Polyvinyl fluoride (PVF); Its Properties, Applications, and Manufacturing Prospects

M. H. Alaaeddin1*, S. M. Sapuan1*, M.Y.M Zuhri1, E.S Zainudin1,2, Faris M. AL-Oqla3

1Department of Mechanical and Manufacturing Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia.
2Laboratory of Biocomposite Technology, Institute of Tropical Forestry and Forest Products, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
3Department of Mechanical Engineering, Faculty of Engineering, The Hashemite University, 13133 Zarqa, Jordan

E-mails: m.h.alaaeddin@gmail.com; sapuan@upm.edu.my +6 01139395070

Abstract. Polyvinyl fluoride (PVF) is a distinctive polymer with excessively studied properties. The wide utilization of this polymer has dramatically escalated due to its outstanding properties (e.g., mechanical strength, chemical resistance, high stability, low permeability, indoor functionality, outdoor durability, light transmissibility, adherence, stain resistance, sunlight degradation, and high temperature endurance). An overview of the chemical, physical, electrical, and thermal properties of PVF is provided. The current applications of PVF/Tedlar®, its polymerization, and the manufacturing processes are also addressed in this work. A comparative analysis has been conducted to explore common and uncommon properties of PVF, fluoropolymers, and competing polymers such as polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE), and polyvinyl chloride (PVC). Overall, PVF is anticipated to have further promising manufacturing prospects in numerous industries and in various fields such as in medical equipment, renewable energy applications, sensitive devices, advanced industrial platforms, and nanotechnology material composites. Therefore, further in-depth analyses and oriented numerical information on the performance of this polymer are required.

1. Introduction
Polyvinyl fluoride is considered to be an exclusive polymer or special plastic employed in numerous industries and manufacturing processes since 1962. It is therefore an important type of polymer due to its excellent properties and high performance; e.g. chemical resistance, stain resistance, outdoor durability, high stability, and adherence [1]. In recent years, different studies have been conducted to scrutinize the wide range of properties of PVF. It is considered as an ideal material with unique electrical, chemical, and mechanical properties [2]. PVF is mainly designed for surface protection due to its toughness, lightness, and high resistivity to moisture, weathering, and ultraviolet rays. It is also inert to chemicals, staining agents, and solvents [1-3]. The functional properties of PVF were investigated in numerous studies which had asserted unswerving facts on its conductivity odds and ferroelectricity. These attributes continuously reveal vital characterizations for the enthalpies and dielectric constant measurements of this polymer and its composites [4]. PVF is applied in a form of films under multiple names: i.e., Teslar®, Tedlar®, and R films. However, PVF films or Tedlar® are the two most common names for polyvinyl fluoride films [1,5]. The high resistivity and excellent properties of PVF (such as resistance to sunlight degradation and solvents, chemical inertness, high
tensile strength, toughness, and dimensional stability) makes it competent for specific functions [3,6]. Initially, PVF films were mainly used for very specific applications such as in PV encapsulation processes, pigmented films for house sidings, and aircraft interiors. This then expanded to printed circuit boards, industrial sidings, roofing films, clear films for skylights with ultraviolet absorber sets, and similar applications [5-7].

2. PVF Structure and Properties

As a fluopolymer, polyvinyl fluoride (PVF) is a thermoplastic polymer with a chemical formula of (C2H3F)n. The polymer consists of a repeating VF unit and is similar in structure to other compounds (e.g. polyvinylidene fluoride PVDF, polyvinyl chloride PVC, polyethylene PE, and polytetrafluoroethylene PTFE) [8,9]. Fluoropolymers are considered to be homopolymers and copolymers; the fluorinated fluoropolymers contain carbon (C), fluorine (F), hydrogen (H), or other atoms such as Chlorine (Cl) and vinyl fluoride (VF). H2C=F undergoes free-radical polymerization leading to C–F and C–H bonds since F is usually the highest electronegative from all elements; it is easy to convert to F−. Likewise, the C–F bond strength is higher than the C–H [10,11]. H is smaller than F and F is smaller than Cl [10]. The distinctive structure of PVF justifies the reason behind its good properties, making it an ideal release for fiber reinforced polymers (FRP). It has the ability to meet the expected long range needs for composites, manufacturing operations, and other specific application requirements [10,11]. Fig. 1 provides the ball-and-stick model for the PVF, PVDF, PTFE, PE, and PVC chemical structures.

![Figure 1. Ball-and-stick model for the structure of referred polymers](image)

PVF properties vary according to manufacturing purposes, but they remain close to original properties; e.g. the commercial one is atactic and the melting point peaks at 190°C. Several transitions are anticipated below the melting temperature in PVF. The polymer is semicrystalline with varied degrees of crystallinity (from 20% to 60%) and a planar zigzag conformation. The solubility in all solvents below 100°C is very low and inert to chemical treatments. PVF is considered to be visible, near-infrared regions, and transparent to radiation in UV [12].

3. Polymerization and Manufacturing of VF

Both the polymerization process and thermal performance in VF significantly contribute to the degree of crystallinity in this polymer. The existing relationship between the polymer structure and the degree of crystallinity and melting point justifies the reason behind the variations found in the melting points, affirming that the head-to-head linkage affects the degree of crystallinity, ranging from 20% to 60%; whereas a 12% head-to-head linkage is anticipated in commercial PVF with an estimated melting point of 190°C [12]. In the polymerization procedure, there are various methods and techniques used to process VF; e.g. suspension polymerization, bulk polymerization, emulsion polymerization, radiation-induced polymerization, and graft polymerization [11,13]. VF is subjected to free-radical polymerization where the thermal initiator is the starter of the polymerization reaction, followed by propagation and termination. With a wide variety of initiators and different polymerization methods, benzoyl peroxide is used to initiate the polymerization process to produce oriented PVF films [13,14]. The initial polymerization includes a saturated solution of VF heated in toluene at 67°C under 600 MPa for 16 hours [12,13]. A combination of oxygen and trisobutylborane can be used to polymerize VF at
less pressure and temperature. The temperature in the polymerization process is anticipated to range between 0 to 85°C. The melting point notoriously drops from 230°C at 0°C polymerization to 200°C at 85°C polymerization, considering the degree of crystallinity and melting temperature. This occurs due to existing divergences of monomer repercussions during the polymerization process. VF volatility insists on interfering in moderate high pressure in the polymerization process; whereby, free-radical initiators are incarnated by the photopolymerization process [13]. However, it is worth mentioning that the copolymers and terpolymers of VF can be prepared with a wide range of monomers; in the last few years, the interpolymers of VF have been classified as highly fluorinated monomers (e.g. perfluoroethylvinylether, hexafluoropropylene, and perfluorobutylethylene) [7,13]. As a result, the yielded polyvinyl fluoride demonstrates high softening temperatures and finite solubility, making it a unique polymer with special characteristics. This opened a major market for its application, leading to the development of the polymerization process and the designed commercialized polymerization of VF.

The table below provides information on the different types of polymerizations and techniques [12,15].

| Polymerization | Technique | Adjutant | Initiator |
|----------------|-----------|----------|-----------|
| Suspension polymerization | VF is suspended in water | Dispersion stabilizer | Organic peroxide |
| Bulk polymerization | Irradiation with UV | Di-tert-butyl peroxide | |
| Emulsion polymerization | Emulsion method (highly reduced pressure and lower temperature) | Emulsifiers | Aqueous emulsion |
| Radiation-Induced polymerization | Radiation (decomposes into free radical) | Gas-phase | Benzyol, lauroyl |
| Graft polymerization | Radiation polymerization | Grafting with monomers | γ-rays |

In the manufacturing process of PVF, converting vinyl fluoride (VF) into polyvinyl fluoride (PVF) is the main process and intended result. At the beginning of the process, VF must be stored in high-pressure storage tanks to be later pumped into a reactor unit. The reacting function should be performed to create PVF slurry in water with unreacted VF. The water then flows into separators to remove the VF. The separators pump the VF using compressors and return it back to the reactor. The procedure is followed by a steam injection process to raise the temperature of PVF and evaporate what remains from VF in the slurry. Subsequently, the heated admixture flows into a small slurry flash tank and there, the residual VF must be released into the atmosphere. The incombustible PVF slurry then flows into a designed insulated tank and out of three tanks for slurry holding. The designed functions of these three tanks form an integrative complementary method; if one reaches a high level, the hot slurry flows into the other. The seal loop is designed in the second slurry tank in order to prevent VF from evaporating as steam when passing from the flash tank into the slurry tank. Once the process is complete, the PVF slurry is impelled from the slurry tanks to a PVF film manufacturing area to produce Tedlar® and undergo further processing [13].

4. PVF Applications and Composite Films

Polyvinyl fluoride offers such tremendous advantages for several applications which can be used in various modern fields and manufacturing platforms. It is an idealistic film for fiber-reinforced polymer (FRP) and carbon-fiber-reinforced polymer (CFRP). It sufficiently works to protect materials under certain conditions and performs well under high pressures and temperatures [16,17]. PVF is used as an encapsulant for the front and back cover of photovoltaic applications and is embedded within the assembly process of solar panels. In 1960s, PVF films were first employed in aircraft interiors, cabin parts, and electronic industries [17,18]. With good capability to withstand various temperatures (up to 204°C), PVF films are useful for numerous aircraft interior parts and are highly recommended for hand lay-up and vacuum bagging, as well as for other related applications. They are available in different colors and forms; whether adhesive materials that function on one side or other bonding materials for laminates. PVF eliminates the production cost and reduces the need for additional processing and other
finishing materials due to its durability and favorable appearance. It is also available in the forms of oriented and unoriented films; the oriented films are made using an extrusion method of Polyvinyl fluoride. In the extrusion process, PVF must be dispersed in a latent solvent and trailed by both transverse directions and biaxial orientations to enhance the mechanical properties of the anticipated films. As for unoriented films, they are more compliant and formable than the oriented ones. Unoriented films are slightly stretched after being cast onto a carrier web. They have a higher elongation at break with lower tensile strength. Nonetheless, PVF adhesives have a wide range of applications which can be applied for coating and lamination substrates (e.g. vinyl fabric, aluminum, galvanized steel, thermoplastic, vinyl wall covering, and cellulosic substrates). The coated polyvinyl fluoride is also useful for different functioning materials in metals, wood, and plastics. It works as a reliable laminated film for substrates. The excellent properties and high specifications of PVF makes it highly recommended for critical applications; this spurred its use in indoor and outdoor applications, either for decorative purposes, protective, or both. In the PV manufacturing process, PVF composite films represent the main materials for the encapsulation process, which must be applied to ensure the safety and reliability of PV modules for long-term function and lasting efficiency [12-15,17,19].

5. PVF and Competing Polymers

Polymers differ from each other through small or large variances that affect their structure. Some polymers slightly differ in their composition or chemical structure from polyvinyl fluoride (e.g. polyvinylidene fluoride, polytetrafluoroethylene, and polyvinyl chloride (not crystalline), while other polymers from the fluoropolymers family consist of semicrystalline materials or homopolymers. These polymers share special characteristics with polyvinyl fluoride in some chemical, mechanical, thermal, or physical properties. They may have independencies or share some basic assumptions in their piezoelectric and pyroelectric properties [20-22].

5.1. Polyvinylidene Fluoride (PVDF)

PVDF is one of the important pyroelectric polymers that could contribute to the concept of heat-generated electricity with a wide range of implementations; from industrial to biomedical utilizations. The pyroelectricity of PVDF can be enhanced using a certain inclusion that is accurately balanced after determining its properties; however, not all additives or inclusions can enhance the pyroelectricity of PVDF. The enhancement process can be increased by the intension of pyroelectric coefficient due to the addition of nonferroelectric inclusion under certain conditions, with the observation of the electric dipole moment [23]. PVDF shares competing characteristics in weather resistance. It does improve the coating quality and adhesion, and it can also be used for the preparation of superhydrophobic membranes and coatings. Besides the high heat resistance properties, PVDF possesses superior mechanical and chemical features which justify its extensive use in the preparation of micro and ultrafiltration membranes [24,25]. PVDF is further used in the fabricating process of electrospun nanofibrous microfiltration-membranes (ENMs), and is a suitable candidate for downstream bioprocessing, bioseparation and biomedical applications. It is a reliable polymer for pH-responsive membranes, water treatment, and purification systems. Nevertheless, the use of this polymer can vary in different directions and applications [26]. Its favorable characteristics such as its mechanical properties, chemical stabilities, high dielectric permittivity, and weatherability represent a key role in determining the methods that can be employed in various optimization conditions for each process. The crystals of PVDF consist of five different polymorphs which come in the form of α, β, γ, δ, and ε [27,28]. However, the crystallization of altered PVDF polymorphs is essential in order to analyze the anticipated changes in its properties upon each process, despite the degree of crystallinity (DOC) or the molecular chain orientation (MCO) which can be evaluated differently [29].

5.2. Polytetrafluoroethylene (PTFE)

Teflon® or PTFE was first discovered in 1938 [11]. It is a fluoropolymer, the fluorocarbon-based chemical formula of (CF2-CF2)n, as illustrated in Fig. 1. The polymer has low or anti-friction
properties, naturally hydrophobic, and non-biodegradable. It is easy to fabricate into a wide range of forms for various applications and industries (e.g. in the healthcare industry, it is used in the fabrication of nerve regeneration tubes, vascular prostheses, maxillofacial surgeries, subcutaneous augmentation materials, and other applications) [30,31]. Furthermore, PTFE is well-known for its good properties and considered as an efficient internal lubricant. The processed and modified features produce a friction film between sliding components to reduce adhesion, whereby the composite is normally treated in a melt mixing process [32]. PTFE thin films with high molecular weight, high melting point of up to 327°C, stability of up to 260°C, elevated chemical and biological inertness, and increased resistance to corrosive solvents and concentrated acids have promising benefits and competitive properties that could be employed in distinct applications [33]. It is one of the most important polymers with a wide range of extensive applications in different sciences and engineering fields. In particular, PTFE with sharp reversible transition at 600 K, changing from the original white substance to a transparent one [34], has a decreased coefficient of friction (CoF). Its low CoF ranges between (0.05-0.08, static) [35]. The C–F bond in PTFE is the strongest bond; it provides the polymer with steady chemical, physical, and thermal properties [11]. These properties lead to the use of PTFE in several important applications for reactive and corrosive chemicals. To alter the surface of PTFE without changing the properties of bulk, the plasma modification technique or the NH3 and H2 plasma treatment is recommended for several reasons: it is considerably easier to control compared to other complex and hazardous techniques, it has a lower processing cost compared to the electron beam and other similar techniques, and more importantly, it is environmentally friendly with a biofunctionalization treatment of the surface. It has much less impact on the environment compared to other techniques [35].

5.3. Polyvinyl Chloride (PVC)
Polyvinyl chloride (PVC) is anticipated to be among the most widely utilized thermoplastic polymer in the world [36]. It displays acceptable mechanical properties, adequate chemical resistance, outstanding process ability, and thermal stability with vast and diversified manufacturing tasks and applications [37]. It was asserted that PVC properties significantly contribute to numerous industrial applications and a wide range of composites. However, there is a need to improve these properties (the mechanical ones in particular). The polymer has shown a positive response to recent improvements accomplished; for example, PVC’s mechanical properties as a function of graphene oxide loading increase both the Young’s modulus and tensile strength. [38]. In addition, PVC membrane properties improved by the introduction of amino groups, further providing active chemical sites to its surface. In composite materials (CMs), PVC has a strong relationship with different types of materials and natural fiber composites (NFCs). It is a highly recommended polymer for reinforcement and blending processes; it shows adequate portability and high compactness when integrated with other materials. However, due to the massive production of PVC and increasing methods of fabrication, widespread debate has been raised to find alternatives in which a reduction in the use of this non-biodegradable polymer can occur [39]. Currently, nanotechnology studies are focusing on the electric and dielectric characterizations of PVC which have reflected on industrial practices, nanocomposite materials, and polymer nanocomposites such as: PVC/CdSE, PVC/ZnS, and PVC/PVDF/ZnO. This opens further prospects for this polymer in nanocomposite technologies [40].

6. Conclusions
PVF has distinguished properties and exceptional performance under harsh conditions. PVF can last in high temperatures and in volatile weather conditions with excellent resistance to sunlight degradation and high solar transmittance rate. In addition, the chemically inert polymer is highly effective in resisting solvents, chemicals, corrosion, and water absorption. Therefore, these important properties have designated this polymer to be utilized in numerous industrial applications; e.g. indoor and outdoor coatings, encapsulation processes, as well as decorative and functional uses. The polymer is meant to be applied where chemical resistance, mechanical strength, strain resistance, release properties, outdoor durability, thermal stability, low permeability, and adherence are required. Despite the complex
polymerization process of PVF, the polymer is anticipated to have a promising future in many other industries and in various fields such as in medical equipment, energy devices, advanced educational platforms, and nanotechnology composites.

Acknowledgments
Acknowledgment goes to the Ministry of Higher Education-Malaysia for the research grant HiCoE project vote number 6369107 and University Putra Malaysia for facilitating the necessary means.

References
[1] S. Ebnesajjad, Introduction to fluoropolymers: materials, technology and applications. William Andrew, 2013.
[2] I. Kaur, V. Kumari, B. Sharma, and N. Gupta, "Characterization and applications of PVF film grafted with binary mixture of methacrylic acid and 4-vinyl pyridine by gamma radiations: Effect of swift heavy ions," Appl. Radiat. Isot., vol. 79, pp. 118-130, 2013.
[3] A. Gupta, R. Bajpai, and J. Keller, "Morphological, crystallization and plasticization studies on isomorphic blends of poly (vinyl fluoride)(PVF) and poly (vinylimide difluoride)(PVDF): using microhardness, XRD and SEM techniques," J. Mater. Sci., vol. 41, no. 18, pp. 5857-5861, 2006.
[4] N. Kumar and R. Nath, "Thermal and ferroelectric properties of potassium nitrate: polyvinyl fluoride composite films," IEEE Trans. Dielectr. Electr. Insul., vol. 12, no. 6, pp. 1145-1150, 2005.
[5] G. Oreski and G. Wallner, "Delamination behaviour of multi-layer films for PV encapsulation," Sol. Energy Mater Sol. Cells, vol. 89, no. 2-3, pp. 139-151, 2005.
[6] V. Simril and B. A. Curry, "The properties of polyvinyl fluoride film," J. Appl. Polym. Sci., vol. 4, no. 10, pp. 62-68, 1960.
[7] S. Ebnesajjad, Fluoroplastics, Volume 2: Melt Processible Fluoropolymers-The Definitive User's Guide and Data Book. William Andrew, 2015.
[8] A. Shu et al., "Cratering studies in Polyvinylidene Fluoride (PVDF) thin films," Planet. Space Sci., vol. 89, pp. 29-35, 2013.
[9] C. L. Yaws, Thermophysical properties of chemicals and hydrocarbons. William Andrew, 2008.
[10] J. E. House and K. A. House, Descriptive inorganic chemistry. Academic Press, 2015.
[11] J. Gardiner, "Fluoropolymers: origin, production, and industrial and commercial applications," Aust. J. Chem., vol. 68, no. 1, pp. 13-22, 2015.
[12] S. Ebnesajjad, "Introduction to fluropolymers," in Applied Plastics Engineering Handbook: Elsevier, 2017, pp. 55-71.
[13] S. Ebnesajjad, Polyvinyl fluoride: Technology and applications of PVF. William Andrew, 2012.
[14] S. Ebnesajjad and A. H. Landrock, Adhesives technology handbook. William Andrew, 2014.
[15] T. Nguyen, "Degradation of poly [vinyl fluoride] and poly [vinylidene fluoride]," Polym. Rev., vol. 25, no. 2, pp. 227-275, 1985.
[16] V. Karbhari, "Rehabilitation of metallic civil infrastructure using fiber-reinforced polymer (FRP) composites: a materials and systems overview at the adhesive bond level," in Rehabilitation of Metallic Civil Infrastructure Using Fiber Reinforced Polymer (FRP) Composites: Elsevier, 2014, pp. 3-10.
[17] M. Alaaeddin, S. Sapuan, M. Zuhri, E. Zainudin, and F. M. Al-Oqla, "Properties and Common Industrial Applications of Polyvinyl fluoride (PVF) and Polyvinylidene fluoride (PVDF)," in IOP Conf. Ser.: Mater. Sci. Eng., 2018, vol. 409, no. 1, p. 012021: IOP Publishing.
[18] J. G. Drobny, "Fluorine-containing polymers," in Brydson's Plastics Materials: Elsevier, 2017, pp. 389-425.
[19] M. DeBergalis, "Fluoropolymer films in the photovoltaic industry," J. Fluor. Chem., vol. 125, no. 8, pp. 1255-1257, 2004.
[20] M. Biron, Thermoplastics and thermoplastic composites. William Andrew, 2018.
[21] J. G. Drobny, Technology of fluoropolymers. CRC Press, 2008.

[22] D. Ghazanfari, D. Bastani, and S. A. Mousavi, "Preparation and characterization of poly (vinyl chloride)(PVC) based membrane for wastewater treatment," J. Water Process Eng., vol. 16, pp. 98-107, 2017.

[23] K. Al Abdullah et al., "The enhancement of PVDF pyroelectricity (pyroelectric coefficient and dipole moment) by inclusions," Energy Procedia, vol. 119, pp. 545-555, 2017.

[24] J. E. Dohany, "Fluorine-containing polymers, poly (vinylidene fluoride)," Kirk-Othmer Encycl. Chem. Technol., 2000.

[25] J. T. Goldbach et al., "The enhancement of PVDF pyroelectricity (pyroelectric coefficient and dipole moment) by inclusions," Energy Procedia, vol. 119, pp. 545-555, 2017.

[26] J. E. Dohany, "Fluorine-containing polymers, poly (vinylidene fluoride)," Kirk-Othmer Encycl. Chem. Technol., 2000.

[27] J. T. Goldbach et al., "Commercial synthesis and applications of poly (vinylidene fluoride)," Fluorinated Polymers Vol. 2: Applications, 2016.

[28] F. Liu, N. A. Hashim, Y. Liu, M. M. Abed, and K. Li, "Progress in the production and modification of PVDF membranes," J. Membr. Sci., vol. 375, no. 1-2, pp. 1-2, 2011.

[29] W. Ma, J. Zhang, S. Chen, and X. Wang, "β-Phase of poly (vinylidene fluoride) formation in poly (vinylidene fluoride)/poly (methyl methacrylate) blend from solutions," Appl. Surf. Sci., vol. 254, no. 17, pp. 5635-5642, 2008.

[30] J. Wang, J. Wu, W. Xu, Q. Zhang, and Q. Fu, "Preparation of poly (vinylidene fluoride) films with excellent electric property, improved dielectric property and dominant polar crystalline forms by adding a quaternary phosphorus salt functionalized graphene," Compos. Sci. Technol., vol. 91, pp. 1-7, 2014.

[31] Y. J. Park et al., "Molecular and crystalline microstructure of ferroelectric poly (vinylidene fluoride-co-trifluoroethylene) ultrathin films on bare and self-assembled monolayer-modified Au substrates," Macromolecules, vol. 41, no. 1, pp. 109-119, 2008.

[32] I. U. Ahad et al., "Extreme ultraviolet (EUV) surface modification of polytetrafluoroethylene (PTFE) for control of biocompatibility," Nucl. Instrum. Methods Phys. Res., Sect. B., vol. 364, pp. 98-107, 2015.

[33] V. Rudnev, A. Vaganov-Vil'kins, T. Yanovsky, and A. Pavlov, "Polytetrafluoroethylene-oxide coatings on aluminum alloys," Surf. Coat. Technol., vol. 307, pp. 1249-1254, 2016.

[34] H. Hunke et al., "Plasma modified Polytetrafluoroethylene (PTFE) lubrication of α-olefin-copolymer impact-modified Polyamide 66," Wear, vol. 338, pp. 122-132, 2015.

[35] N. Benipal, J. Qi, J. C. Gentile, and W. Li, "Direct glycerol fuel cell with polytetrafluoroethylene (PTFE) thin film separator," Renewable energy, vol. 105, pp. 647-655, 2017.

[36] H. Hunke, N. Soin, T. Shah, E. Kramer, K. Witan, and E. Siores, "Influence of plasma pretreatment of polytetrafluoroethylene (PTFE) micropowders on the mechanical and tribological performance of Polyethersulfone (PESU)-PTFE composites," Wear, vol. 328, pp. 480-487, 2015.

[37] M. Pandey, G. M. Joshi, A. Mukherjee, and P. Thomas, "Electrical properties and thermal degradation of poly (vinyl chloride)/polyvinylidene fluoride/ZnO polymer nanocomposites," Polym. Int., vol. 65, no. 9, pp. 1098-1106, 2016.