
- (12) Ngo, C.-V.; Liu, Y.; Li, W.; Yang, J.; Guo, C. Scalable Wettability Modification of Aluminum Surface through Single-Shot Nanosecond Laser Processing. *Nanomaterials* **2023**, *13*, 1392.
- (13) Moldovan, E.-R.; Doria, C.-C.; Ocaña, J.-L.; Baltes, L.-S.; Stanciu, E.-M.; Croitoru, C.; Pascu, A.; Roata, I.-C.; Terean, M.-H. Wettability and Surface Roughness Analysis of Laser Surface Texturing of AISI 430 Stainless Steel. *Materials* **2022**, *15*, 2955.
- (14) Liu, Z.; Niu, T.; Lei, Y.; Luo, Y. Metal surface wettability modification by nanosecond laser surface texturing: A review. *Biosurf. Biotribol.* **2022**, *8*, 95–120.
- (15) Samanta, A.; Wang, Q.; Shaw, S.-K.; Ding, H. Roles of chemistry modification for laser textured metal alloys to achieve extreme surface wetting behaviors. *Mater. Des.* **2020**, *192*, No. 108744.
- (16) Kim, V.-V.; Yalishev, V.-S.; Khan, S.-A.; Iqbal, M.; Boltaev, G.-S.; Ganeev, R.-A.; Alnaser, A.-S. Influence of gas environment on the dynamics of wetting transition of laser-textured stainless steel meshes. *AIP Adv.* **2021**, *11*, No. 075221.
- (17) Khan, S.-A.; Boltaev, G.; Iqbal, M.; Kim, V.; Ganeev, R.; Alnaser, A. Ultrafast fiber laser-induced fabrication of superhydrophobic and self-cleaning metal surfaces. *Appl. Surf. Sci.* **2021**, *542*, No. 148560.
- (18) He, Y.; Wang, L.; Wu, T.; Wu, Z.; Chen, Y.; Yin, K. Facile fabrication of hierarchical textures for substrate-independent and durable superhydrophobic surfaces. *Nanoscale* **2022**, *14*, 9392–9400.

- (19) Yin, K.; Wang, L.; Deng, Q.; Huang, Q.; Jiang, J.; Li, G.; He, J. Femtosecond Laser Thermal Accumulation-Triggered Micro-/Nanostructures with Patternable and Controllable Wettability Towards Liquid Manipulating. *Nano-Micro Lett.* **2022**, *14*, 97.
- (20) Isimjan, T.-T.; Wang, T.; Rohani, S. A novel method to prepare superhydrophobic, UV resistance and anti-corrosion steel surface. *Chem. Eng. J.* **2012**, *210*, 182.
- (21) Chen, F.; Zhang, D.; Yang, Q.; Yong, J.; Du, G.; Si, J.; Yun, F.; Hou, X. Bioinspired Wetting Surface via Laser Microfabrication. *ACS Appl. Mater. Interfaces* **2013**, *5*, 6777.
- (22) Khan, S.-A.; Lalyshv, V.; Kim, V.; Iqbal, M.; Harmi, H.; Boltaev, G.-S.; Ganeev, R.-A.; Alnaser, A.-S. Expedited Transition in the Wettability Response of Metal Meshes Structured by Femtosecond Laser Pulses for Oil-Water Separation. *Front Chem.* **2020**, *8*, 768.
- (23) Yalishev, V.; Iqbal, M.; Kim, V.; Alnaser, A.-S. Effect of processing environment on the wettability behavior of laser-processed titanium. *J. Phys. D: Appl. Phys.* **2022**, *55*, No. 045401.
- (24) Ren, N.; Li, J.-H.; Qiu, J.-C.; Sang, Y.-H.; Jiang, H.-D.; Boughton, R.-I.; Huang, L.; Huang, W.; Liu, H. Nanostructured titanate with different metal ions on the surface of metallic titanium: a facile approach for regulation of rBMSCs fate on titanium implants. *Small* **2014**, *10*, 3169–3180.
- (25) Kim, P.; Wong, T.-S.; Alvarenga, J.; Kreder, M.-J.; Adorno-Martinez, W.-E.; Aizenberg, J. Liquid-infused nanostructured surfaces with extreme anti-ice and anti-frost performance. *ACS Nano* **2012**, *6*, 6569–6577.
- (26) Lopez, A.-J.; Torres, B.; Taltavull, C.; Rams, J. Influence of high velocity oxygen-fuel spraying parameters on the wear resistance of Al–SiC composite coatings deposited on ZE41A magnesium alloy. *Mater. Des.* **2013**, *43*, 144–152.
- (27) Kostal, E.; Stroj, S.; Kasemann, S.; Matylitsky, V.; Domke, M. Fabrication of biomimetic fog-collecting superhydrophilic-superhydrophobic surface micropatterns using femtosecond lasers. *Langmuir* **2018**, *34*, 2933–2941.
- (28) Ren, F.; Li, G.; Zhang, Z.; Zhang, X.; Fan, H.; Zhou, C.; Wang, Y.; Zhang, Y.; Wang, C.; Mu, K.; Su, Y.; Wu, D. A single-layer Janus membrane with dual gradient conical micropore arrays for self-driving fog collection. *J. Mater. Chem. A* **2017**, *5*, 18403–18408.
- (29) Liu, W.; Fan, P.; Cai, M.; Luo, X.; Chen, C.; Pan, R.; Zhang, H.; Zhong, M. An integrative bioinspired venation network with ultra-contrasting wettability for large-scale strongly self-driven and efficient water collection. *Nanoscale* **2019**, *11*, 8940–8949.
- (30) Lu, L.; Ngo, C.-V.; Singh, S.; Yang, J.; Xin, W.; Yu, Z.; Guo, C. Bioinspired hierarchical surfaces fabricated by femtosecond laser and hydrothermal method for water harvesting. *Langmuir* **2019**, *35*, 3562–3567.
- (31) Yang, J.; Li, H.-N.; Chen, Z.-X.; He, A.; Zhong, Q.-Z.; Xu, Z.-K. Janus membranes with controllable asymmetric configurations for highly efficient separation of oil-in-water emulsions. *J. Mater. Chem. A* **2019**, *7*, 7907–7917.
- (32) Jagdheesh, R.; Diaz, M.; Marimuthu, S.; Ocana, J.-L. Robust fabrication of μ -patterns with tunable and durable wetting properties: hydrophilic to ultrahydrophobic via a vacuum process. *J. Mater. Chem. A* **2017**, *5*, 7125.
- (33) Tian, Y.; Jiang, L. Intrinsically robust hydrophobicity. *Nat. Mater.* **2013**, *12*, 291–292.
- (34) Pou, P.; del Val, J.; Riveiro, A.; Comesaña, R.; Arias-González, F.; Lusquiños, F.; Bountinguiza, M.; Quintero, F.; Pou, J. Laser texturing of stainless steel under different processing atmospheres: From superhydrophilic to superhydrophobic surfaces. *Appl. Surf. Sci.* **2019**, *475*, 896–905.
- (35) Yang, Z.; Liu, X.; Tian, Y. Insights into the wettability transition of nanosecond laser ablated surface under ambient air exposure. *J. Colloid Interface Sci.* **2019**, *533*, 268–277.
- (36) Li, B.; Li, H.; Huang, L.; Ren, N.; Kongda, X. Femtosecond pulsed laser textured titanium surfaces with stable superhydrophilicity and superhydrophobicity. *Appl. Surf. Sci.* **2016**, *389*, 585–593.
- (37) Guan, Y.-C.; Luo, F.-F.; Lim, G.-C.; Hong, M.-H.; Zheng, H.-Y.; Qi, B. Fabrication of metallic surfaces with long-term superhydrophilic property using one-stop laser method. *Mater. Des.* **2015**, *78*, 19–24.
- (38) Accogli, A.; Gibertini, E.; Panzeri, G.; Lucotti, A.; Magagnin, L. Understanding the Failure Mode of Electroless Nickel Immersion Gold Process: In Situ-Raman Spectroscopy and Electrochemical Characterization. *J. Electrochem. Soc.* **2020**, *167*, No. 082507.
- (39) Yalishev, V.-S.; Iqbal, M.; Kim, V.-V.; Khan, S.-A.; Ganeev, R.-A.; Alnaser, A.-S. Reversible wettability transition of laser-textured metals after vacuum storing and low-temperature annealing. *Appl. Phys. A* **2021**, *127*, 393.
- (40) Yalishev, V.-S.; Abbasi, N.-S.; Iqbal, M.; Alnaser, A.-S. Comparing Water Transport Properties of Janus Membranes Fabricated from Copper Mesh and Foam Using a Femtosecond Laser. *Langmuir* **2023**, *39*, 1815–1825.
- (41) Tian, X.; Li, J.; Wang, X. Anisotropic liquid penetration arising from a cross-sectional wettability gradient. *Soft Matter* **2012**, *8*, 2633.