Fabrication of Antireflection-structured Film by Ultraviolet Nanoimprint Lithography and its Mold Lifetime Amelioration

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ABSTRACT

Antireflection (AR) nanostructures from biomimetic moth-eye structures which can eliminate the undesirable reflection and increase the light transmission on the film surface have various applications in micro-nano discipline and nanophotonics fields. The significant applications of AR nanostructures ranging from improving the visibility of flat panel displays (FPDs), enhancing the performance of solar cells, enhancing the optical data storage, ameliorating light extraction in LEDs, and increasing the performance of optical lenses. Bernhard first discovered the function of antireflection of moth-eye structures in the nanophotonics fields in 1967. Several methods are available for fabricating AR nanostructures, for instance, interface holographic lithography, electron-beam lithography, nanosphere lithography, anodic oxidation porous alumina and so on. Nevertheless, these methods involve expensive and sophisticated apparatus when it comes to large scale fabrication of AR film.

Therefore, ultraviolet nanoimprint lithography or known as UV-NIL is a promising technique to fabricate AR films with excellent properties in large scale and with cost effective fabrication. The merits of using UV-NIL technique are its simple and room-temperature process, high-throughput, rapid and low-pressure process, and high-accuracy replication of pattern. One of the major obstacles for fabricating an AR film is to eliminate the reflection of interface that disrupts the optical performance of fabricated AR film. Interface reflection commonly occurs between the film substrate and the AR structures layer of fabricated AR film, which results in degradation of AR effect. Our laboratory i.e. Taniguchi *et al.* previously succeeded to fabricate a self-supporting AR-structured film which can reduce the reflection of interface to as low as 0.5% from UV-curable resin by UV-NIL. In this method, an intermediator film from film type of polyvinyl alcohol (PVA) material was employed to acquire a layer of self-supporting antireflection-structured film. However, the intermediator film that was used to support the replicated AR structure was from a low durability material. Due to the water-soluble synthetic polymer material of PVA film, a few regions from the replicated AR-structured layer were deteriorated and creases appeared during the

dissolving process of PVA film. This phenomenon affects the performance of fabricated AR film. The improvement in AR film method fabrication is highly needed. The industry goal of AR film fabrication is to reduce the reflectivity as low as 0.1%.

Therefore, to enhance the performance of fabricated AR film, the film type from polypropylene (PP) material is proposed to replace the intermediator film from PVA. This film gives better stability and better releasability during UV-NIL in concerning to get a single layer of self-supporting film with an excellent AR property. As a result, we successfully improved the fabricated self-supporting AR-structured film by reducing the reflectivity to as low as 0.3% and allows 94 ± 0.5% of light transmission. The advantages of the fabricated AR film; it is flexible, disposable, simple process, and time-effectiveness. Then, an adhesive material is required in order to apply this replicated AR film to any surfaces. Nevertheless, reflection of interface still arises due to the difference in refractive index between the self-supporting AR-structured film and the adhesive material. This phenomenon exhibit high reflectivity which affects the optical performance of fabricated AR film.

A study of film with AR property that capable to eliminate the interface reflection of the front and back surfaces is required. This type of AR film offers an excellent quasi-omnidirectional AR property on its film. Therefore, we proposed a double-sided self-supporting antireflection-structured; hereafter, DSARS film that fabricated by UV-NIL, as a promising solution. In this method, the DSARS film was fabricated by sandwiching the film between two different material of molds i.e., from glass carbon (GC) as master mold and the replica film mold of the master mold. As a result, we successfully further improved the optical performance of fabricated DSARS film by eliminating the reflectivity as minimum as 0.1% at the wavelength of visible light. The fabricated DSARS film also presents excellent transmission of light, which presents 97.00 ± 1.25% in the spectral range of wavelength. In this study, the determination of the commercial usability of fabricated DSARS film that specifically used in the surfaces of photovoltaic and LED was executed by demonstrating the application of fabricated DSARS film. The demonstration was executed by adhering the DSARS layer with adhesive material on top of the substrate of glass that had different refractive index. The merit of fabrication of DSARS film: it can be benefited in different shape of substrate. It also can effectively suppress the mismatch of the adhesive interface reflection that occurred during the multistacking of different refractive indices of film materials.

Due to the high demand of the industry in mass fabrication of AR film, the improvement of UV-NIL technique is necessary. Based on the principle of UV-NIL, the presence of release-coating-layer (RCL) on the AR mold is very crucial in order to impede the adhesion of resin during the mass fabrication of AR film. Nevertheless, one of the obstacles in mass fabrication of AR film is to extend the lifetime of RCL on the surface of AR mold. This issue arises due to the factors of chemical and mechanical that deteriorate the components of RCL from the surface gradually during the repetitive UV-NIL. According to the previous research reported by Takahashi *et al.*, force that resulting from the complete filling of the resin into the mold of high aspect ratio will generate the strong release force (RF). The resulted strong RF will possibly shorten the lifetime of the mold. In addition, the strong RF also resulted from the large surface area of the complete filling of resin. Osari *et al.* also reported that the difference in filling pressure results in different resin filling behavior in UV-NIL mold. This phenomenon can affect the durability of RCL on the UV-NIL mold. They also claimed that the presence of capillary force in the RCL mold allows the management of resin filling. Taking into account the concept of partial-filling of polymer that used by Bogdanski *et al.*, we considered that partial filling of resin will weaken the RF and reduce the aspect ratio of the fabricated AR film, eventually ameliorates the AR mold lifetime.

Thus, by employing the technique of partially filling the UV-curable resin during UV-NIL, the amelioration of the lifetime of AR mold was proposed. We ameliorated the lifetime of the mold of ARS that was made by GC mold and coated with RCL by partially filling technique of UV-NIL. Then, we evaluated the amelioration of the lifetime of AR mold by evaluating the repeatability of the AR mold to fabricate the AR film with low reflectivity. For comparison, we also evaluated the complete-filling technique of UV-NIL. By utilizing the technique of partial-filling UV-NIL, as a result, 0.25 ± 0.15% reflectivity and 94.0 ± 0.50% transmittance at the spectral range of wavelengths of replicated AR films were successfully obtained in average up to the 150th imprint. While, by employing the complete-filling UV-NIL, low reflectivity of replicated AR film was only obtained up to the 50th imprint.

Producing AR film using GC mold will allow only one sided fabrication due to the opacity of GC mold and GC is from brittle material. Besides, direct mass fabrication of DSARS film is not possible by using GC mold. It is necessary to reduce the dependency on GC mold and shift to the fabrication of AR film by using replica mold. It is also significant to produce the replica mold from

the material that can contribute resilience and flexibility, translucent, excellent releasability, and environmental-friendly. Thus, replicating the ARS film from replica mold was proposed. The proposed replica mold is from release-agent-free-antireflection-structured (RAF-ARS) type mold. This replica mold is made from UV-curable resin that consists of fluorinated components. This replica mold also offers antisticking effect, antifouling effect, and tested durability. Nevertheless, extending the lifetime of fluorinated components in the replica molds of RAF-ARS becomes an issue in their mass fabrication.

Therefore, by employing the same technique of partial-filling UV-NIL, we ameliorated the life-expectancy of RAF-ARS replica mold. For comparison, complete-filling technique was also evaluated. The investigation of the filling ratio effects on an RAF-ARS replica mold was also carried out. As a result, the lifetime of the RAF-ARS replica mold was successfully be prolonged up to the 100th imprint by employing the technique of partial-filling UV-NIL. In comparison, by employing the complete-filling UV-NIL technique, we can only fabricate up to the 75th imprint.

REFERENCES

[1] J.V. den B. Jan Haisma, Martin Verheijen, Kees van den Heuvel, J. Vac. Sci. Technol. B Microelectron. Nanom. Struct. **14**, (1996) 4124.

[2] A.M. Yasuhiko Tada, Hiroshi Yoshida, J. Photopolym. Sci. Technol. **4**, (2007) 545.

[3] D. Truffier-Boutry, M. Zelsmann, J. De Girolamo, J. Boussey, C. Lombard, B. Pépin-Donat, Appl. Phys. Lett. **94**, (2009) 044110.

[4] J. Takahashi, J. Taniguchi, Microelectron. Eng. **88**, (2011) 2141.

[5] J. Taniguchi, K. Machinaga, N. Unno, N. Sakai, Microelectron. Eng. **86**, (2009) 676.

[6] A. Mayer, K. Dhima, S. Möllenbeck, S. Wang, H.-C. Scheer, J. Sakamoto, H. Kawata, Y. Hirai, J. Vac. Sci. Technol. B Microelectron. Nanom. Struct. **30**, (2012) 06FB03.

[7] N. Bogdanski, M. Wissen, S. Möllenbeck, H.-C. Scheer, J. Vac. Sci. Technol. B Microelectron. Nanom. Struct. **24**, (2006) 2998.

[8] M. Otto, M. Bender, B. Hadam, B. Spangenberg, H. Kurz, Microelectron. Eng. **57**, (2001) 361.

[9] L.. Heyderman, H. Schift, C. David, J. Gobrecht, T. Schweizer, Microelectron. Eng. **54**, (2000) 229.

[10] R.N. Wenzel, Ind. Eng. Chem. **28**, (1936) 988.

[11] A.B.D.Cassie, S. Baxter, Trans. Faraday Soc. **40**, (1944) 546.

[12] E.E. Miller, R.D. Miller, J. Appl. Phys. **27**, (1956) 324.

[13] K. Osari, N. Unno, J. Taniguchi, K. Machinaga, T. Ohsaki, N. Sakai, Microelectron. Eng. **87**, (2010) 918.

[14] K. Machinaga, J. Taniguchi, N. Unno, N. Sakai, T. Ohsaki, Seikei-Kakou **21**, (2010) 346 [in Japanese].

[15] N. Unno, K. Osari, K. Machinaga, J. Taniguchi, T. Ohsaki, N. Sakai, J. Photopolym. Sci. Technol. **22**, (2009) 161.

[16] A. del Campo, E. Arzt, Chem. Rev. **108**, (2008) 911.

[17] W.K. Neo, M.B. Chan-Park, Macromol. Rapid Commun. **26**, (2005) 1008.

[18] J. Taniguchi, Y. Nemoto, Y. Sugiyama, J. Nanosci. Nanotechnol. **9**, (2009) 445.

[19] N.B. Abu Talip[a]Yusof, J. Taniguchi, Microelectron. Eng. **110**, (2013) 163.

[20] D. Truffier-Boutry, a. Beaurain, R. Galand, B. Pelissier, J. Boussey, M. Zelsmann, Microelectron. Eng. **87**, (2010) 122.

[21] J. Taniguchi, Y. Kamiya, T. Ohsaki, N. Sakai, J. Photopolym. Sci. Technol. **22**, (2009) 175.

[22] J. Taniguchi, Y. Kamiya, T. Ohsaki, N. Sakai, Microelectron. Eng. **87**, (2010) 859.

[23] P.B.Clapham, M.C.Hutley, Nature **244**, (1973) 281.

[24] D.J.Arrowsmith, Trans. Inst. Met. Finish. **48**, (1970) 88.

[25] K.X. Wang, Z. Yu, V. Liu, Y. Cui, S. Fan, Nano Lett. **12**, (2012) 1616.

[26] N.B. Abu Talip[a]Yusof, J. Taniguchi, Jpn. J. Appl. Phys. **53**, (2014) 06JK03.

[27] F.L. Gonzalez, M.J. Gordon, Opt. Express **22**, (2014) 12808.

[28] Z. Chen, Y. Gao, **53**, (2014) 3673.

[29] J.Y.Y.Loh, D.P.Puzzo, P.G. O'Brien, G. A. Ozin, N.P. Kherani, RSC Adv. **4**, (2014) 31188.

[30] K.-H. Tsui, Q. Lin, H. Chou, Q. Zhang, H. Fu, P. Qi, Z. Fan, Adv. Mater. **26**, (2014) 2805.

[31] Y. Li, F. Li, J. Zhang, C. Wang, S. Zhu, H. Yu, Z. Wang, B. Yang, Appl. Phys. Lett. 96, **43**, (2010) 255101.

[32] Y.-H. Ho, H. Liang, S.-W. Liu, W.-C. Tian, F.-C. Chen, P.-K. Wei, RSC Adv. **4**, (2014) 9588.

[33] P. Gao, M. Gu, X.L. Liu, B. Liu, S.M. Huang, Appl. Phys. Lett. **95**, (2009) 2013.

[34] Z. Zhu, B. Liu, C. Cheng, H. Chen, M. Gu, Y. Yi, R. Mao, Phys. Status Solidi **211**, (2014) 1583.

[35] X. Li, L. Xue, Y. Han, J. Mater. Chem. **21**, (2011) 5817.

[36] J. Cai, J. Ye, S. Chen, X. Zhao, D. Zhang, S. Chen, Y. Ma, S. Jin, L. Qi, Energy Environ. Sci. **5**, (2012) 7575.

[37] S. Ravipati, J. Shieh, F.-H. Ko, C.-C. Yu, H.-L. Chen, C.-T. Wu, S.-H. Chen, Energy Environ. Sci. **5**, (2012) 7601.

[38] M. Bender, M. Otto, B. Hadam, B. Spangenberg, H. Kurz, Microelectron. Eng. **61**, (2002) 407.

[39] S. Ahn, J. Cha, H. Myung, S.M. Kim, S. Kang, Appl. Phys. Lett. **89**, (2006) 9.

[40] S.H. Ahn, L.J. Guo, Adv. Mater. **20**, (2008) 2044.

[41] S.H. Ahn, L.J. Guo, ACS Nano **3**, (2009) 2304.

[42] M. Moro, J. Taniguchi, S. Hiwasa, J. Vac. Sci. Technol. B, Nanotechnol. Microelectron. Mater. Process. Meas. Phenom. **32**, (2014) 06FG09.

[43] J. Taniguchi, T. Kawasaki, Y. Tokano, Y. Kogo, I. Miyamoto, M. Komuro, H. Hiroshima, N. Sakai, K. Tada, Jpn. J. Appl. Phys. **41**, (2002) 4194.

[44] S. Garidel, M. Zelsmann, N. Chaix, P. Voisin, J. Boussey, A. Beaurain, B. Pelissier, J. Vac. Sci. Technol. B Microelectron. Nanom. Struct. **25**, (2007) 2430.

[45] F. A. Houle, C.T. Rettner, D.C. Miller, R. Sooriyakumaran, Appl. Phys. Lett. **90**, (2007) 213103.

[46] F. A. Houle, E. Guyer, D.C. Miller, R. Dauskardt, J. Vac. Sci. Technol. B Microelectron. Nanom. Struct. **25**, (2007) 1179.

[47] D. Yamashita, J. Taniguchi, H. Suzuki, Microelectron. Eng. **97**, (2012) 109.

[48] K. Yajima, K. Adachi, Y. Tsukahara, J. Taniguchi, Microelectron. Eng. **110**, (2013) 188.

[49] K.D. Kim, H.J. Kwon, D.G. Choi, J.H. Jeong, E.S. Lee, Jpn. J. Appl. Phys. **47**, (2008) 8648.

[50] M. Im, H. Im, J.-H. Lee, J.-B. Yoon, Y.-K. Choi, Soft Matter **6**, (2010) 1401.

[51] P. Glaris, J.F. Coulon, M. Dorget, F. Poncin-Epaillard, Compos. Part B Eng. **73**, (2015) 10.

[52] N. Nakamura, T. Yamashita, T. Kitagawa, H. Kawata, M. Shirai, Y. Hirai, J. Photopolym. Sci. Technol. **27**, (2014) 111.

[53] K. Wu, X. Wang, E.K. Kim, C.G. Willson, J.G. Ekerdt, Langmuir **23**, (2007) 1166.

[54] Y. Otsuka, S. Hiwasa, J. Taniguchi, Microelectron. Eng. **123**, (2014) 192.

[55] K. Sogo, M. Nakajima, Y. Hirai, J. Photopolym. Sci. technol. **19**, (2006) 647.

[56] T. Yanagishita, T. Kondo, K. Nishio, H. Masuda, J. Vac. Sci. Technol. B Microelectron. Nanom. Struct. **26**, (2008) 1856.

LIST OF PUBLICATIONS

A. Accepted/Published papers

1. N. B. Abu Talip[a]Yusof, T. Hayashi, J. Taniguchi, and S. Hiwasa, "Lifetime prolongation of release agent on antireflection structure molds by means of partial-filling ultraviolet nanoimprint lithography." 2015 International Conference on Electronic Packaging and iMAPS All Asia Conference (ICEP-IACC), IEEE pp. 418-421 (2015).
2. N. B. Abu Talip[a]Yusof, T. Hayashi, J. Taniguchi, and S. Hiwasa, "Lifetime amelioration of release-agent-free antireflection-structured replica molds by partial-filling ultraviolet nanoimprint lithography", Jpn. J. Appl. Physc. **54**, (2015) 06FM04.
3. N. B. Abu Talip[a]Yusof, T. Hayashi, J. Taniguchi, and S. Hiwasa, "Lifetime amelioration of antireflection structure molds by means of partial-filling ultraviolet nanoimprint lithography", Microelectron. Eng. **141**, (2015) 81.
4. N. B. Abu Talip[a]Yusof and J. Taniguchi, "Fabrication of double-sided self-supporting antireflection-structured film by ultraviolet nanoimprint lithography", Jpn. J. Appl. Physc. **53**, (2014) 06JK03.
5. N. B. Abu Talip[a]Yusof and J. Taniguchi, "Fabrication of self-supporting antireflection-structured film by UV-NIL", Microelectron. Eng. **110**, (2013) 163.

B. Conference oral presentation

1. N. B. Abu Talip[a]Yusof, T. Hayashi, J. Taniguchi, and S. Hiwasa, "Lifetime prolongation of release agent on antireflection structure molds by means of partial-filling ultraviolet nanoimprint lithography", 2015 International Conference on Electronic Packaging & iMAPS All Asia Conference, Kyoto Terrsa, Kyoto, Japan. (14/04/2015-17/04/2015)

C. Conference poster presentation

1. N. B. Abu Talip[a]Yusof, T. Hayashi, J. Taniguchi, and S. Hiwasa, "Lifetime amelioration of release-agent-free antireflection-structured replica molds by partial-filling ultraviolet

nanoimprint lithography", 27th International Microprocesses and Nanotechnology Conference, Hilton Fukuoka Sea Hawk, Fukuoka, Japan. (04/11/2014-07/11/2014)

2. N. B. Abu Talip[a]Yusof, T. Hayashi, J. Taniguchi, and S. Hiwasa, "Lifetime amelioration of antireflection structure molds by means of partial-filling ultraviolet nanoimprint lithography", 40th International Conference on Micro and Nano Engineering, Lausanne, Switzerland. (22/09/2014-26/09/2014)
3. N. B. Abu Talip[a]Yusof and J. Taniguchi, "Fabrication of double-sided self-supporting antireflection-structured film by ultraviolet nanoimprint lithography", 26th International Microprocesses and Nanotechnology Conference, Royton Sapporo, Hokkaido, Japan. (05/11/2013-08/11/2013)
4. N. B. Abu Talip[a]Yusof and J. Taniguchi, "Fabrication of self-supporting antireflection-structured film by UV-NIL", 38th International Conference on Micro and Nano Engineering, Toulouse, France. (16/09/2012-20/09/2012)