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Journal Name:	International Research Journal of Pure and Applied Chemistry
Manuscript Number:	2014_IRJPAC_9624
Title of the Manuscript:	Support Material Effect for Pt Catalytic Activity
Type of the Article	Original Research Article

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This journal's peer review policy states that **NO** manuscript should be rejected only on the basis of '**lack of Novelty**', provided the manuscript is scientifically robust and technically sound.

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PART 1: Review Comments

	Reviewer's comment	Author's comment (if agreed with reviewer, correct the manuscript and highlight that part in the manuscript. It is mandatory that authors should write his/her feedback here)
<u>Compulsory REVISION</u> comments	Support Material Effect for Pt Catalytic Activity I recommend that the manuscript be published after a few changes have been made. 1.The quality of the English language requires improvement throughout the manuscript. An example is the sentence on lines 132-134. That sentence has no verb. 2.Acronymns are sometimes used without being defined immediately. An example is line 56 where Pt/CB is used. It should be platinum on carbon black (Pt/CB). Other instances are: GNS in the first line of the abstract N-G in the first line of the abstract HPOG line 113	Response: Thank you very much for your comments. I already modified below as: “The amounts of catalysts loading on the glassy carbon are 20 µg for both GNS and N-G. The rotating ring disk electrode (RRDE) polarization curves of GNS and N-G was obtained in O₂-saturated 0.1 M HClO₄ solution”. Please see in the manuscript on lines 132-134 (yellow part). Thank you very much for your comments. I modified in my manuscript (yellow part), below as: “graphene nano sheets (GNS) and nitrogen doped graphene (N-G)” “platinum on carbon black (Pt/CB) commercial catalyst” (line 56, please see in manuscript, yellow part)” “highly oriented pyrolytic graphite (HOPG)” (line 113, please see in manuscript, yellow part)



SDI Review Form 1.6

	3.Line 97 The BET surface area of the commercial catalyst should be provided. 4.On lines 136 and 150, the supplier of the commercial catalyst should be identified. 5.Current density is compared at 0.9 V by the authors. If the comparison were made at 0.7 V, a different result would have been obtained. An explanation for the choice of the potential should be given. 6.Table 1 – Current density, j, has units of [mA	Thank you very much for your comments. I modified in my manuscript (yellow part), below as: “the surface area of carbon black (CB), GNS and N-G was determined by the Brunauer Emmett and Teller (BET) adsorption method are 45, 318 and 594 m² g⁻¹, respectively”. Thank you very much for your comments. I modified in my manuscript (yellow part), below as: “Pt/CB commercial catalysts (Johnson Matthey)” Thank you very much for your comments. I choose current density at 0,9 V due to the maximum oxidation reduction reaction occur at that voltage. In addition at that voltage I can estimate the mass lost of my catalyst as well as effect of the maximum voltage. If the mass lost catalyst is low at 0,9 V, indicating the catalytic activity of catalyst is high and not easy to oxidize during the extend usage. Thank you very much for your comments.
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	cm⁻²]. I assume the area is the “cm⁻² “ of the area of the glassy carbon working electrode, 0.283 cm² . That should be stated clearly. The other area that might have been used is the BET surface area. 7.The sentence on lines 195-197 cites [reference 29] by Shao et al. (2011). I read this reference. Shao et al. explain the effect in terms of strong oxygen binding, not the “strong adsorbed intermediates” explanation that the authors have used. Furthermore, Shao et al. indicate that optimum ORR occurs at Pt particle sizes of 2.5 nm. The author’s results are consistent with the findings of Shao et al. if the authors were to make their current density comparison at 0.7 V.	The current density was determined by using 3 electrode systems, namely reference electrode (hydrogen electrode), counter electrode (Pt wire) and working electrode (rotating ring disk electrode (RRDE). The ink solution as a sample was put on working electrode. Therefore the current result should be divided by area of RRDE (0,283 cm2) as shown in Table 1. On the other hand, the BET measurement is a method that used to know surface area of a mass sample. Its unit is mg cm-2. : Thank you very much for your comments. I have modified in my manuscript as your suggestion. The strong adsorbed intermediates sentences have been changed to be strong oxygen binding (yellow part on lines 195-197 in manuscript). I agree with your opinion, in term of oxidation performance of support material, it can be known by measuring current density of support material at 0,7 V. It is caused at that voltage, the carbon as representative of support material component material should be oxidized. Thereby, we can know the performance of support material at 0,7 V.
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	8.Lines 224-228 Do the authors mean that subnano clusters have diameters less than 1 nm. Perhaps that is obvious. Nevertheless it should be stated. 9. Lines 227-228 The authors wrote “It is responsible causing the ORR activity of Pt/GNS is higher than Pt/N-G.” Do they mean “Subnano clusters are responsible for causing the ORR activity of Pt/GNS to be higher than Pt/N-G.”? Shao et al. [reference 29] state that the Pt cluster size that gives optimum ORR is 2.5 nm, not subnano clusters. That contradicts the statement by the authors in lines 227-228. The authors need to explain this better. 10.Lines 246-248 I know the authors claim that the XPS binding energies are accurate to plus or minus 0.05 eV. Nevertheless the difference in binding energies between GNS (71.6 eV) and N-G (71.7 eV) is not large enough to really convince	Thank you very much for your comments. I have modified my manuscript as your suggestion in yellow part below as: “Interestingly, the Pt diameters less than 1 nm (Pt subnano-clusters) were only formed on GNS and N-G, but not on CB”. Thank you very much for your comments. I have modified my manuscript as your suggestion in yellow part below as: “I suppose there are two factors which affect it namely the support material effect and Pt subnano-clusters. At the same amount (20 wt % Pt), Pt supported on GNS has the highest ORR catalytic activity among Pt/N-G and Pt/CB. It is caused the support material effect. In addition, in term of Pt/CB, there is no Pt subnano clusters are formed. On the other hand, Pt subnano clusters are formed on GNS and N-G. It is responsible causing the ORR catalytic activity of Pt/GNS and Pt/N-G is higher than Pt/CB”. Thank you very much for your comments. I have modified my manuscript as your suggestion in yellow part below as: “In addition, there are three possibilities nitrogen species doped in graphene layer,
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	me that they might not be the same within experimental error ($71.6 + 0.05$ is the same as $71.7 - 0.05$). In other words the interaction between the Pt particles and the support is not the only explanation. Another explanation for the difference between GNS and N-G might be the fact that GNS has a BET surface area of $318 \text{ m}^2/\text{g}$ and N-G has a surface area of $594 \text{ m}^2/\text{g}$. That suggests that the graphene sheets in N-G have fewer atomic layers than GNS. Perhaps the increased surface area is associated with more sites (edge sites?) with the very strong oxygen binding, that Shao et al. [reference 29] indicated were responsible for a low ORR.			namely pyridine, graphitic and amino types. It suggests that the graphene sheets in N-G have fewer atomic layers than GNS. It also indicates that the increasing surface area is associated with more edge sites. The large surface area of N-G causes the strong interaction between oxygen and carbon [29], resulting a low ORR activity. Therefore, the Pt subnano-clusters and graphene are responsible for causing high ORR catalytic activity of Pt/GNS. I consider two factors, those are i) Enthalpy factor ascribes the interface interaction Pt-C via the π-d hybridization. It is possible due to the reactivity of graphene's π-conjugated states, and ii) Entropy factor. GNS has large surface area. It is required for high dispersion of small Pt Particles".																	
	11.Conclusion and Abstract – Consider the following:			Thank you very much for your comments. I have modified my abstract and conclusions in my manuscript as your suggestion in yellow part below as: Abstract: "Therefore, one can control the electronic structure of graphene and Pt by doping of some elements. In addition, the large surface area of N-G due to it has more sites on which the oxygen binding is very strong. It causes the ORR catalytic																	
	<table><tr><td></td><td>Pt particle</td><td>XPS</td><td>BET</td></tr><tr><td>Surface</td><td>Size [nm]</td><td>BE [eV]</td><td>Area</td></tr><tr><td>[m^2/g]</td><td></td><td></td><td></td></tr><tr><td>GNS</td><td>1.2</td><td>71.6</td><td>318</td></tr></table>		Pt particle	XPS	BET	Surface	Size [nm]	BE [eV]	Area	[m^2/g]				GNS	1.2	71.6	318				
	Pt particle	XPS	BET																		
Surface	Size [nm]	BE [eV]	Area																		
[m^2/g]																					
GNS	1.2	71.6	318																		



SDI Review Form 1.6

	N-G 1.3 71.7 594 Since the particle sizes and binding energies are similar and the surface areas are different, perhaps the explanation for the difference between GNS and N-G is related to surface area. Perhaps the greater surface area has more sites on which the oxygen binding is very strong, as reported by Shao et al. I suggest that the conclusion and abstract be modified.	activity of N-G is lower than GNS”. “In addition, there are three possibilities nitrogen species doped in graphene layer, namely pyridine, graphitic and amino types. It suggests that the graphene sheets in N-G have fewer atomic layers than GNS. It also indicates that the increasing surface area is associated with more edge sites. The large surface area of N-G causes the strong interaction between oxygen and carbon [29], resulting a low ORR activity.”. Conclusion: “On the other hand, since the particle sizes and binding energies are similar and the surface areas are different, the difference between GNS and N-G is related to surface area. The greater surface area has more sites on which the oxygen binding is very strong. It also causes the ORR catalytic activity of Pt/N-G is lower than Pt/GNS”.
<u>Minor</u> REVISION comments		
<u>Optional/General</u> comments		