# Non-linear creep of polypropylene utilizing multiple integral

# Mahmoud Fadhel Idan

Department of Civil Engineering, Al-Maarif University College, Ramadi, Iraq

# Article Info

# ABSTRACT

# Article history:

Received Feb 19, 2021 Revised Jun 4, 2021 Accepted Jun 18, 2021

### Keywords:

Creep Integral method Multiple integral representation Non-linear Polypropylene Multiple integral representation (MIR) has been used to represent studying the effect of temperature on the amount of nonlinear creep on the semicrystalline polypropylene (PP) under the influence of axial elastic stress. To complete this research, the kernel functions were selected, for the purpose of performing an analogy, and for arranging the conditions for the occurrence of the first, second and third expansion in a temperature range between 20 °C-60 °C, i.e., between the glass transition and softening temperatures, within the framework of the energy law. It was observed that the independent strain time increased non-linearly with increasing stress and non-linearly decreased with increase in temperature, although the time parameter increased nonlinearly with stress and temperature directly. In general, a very satisfactory agreement between theoretical and practical results on the MIR material was observed.

This is an open access article under the <u>CC BY-SA</u> license.



#### Corresponding Author:

Mahmoud Fadhel Idan Department of Civil Engineering Ma'arif University College Al-Ramadi Road, Al-Ramadi 31001, Iraq Email: dr.mafa57@gmail.com

#### Nomenclature:

- e in Strain tensor
- e (t) Tensile strain
- e<sub>o</sub> Time independent strain
- F<sub>ij</sub> Non-linear continues functional
- F<sub>i</sub> Kernel functions
- F<sub>oi</sub> Time independent component of kernels
- F<sub>mi</sub> Time dependent coefficients of kernels
- m Time dependent coefficient
- N Time exponent
- σ Tensile stress
- $\xi$  Time parameter (s)

# 1. INTRODUCTION

After polymers have been widely used in various industrial and commodity fields, polymeric materials are now widely used in structural and construction applications, as its performance meets the necessary maximum mechanical requirements, hence the importance of research and study. Another parameter of paramount importance is that the creep resistance is uniform when under the influence of a long-term load, as it changes from linear to non-linear behavior. Chronological and historical sequences can be linked to reasonably and acceptably determine the non-linear behavior of the polymer material [1]-[4].

ISSN: 2252-8814

**2**89

Brinson and Brinson [5], Torrens and Castellano [6] used a one-dimensional equation containing first and third order integrals to describe the mechanical behavior characterized by the second function of kernel and up to the third function, for polypropylene (PP) material [7]-[10]. Touati and Cederbaum [11], Lai and Findley [12] identified nine kernel functions to describe the behavior of polyvinyl chloride (PVC) under the influence of both tensile and twisting stress. For further work, only the first and second kernel order of pure tension was assumed, and only the first and third order of pure twist [13]-[17]. When studying the properties of polietilena (PE), "the second kernel arrangement suffices to describe the effect of pure shear distortion and the third kernel order to characterize the effect of pure tensile strength" [18]-[24]. Others researches use multiple integral representation (MIR) and power law to describe the large distortion occurring in nylon-6, in which the mechanical behavior changes from one mode to another, and in each mode the mechanical behavior remains subject to the power law and for all kinds of different loads [25]-[36]. The goal of all of this is to think using a combination of MIR and power law to investigate how temperature affects the mechanical behavior of viscous materials (0.909 g/cm3 at 20 °C) under the influence of uniaxial load.

# 2. RESEARCH METHOD

For the purpose of developing a suitable characterization of nonlinear behavior, it is assumed that the material elongation rate with respect to time (t) depends mainly on the values of the load rate previously placed on the material or sample. In other words, the elongation that occurs in the sample is a function of the rated loads [1].

$$e_{ij} = F_{ij} \left[ \frac{d\sigma(\xi)}{d\xi} \right]_{-\infty}^{t}$$
(1)

Where  $F_{IJ}$  is represent the continuous nonlinear function. The F function can be represented as indicating the degree of precision within the multiple integration equation [5], [6].

$$e(t) = \int_{-\infty}^{t} (t - \xi_1) \frac{d\sigma}{d\xi_1} d\xi_1 + \int_{-\infty}^{t} \int_{-\infty}^{t} F(t - \xi_1 \cdot t - \xi_2) \frac{d\sigma}{d\xi_1} \frac{d\sigma}{d\xi_2} d\xi_1 d\xi_2 + \int_{-\infty}^{t} \int_{-\infty}^{t} \int_{-\infty}^{t} F(t - \xi_1 \cdot t - \xi_2) \frac{d\sigma}{d\xi_1} \frac{d\sigma}{d\xi_2} d\xi_1 d\xi_2 d\xi_2 d\xi_3$$

$$(2)$$

According to (2), the time dependent elongation produced by uniaxial tensile loading applied at time  $\xi_1 = \xi_2 = \xi_3 = 0$  is given by the expression (3).

$$e(t) = F_1(t)\sigma + F_2(t)\sigma^2 + F_3(t)\sigma^3$$
(3)

By substituting the responses obtained from three tensile tests at different levels of stress, (3) becomes a system of simultaneous algebraic equations with the three unknowns,  $F_1$ ,  $F_2$ , and  $F_3$ . These equations have been solved for each step time using "Gaussian elimination" to find these kernels in the form of Findlay's power [8] as (4a), (4b), and (4c).

$$F_1 = F_{01} + F_{m1} t^N (4a)$$

$$F_2 = F_{02} + F_{m2} t^N (4b)$$

$$F_3 = F_{03} + F_{m3} t^N \tag{4c}$$

The equation of tensile strain can be obtained by substituting (4) into (3).

$$e(t) = (F_{01} + F_{m1}t^N)\sigma + (F_{02} + F_{m2}t^N)\sigma^2 + (F_{03} + F_{m3}t^N)\sigma^3 = e_0 + m_0t^N$$
(5)

Where  $e_0$  and  $m_0$  are time independent strain and time dependent coefficient respectively, and both are functions of stress and material constants. N is constant (5) emphasizes that creep reaction of non-linear viscoelasticity materials can be partitioned into time, stretch, and temperature reliance components.

A "creep test" is performed on a sample using a creep meter (note the device in Figure 1). In simple terms, the sample is heated to a temperature. Once the temperature set point is reached, a constant load is applied to exert a longitudinal force on the material's grain structure. Pregnancy is maintained for the duration of the test or until the sample is ruptured. During testing, data is continuously monitored and recorded to qualify for temperature stability, load, and sample elongation.





Figure 1. SM1006 creep test device

#### **RESULTS AND DISCUSSION** 3.

The deformation behavior available at 20 °C and 60 °C only and linear interpolation between them can be made to obtain the deformation at 30, 40, and 50 °C [8]. Kernel functions have been determined at each temperature using three different stress levels ( $\sigma = 1.378$ , 4.136, and 6.897 MPA) in the form of power law, in which the value of time exponent is valid for all levels of loading. From Table 1 it can be noted that the time exponent increases as temperature increases. Kernel functions given in Tables 2, 3, and 4 are presented in Figures 2, 3, and 4. These figures show that, first and third kernels increase non-linearly with time, whereas the second kernel decreases non-linearly. This result is emphasized by the presence of negative values of time dependent coefficients in Table 1. This behavior was observed at each temperature. Also, it was found that temperature increasing caused a shift in values of all kernels. It is critical here to specify that non-linear viscoelasticity can be related to the time dependent on terms of these parts, since the time autonomous terms allude to starting reactions.

Table 1. Variation of time exponent with temperature Table 2. First kernel at different temperature

1

1

2

Temp. °C 20 30

40

50

60

_		-		-
С	Ν		Temp. °C	F1 (M Pa <sup>-1</sup> )
	0.070	_	20	-0.0141241 + 0.132254 t N
	0.0765		30	-0.01284681 + 0.162771 t N
	0.0817		40	-0.007553014 + 0.1886007t N
	0.086		50	-0.008763126 + 0.2219725t N
	0.090		60	-0.1184285 + 0.2514425t N

Table 3. Second kernel at different temperature

.1	able 4	<b>1</b> . '	Third	kernel	at	different	tem	pera	ture

F2 X 10-3 (M Pa <sup>-2</sup> )	Temp. °C	F3 X 10-3 (M Pa <sup>-3</sup> )
.320227 – 9.034076 t N	20	-0.5189451 + 1.540565 t N
.992685 – 6.130732 t N	30	-0.4471407 + 1.608113 t N
.269982 – 2.563623 t N	40	-0.2774595 + 1.613862 t N
.1936700 – 1.05282 t N	50	-0.3519898 + 1.8840761 t N
290401 – 0.3867674 t N	60	-0.3637092 + 2.077798 t N

Non-linearity degree can be related to the temperature increase since time subordinate coefficients are shifted with temperature. From Tables 2, 3, and 4, time dependent coefficients of first and third kernels are increased as temperature increases, whereas decreased for second kernel. Thus, the non-linearity degree increases for first and third kernels, and decreases for second kernel shown in Figures 2, 3, and 4.





Figure 2. First kernel function of PP for axial tensile loading at different temperature

Figure 3. Second kernel function of PP for uniaxial tensile loading at different temperature



Figure 4. Third kernel function of PP for uniaxial tensile loading at different temperature

From these figures, it can be noted that, effects of F1 and F2 are opposite that of F2 up to 40° C. Beyond this temperature, Figure 3 shows that the values of F2 change from negative to positive (i.e., effects of all kernels are gathered). As temperature, the crystalline degree diminishes and causes a decrease in solidness and tensile quality [37], which increases the inclination of materials to deform (strain rate expanding). This change in properties happens due to the holding powers between the atomic chains ending up weaker. As strain rate increasing, the molecular chain is gradually aligned closer together and oriented in the direction of applied stress, thus, the capacity of these adjacent closer chains to bond again increases and causes increasing tensile and stiffness (strain rate decreasing). Crystalline behavior characterized by  $\alpha$  and  $\gamma$ relaxation. Local twisting of atomic chains is represented by  $\gamma$  relaxation, which related with non-linear defects occur in the crystal phase inside the crystal and then move in the form of distortions in the amorphous phase associated with F1 and F2, which increases the tensile strength and reduces the strain rate. This deformation is related to alpha relaxation, and can be represented by F1 and F2. The change in the value of the kernel function agrees with the results of Seeker [37]. Figures 5, 6, 7, 8, and 9 show the results of this representation. Contributions of F1 and F3 at ( $\sigma = 1.378$  MPA) to total F2 opposite strain up to 40 °C. Beyond this temperature, this behavior does not hold up due to distortion of most of the molecular bonds. It was also found that, over a full timescale, F1 contributed significantly to total stress. With reference to Figures 2, 3 and 4, and based on the equation of temperature and time, the effect of time on the kernel function is similar to that of temperature.

For each temperature, substituting time exponent and kernel functions given in Table 1 and Table 2 in (5), creep strain at different stress levels is shown in Figures 10, 11, 12, 13, and 14. Comparison of MIR and experimental results shows that agreement between them was very satisfactory. Also, it was found that, for each temperature, the strain increases non-linearly with time (since the time exponent is not equal to 1) and stress increment causes a shift in these curves. Net impacts of these parts are given in Figure 15 and Figure 16 through the behavior of time free strain e o and time subordinate coefficient m. Figure 15 appears that, for each temperature,  $e_0$  increment non-linearly with stress, and temperature expanding caused a reduction move.

e(t)

- F<sub>1</sub> σ - F<sub>2</sub>σ<sup>2</sup>

• F<sub>3</sub> σ<sup>3</sup>



Figure 5. Contribution of kernel function of PP to total strain at  $\sigma = 1.3788$  MPa and T = 20 °C



Figure 7. Contribution of kernel function of PP to total strain at  $\sigma$  = 1.3788 MPA and T = 40  $^{\circ}C$ 



Figure 9. Contribution of kernel function of PP to total strain at  $\sigma = 1.3788$  MPA and T = 60 °C



10<sup>0</sup> Time(hr) 10<sup>1</sup>

0.3

0,3

0.25

0,1

0

0.05



Figure 8. Contribution of kernel function of PP to total strain at  $\sigma = 1.3788$  MPa and T = 50 °C



Figure 10. Creep curves of PP in different uniaxial tensile loading at 20 °C

-σ = 1.378 MPa

---σ = 2.757 MPa ---σ = 4,136 MPa



Figure 11. Creep curves of PP in different uniaxial tensile loading at 30 °C



Figure 12. Creep curves of PP in different axial tensile loading at 40 °C



Figure 13. Creep curves of PP in different uniaxial tensile loading at 50 °C

Figure 14. Creep curves of PP in different axial tensile loading at 60 °C

Figure 16 shows that m increases non-linearly with stress and temperature increasing caused a shift in m vs stress curves. The great shift occurred between 30 °C and 40 °C, which insulates more in Figure 17 and reflects the effect of temperature on molecular structure. The reduction of  $e_0$  (which represent initial strain) between 30 °C and 40 °C, is related to decreasing of stiffness due to network deformation. Figure 18 show that m is linearly increments with temperature and its effect with increased stress. These results emphasize that PP exhibits nonlinear viscous-elasticity for any deformation of practical importance [13]. Figure 19 shows that time exponent (N) is increasing non-linearly with temperature.



Figure 15. Tensile component of time independent strain  $e_0$  % vs stress at different temperature

Figure 16. Coefficient of time dependent strain mo% vs stress at different temperature



Figure 17. Tensile component of time independent strain  $e_0$ % vs temperature at different tensile stresses

Figure 18. Coefficient of time dependent strain mo% vs temperature at different tensile stresses



Figure 19. Time exponent (N) vs temperature

## 4. CONCLUSION

From the above discussion, we can draw: a) The non-linear behavior of PP is adequately described by power law, which the time exponent is valid for all stress levels; b) Non-linear creep of PP is enough spoken to by the first, second, and third arrange stretch of MIR; c) Time exponent n increase as temperature increases; d) First and third kernels increase non-linearly with time whereas F2 decreases non-linearly; e) All kernels increase non-linearly as temperature increases; f) First and third kernels can represent the deformation of non-crystalline phase, whereas second represent that of crystalline phase, which both occur beyond 40  $^{\circ}$ C; g) Stress increasing caused non-linearly increases in time independent strains and time dependent coefficients; h) Temperature increases cause non-linearly decreases in time independent strains and linearly increases in time dependent coefficient; and h) The time exponent (N) increases non-linearly with temperature.

#### REFERENCES

- H. F. Brinson and L. C. Brinson, "Nonlinear Viscoelasticity," in *Polymer Engineering Science and Viscoelasticity*, Springer, Boston, MA, 2015, pp. 339-377, doi: 10.1007/978-1-4899-7485-3\_10.
- [2] R. H. Ewoldt, A. E. Hosoi, and G. H. McKinley, "New measures for characterizing nonlinear Viscoelasticity in large amplitude oscillatory shear," *Journal of Rheology*, vol 52, no. 6, p. 1427, 2008, doi: 10.1122/1.2970095.
- [3] K. Miyazaki1, H. M. Wyss, D. A. Weitz and D. R. Reichman, "Nonlinear Viscoelasticity of met stable complex fluids," *EPL (Euro physics Letters)*, vol. 75, no. 6, p. 915, 2006, doi: 10.1209/epl/i2006-10203-9.
- [4] P. Fernández, P. A. Pullarkat and A. Ott, "A Master Relation Defines the Nonlinear Viscoelasticity of Single Fibroblasts," *Biophysical journal*, vol. 90, no. 10, pp. 3796-3805, 2006, doi: 10.1529/biophysj.105.072215.
- [5] H. F. Brinson and L. C. Brinson, "Polymerization and classification," in *Polymer Engineering Science and Viscoelasticity*, Springer, Boston, MA, 2008, pp. 99-157, doi: 10.1007/978-0-387-73861-1\_.
- [6] F. Torrens and G. Castellano, "Molecular Classification of 5-amino-2-aroylquinolines and 4-aroyl-6, 7, 8trimethoxyquinolines as Highly Potent Tubulin Polymerization Inhibitors," *International Journal of*

Chemoinformatics and Chemical Engineering (IJCCE), vol. 3, no. 2, pp. 1-26, 2013, doi: 10.4018/ijcce.2013070101.

- [7] H. F. Brinson and L. C. Brinson, Polymer engineering science and Viscoelasticity, Springer, Boston, MA, 2008.
- [8] L. C. S., Nuns, F.W.R. Dias and H.S. da Costa Mattos, "Mechanical behavior of poly tetra fluoro ethylene in tensile loading under different strain rates," *Polymer testing*, vol. 30, no. 7, pp. 791-796, 2011, doi: 10.1016/j.polymertesting.2011.07.004.
- [9] M. C. Koetting, J. T. Peters, S. D. Steichen and N. A. Peppas, "Stimulus-responsive hydro gels: Theory, modern advances, and applications," *Materials Science and Engineering: R: Reports*, vol. 93, pp. 1-49, 2015, doi: 10.1016/j.mser.2015.04.001.
- [10] George Dvorak, Micro mechanics of composite material, Springer, Dordrecht, 2012.
- [11] D. Touati and G. Cederbaum, "On the prediction of stress relaxation from known creep of nonlinear materials," *Journal of Engineering Materials and Technology*, vol. 119, no. 2, pp. 121-124, 1997, doi: 10.1115/1.2805982.
- [12] J. S. Y. Lai and W. N. Findley, "Prediction of Uniaxial Stress Relaxation from Creep of Nonlinear Viscoelasticity Material," *Transactions of the Society of Rheology*, vol. 12, no. 2, p. 243, 1968, doi: 10.1122/1.549120.
- [13] J. S. Y. Lai and W. N. Findley, "Creep of Polyurethane at Varying Temperature under Nonlinear axial stress," *Transactions of the Society of Rheology*, vol. 17, no. 1, p. 63, 1973, doi: 10.1122/1.549296.
- [14] R. Mark and W. N. Findley, "Nonlinear Creep of Polyurethane under Combined Stresses and Elevated Temperature," *Transactions of the Society of Rheology*, vol. 18, no. 4, p. 563, 1974, doi: 10.1122/1.549361.
- [15] M. Garrido, J. R. Correia and T. Keller, "Effect of service temperature on the shear creep response of rigid polyurethane foam used in composite sandwich floor panels," *Construction and Building Materials*, vol. 118, pp. 235-244, 2016, doi: 10.1016/j.conbuildmat.2016.05.074.
- [16] A.G. Denay, S. Castagnet, A. Roy and G. Alise, "Creep compression behavior of a polyurethane foam from cryogenic temperatures: size effect and long-term prediction," *EPJ Web of Conferences*, vol. 6, p. 25006, 2010, doi: 10.1051/epjconf/20100625006.
- [17] R. Mark and W. N. Findley, "Temperature History Dependence in Combined Tension Torsion Creep of Polyurethane under Varying Temperature," *Transactions of the Society of Rheology*, vol. 19, no. 2, pp. 201-213, 1975, doi: 10.1122/1.549369.
- [18] D. A. Siginer and M. F. Letelier, "Heat transfer asymptote in laminar flow of non-linear Viscoelasticity fluids in straight non-circular tubes," *International journal of engineering science*, vol. 48, no. 11, pp. 1544-1562, 2010, doi: 10.1016/j.ijengsci.2010.07.010.
- [19] M. F. Letelier, D. A. Siginer, G. Arriagada and A. González, "Heat Transfer Optimization in Laminar Flow of Non-Linear Viscoelasticity Fluids in Asymmetric Straight Ducts with Inclusions," ASME 2017 International Mechanical Engineering Congress and Exposition at Tampa, 2017, doi: 10.1115/IMECE2017-70994.
- [20] F. Hagani, M. Boutaous, R. Knikker, S. Xin and D. Siginer, "Numerical Modeling of Non-Affine Viscoelasticity Fluid Flow Including Viscous Dissipation Through a Square Cross-Section Duct: Heat Transfer Enhancement due to the Inertia and the Elastic Effects," ASME 2020 International Mechanical Engineering Congress and Exposition, 2020, doi: 10.1115/IMECE2020-23558.
- [21] M. F. Letelier, C. B. Hinojosa and D. A. Siginer, "Analytical solution of the Great problem for non-linear Viscoelasticity fluids in tubes of arbitrary cross-section," *International Journal of Thermal Sciences*, vol. 111, pp. 369-378, 2017, doi: 10.1016/j.ijthermalsci.2016.05.034.
- [22] S. D. Abramowitch, X. Zhang, M. Curran and R. Kilger, "A comparison of the quasi-static mechanical and nonlinear Viscoelasticity properties of the human semi tedious and tendons," *Clinical biomechanics*, vol. 25, no. 4, pp. 325-331, 2010, doi: 10.1016/j.clinbiomech.2009.12.007.
- [23] W. Qi-Ye, W. Jin-Gan, X. M. Wen, X. Wang, L. I Peng and S. Y. Wang, "The Extra date Distortion and the Non-linear Viscoelasticity of Linear PE and its Copolymer Melts [J]," *Polymer Bulletin*, vol. 1, p. 66, 2005.
- [24] Jianye Liu, "Polymer chain topological map as determined by linear Viscoelasticity,", *Journal of Rheology*, vol. 55, no. 3, p. 545, 2011, doi: 10.1122/1.3569136.
- [25] S. M. Beňová, Ľ. Miková and P. Kaššay, "Material properties of rubber-cord flexible element of pneumatic flexible coupling," *Metallurgical*, vol. 54, no. 1, 194-196, 2015.
- [26] S. R. Ryu and D. J. Lee, "Effects of interphone and short fiber on puncture and burst properties of short-fiber reinforced chloroprene rubber," *Journal of Elastomers & Plastics*, vol. 42, no. 2, vol. 181-197, 2010, doi: 10.1177/0095244310362397.
- [27] R. M. Ogorkiewicz, Engineering properties of thermoplastic, John Wily & sons' ltd., 1970.
- [28] V. P. Cyras, L. B. Manfredi, M.-T. Ton-That and A. Vázquez, "Physical and mechanical properties of thermoplastic starch/montmorillonite Nano composites films," *Carbohydrate Polymers*, vol. 73, no. 1, pp. 55-63, 2008, doi: 10.1016/j.carbpol.2007.11.014.
- [29] W. Brostow and H. E. H. Lobland, "Predicting wear from mechanical properties of thermoplastic polymers," *Polymer Engineering and Science*, vol. 48, no. 10, 2008, doi: 10.1002/pen.21045.
- [30] L. Averous, L. Moro, P. Dole and C. Fringant, "Properties of thermoplastic blends: starch- polycaprolactone," *Polymer*, vol. 41, no. 11, pp.4157-4167, 2000, doi: 10.1016/S0032-3861(99)00636-9.
- [31] N. Kohda, M. Iijima, T. Muguruma, W. A. Brantley, K. S. Ahluwalia and I. Mizoguchi, "Effects of mechanical properties of thermoplastic materials on the initial force of the thermoplastic appliances," *The Angle Orthodontist*, vol. 83, no. 3, pp. 476-483, 2013, doi: 10.2319/052512-432.1.
- [32] A. Boubakri, N. Haddar, K. Elleuch and Y. Bienvenu, "Impact of aging conditions on mechanical Properties of thermoplastic Polyurethane," *Materials & Design*, vol. 31, no. 9, pp. 4194-4201, 2010, doi: 10.1016/j.matdes.2010.04.023.

- [33] M. Dong, "Enhanced solid particle erosion properties of thermoplastic polyurethane carbon Nano composites," *Macromolecular Materials and Engineering*, vol. 304, no. 5, p. 1900010, 2019, doi: 10.1002/mame.201900010.
- [34] H. M.C. de Azeredo, "Nano composites for food Packaging applications," *Food Research International*, vol. 42, no. 9, pp. 1240-1253, 2009, doi: 10.1016/j.foodres.2009.03.019.
- [35] C. Silvestre, D. Duraccio and S. Cimmino, "Food Packaging based on Polymer nonmaterial," *Progress in Polymers Science*, vol. 36, no. 12, pp. 1766-1782, 2011, doi: 10.1016/j.progpolymsci.2011.02.003.
- [36] F. Chivrac, E. Pollet and L. Avérous, "Progress in nano-biocomposites based on polysaccharides and nanoclays," *Materials Science and Engineering: R: Reports*, vol. 67, no. 1, pp.1-17, 2009, doi: 10.1016/j.mser.2009.09.002.
- [37] R. Hemanth, M. Sekar and B. Suresha, "Effects of fibers and fillers on mechanical properties of thermoplastic composites," *Indian Journal of Advances in Chemical Science*, vol. 2, pp. 28-35, 2014.

# **BIOGRAPHIES OF AUTHORS**



**Mahmoud Fadhel Idan**. B.Sc. (Mech. Eng.), H.D., M. Sc., PH. D (Mech. Eng. – Design). The author has conducted many researches in the field's space, Engineering of Materials, Physics, and satellite through his work at the Center for Space and Physics Research (BATANI CENTER), and the Universities. These Researches has been published in various journals.