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Field Dependent Free Ion Yields of Room Temperature Tetramethyl Liquids and Their Mixtures

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ABSTRACT

Ionization currents and free ion yields have been measured as a function of the applied electric field in tetramethylgermanium and tetramethyltin exposed to energetic gamma rays, and in binary mixtures of 2,2,4,4 tetramethylpentane, tetramethylsilane, tetramethylgermanium and tetramethyltin. The ion yields in tetramethylgermanium and tetramethyltin are high, making these room temperature liquids interesting candidates for use in liquid ionization detectors.

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1. Introduction

Recently there has been considerable interest in using room temperature liquids as the active medium in large total absorption ionization calorimeters. Small calorimeter modules using tetramethylsilane (TMS) or 2,2,4,4 tetramethylpentane (TMP) have been tested and give encouraging results [1-4]. Larger room temperature liquid ionization calorimeters are presently being assembled [3,4], or are under consideration for future experiments [5].

Some of the properties of TMS and TMP which are relevant to their use in ionization detectors are listed in table 1. Two essential properties of any liquid to be used in an ionization detector are that, at moderate electric fields ($E \sim 10^4$ KV/cm), the liquid must have a large free ion yield G_{fi} and a high electron mobility μ_e. G_{fi} and μ_e are larger for TMS and TMP than most other liquid hydrocarbons [6]. However, at moderate fields the drift velocity of electrons in TMP is probably too low to be of use in the next generation of fast calorimeters required for future hadron colliders [5]. TMS has a higher electron mobility, but its boiling point is uncomfortably low for the safe operation of a large TMS calorimeter. There is a clear need to find a more suitable room temperature liquid having a higher boiling point than TMS, a high G_{fi}, and a higher μ_e than TMP.

It is natural to consider as potential candidates for liquids to be used in room temperature ionization detectors (i) tetramethyl liquids similar in structure to TMS but with a central group IV atom heavier than Silicon (giving a higher boiling point than TMS), and (ii) mixtures of the tetramethyl liquids. We are led to consider tetramethylgermanium (TMG), tetramethyltin (TMSn), and tetramethyllead (TMPb). Pure TMPb is unstable [7]. This leaves TMG and TMSn for further consideration. Some relevant properties of TMG and TMSn are listed in table 1. Both liquids have boiling points significantly higher than TMS. Furthermore, both TMG and TMSn are known to have high values of μ_e [8], higher than TMP and comparable to that of TMS.

In this paper we report measured values of $G_{fi}(E)$ for TMG and TMSn from low electric fields up to fields of 40 KV/cm, and compare these field dependent yields with those of TMS and TMP. We also report measurements of G_{fi} for binary mixtures of TMS, TMG, TMSn and TMP.

2. Method

The ionization current produced in the tetramethyl liquids by γ radiation was measured as a function of the applied electric field using a guard ring type parallel plate ionization chamber. A schematic drawing of the chamber is shown in figure 1. It consists of a 19.05 mm outer diameter stainless steel tube, 50 mm long, with 1.6 mm thick walls. There is a guard ring half way down the tube, and flanges at both ends. The chamber was closed using end flanges sealed with copper O-rings. The end flanges support low and high voltage electrodes on central pins which are fed through to external BNC signal and SHV connectors. The low and high voltage electrodes are cylindical with polished faces and have diameters of 11.8 mm and 13.8 mm respectively. The distance between the electrodes and the flanges is adjustable, allowing the electrode gap to be changed. The electrode positions were set as follows: (i) the low voltage electrode was adjusted on its supporting pin so that when mounted in the chamber the electrode face would be aligned with the guard ring, (ii) the low voltage end flange was mounted on the chamber, (iii) the high voltage electrode was adjusted so that the electrode gap would be approximately 1 mm when the chamber was closed, (iv) the high voltage end flange was then mounted on the chamber with the electrode gap defined using a 0.99 mm spacer inserted between the electrodes, (v) the separation between the two outer flanges was measured, and finally (vi) the high voltage flange was dismounted, the spacer removed, and the high voltage end flange remounted on the chamber. The electrode separation could then be found by measuring the final separation between the end flanges. The electrode gap was 0.98 ± 0.02 mm. This value was confirmed by measuring the capacitance between the electrodes.

Two fill tubes enable liquid to be inserted into or removed from the chamber, which held 10 cm^3 of liquid. The tetramethyl liquids were dried by passage through a column of activated silica gel, and then pipetted into the chamber. To identify any significant impurities in the liquids they were analyzed with a mass spectrometer. All the liquids used (listed in table 2) were better than 99.8% pure. Binary mixtures of TMP, TMS, TMG and TMSn were prepared by pipetting the desired volume of each liquid into a dry beaker, stirring, and then pipetting the mixture into the chamber.

After filling with liquid the ionization chamber was exposed to γ radiation produced by 1.95 MeV electrons incident on a 0.25 mm tungsten target. Low energy photons (less than ~ 400 KeV) were suppressed using a filter consisting of 12 mm of lead and 2 mm of tin. The ionization chamber was placed downstream of the filter, 40cm from the target, and received a dose rate of about 0.5 mrad/sec. The ionization

current was measured using a conventional pico-ammeter (Keithley Instruments Model 610BR) calibrated so that its absolute response was known to better than 1%. To check the reproducibility of our results the chamber was filled with TMP and the ionization current at 10 KV/cm measured before and after each of the other liquids (TMS, TMG, and TMSn) was put into and taken out of the chamber. The rms spread on these TMP measurements was 1.6 %, which we take as the relative (liquid to liquid) uncertainty on our results. As a final check we measured the field dependent free ion yield in n-hexane and found good agreement with theoretical predictions [9].

3. Results

Our measured ionization currents are shown as a function of electric field in figure 2 for TMP, TMS, TMG, and TMSn. We also compare our measurements with previously published results for TMP (Ryan and Freeman [11]) and TMS (Jungblut and Schmidt [12]) normalized to our measurements at an applied field of 9.2 KV/cm. There is good agreement between the different measurements of the field dependence of the ionization current in TMP and TMS. At high fields the ionization currents measured in TMG are comparable to those observed in TMP, whilst the currents measured in TMSn are significantly larger than those measured in TMP. We note that due to the contribution from photoelectric absorption removal of the lead-tin low energy photon filter results in an increase in the observed ionization currents for TMS, TMG, and TMSn relative to TMP. This increase was found to be approximately 100% for TMSn, 30% for TMG, and 3% for TMS. Removal of 5 mm of lead from the lead-tin filter resulted in an increase in the ionization current of only 5%, indicating that the filter was reasonably efficient.

To extract the field dependent G_{fi} from our measured ionization currents we normalize our results to the measured G_{fi} at 9.2 KV/cm for TMP from ref. [11]. The ion yield for liquid L is then given by :

$$G_{fi}^{L}(E) = G_{fi}^{TMP}(9.2\ \text{KV/cm}) \frac{I^{L}(E)}{I^{TMP}(9.2\ \text{KV/cm})} \frac{\rho_e^{TMP}}{\rho_e^{L}}$$

where I(E) are the measured ionization currents at the applied field E, and the electron densities ρ_e are given by :

$$\rho_e = \frac{\rho Z N_A}{M}$$

where ρ, Z, and M are the density, atomic number, and molecular weight of the liquid, and N_A is the Avagadro number. The resulting field dependent G_{fi} for the four tetramethyl liquids are shown in figure 3. Our results for TMP and TMS are in agreement with previous measurements [11,12].

We have also measured the ionization currents for binary mixtures of TMP, TMS, TMG and TMSn. The resulting G_{fi} for an applied field of 6.1 KV/cm are shown in figures 4 and 5 as a function of the mole fraction of the component liquids. In extracting G_{fi} from the ionization currents we have calculated the electron density assuming that there is no volume change on mixing the component liquids. The G_{fi} for binary mixtures of TMP with TMS, TMG, and TMSn (figure 4) decreases from a maximum for the pure higher-yield liquid through a minimum as more lower-yield liquid is added, and then increases again to the value for the pure lower-yield liquid. This behaviour is also observed for TMS-TMSn (figure 5b), and TMG-TMSn (figure 5c) mixtures. In contrast the ion yield for TMS-TMG mixtures (figure 5a) exhibits a small enhancement corresponding to a TMS mole fraction of about 0.8 to 0.9.

4. Conclusions

The high field ionization currents are higher in TMG and TMSn than in TMS or TMP. TMG and TMSn also have higher boiling points than TMS, and higher electron mobilities than TMP, making them interesting liquids for further study as the sensitive medium in the next generation of room temperature liquid ionization calorimeters. To compare the relative performance of liquids used in ionization detectors we note that for a fixed detector geometry and applied electric field (i) the signal from the passage of a minimum ionizing particle will be proportional to the energy deposited in the liquid per unit path length, $(dE/dx)_L$ (MeV cm^{-1}), and to G_{fi}^L (ion pairs per 100 eV deposited), (ii) the noise will be proportional to the detector capacitance, and therefore to the dielectric constant of the liquid ε_L, and (iii) the noise will be inversely proportional to the length of time over which the signal is integrated, and therefore inversely proportional to the electron drift velocity v_D^L (cm s^{-1}). We define as a figure of merit for liquid L :

$$F_L(E) \equiv \frac{G_{fi}^L(E)\,(dE/dx)_L\,v_D^L(E)}{\varepsilon_L}$$

At E = 10 KV/cm we obtain F = 2.5, 6.6, 7.5, and 7.5 ion pairs ns^{-1} for TMP, TMS, TMG and TMSn respectively.

The ion yields in binary mixtures of TMP with TMS, TMG, and TMSn are less than the yields measured in the component liquids, and exhibit a minimum when the mole fraction of TMP is about 0.5 to 0.7. A similar behaviour is observed in mixtures of TMSn with TMS and TMG, where the minimum is observed when the mole fraction of TMSn is about 0.2. Since the ion yield depends on the range of geminate electrons before they are thermalized, these results may indicate an enhanced scattering of epithermal electrons in these mixtures which results in a shorter range.

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Table 1: Properties of the tetramethyl liquids.

	TMP	TMS	TMG	TMSn
Formula	C_9H_{20}	$C_4H_{12}Si$	$C_4H_{12}Ge$	$C_4H_{12}Sn$
Molecular Wt	128.3	88.2	133.5	178.9
Density (g cm^{-3})	0.720	0.645	1.006	1.314
Boiling Point (°C)	123	27	43	78
Melting Point (°C)	-67	-100	-88	-54
Dielectric Constant*)	1.98	1.92	2.01	2.25
μ_e(cm^2 V^{-1} s^{-1})**)	28[a]	98[a]	90[b]	70[c]
G_{fi} (ion pairs per 100 eV)**)	1.14*)	0.98*)	0.95*)	1.15*)
dE/dx (MeV/cm)	1.55	1.33	1.78	2.09
Radiation Length (cm)	62.3	52.2	18.2	9.2
Interaction Length (cm)	108	129	101	89
F(10 KV) pairs ns^{-1} ***)	2.5	6.6	7.5	7.5

a) Reference [13], b) Reference [8], c) Reference [10]

*) Our measurements.

**) Measured at E=10 KV/cm. Absolute normalization from the data of Ref. [11] giving G_{fi}^{TMP}(9.2 KV/cm) = 1.10.

***) Figure of merit defined in text.

Table 2: Tetramethyl liquids used; their source and the impurites observed in a mass spectrometer analysis.

Liquid	Source	Purity	Trace Impurities
TMP	Wiley Organics	≥ 99.9%	None detectable
TMS	Aldrich	≥ 99.9%	Isooctane
TMG	Wiley Organics	≥ 99.9%	None detectable
TMSn	a) Alfa (Electronic Gr)	≥ 99.9%	Toluene, TMP, TMG
	b) Aldrich	≥ 99.8%	Benzene, Ethylether

FIGURE CAPTIONS

Fig. 1 Schematic drawing of the ionization chamber. The electrodes and guard-ring are shown hatched.

Fig. 2 Ionization currents shown as a function of applied electric field for TMP (our measurement open squares, Ref. [11] closed squares), TMS (our measurement open triangles, Ref. [12] closed triangles), TMG (open circles), and TMSn (closed circles).

Fig. 3 Free Ion Yields shown as a function of applied field for TMP (our measurements open squares, Ref [11] closed squares), TMS (our measurements open triangles, Ref. [12] closed triangles), TMG (open circles), and TMSn (closed circles).

Fig. 4 Free Ion Yields at 6.1 KV/cm for (a) TMS-TMP, (b) TMG-TMP, and (c) TMSn-TMP mixtures shown as a function of mole fraction of TMP.

Fig. 5 Free Ion Yields at 6.1 KV/cm for (a) TMS-TMG, (b) TMS-TMSn, and (c) TMG-TMSn mixtures shown as a function of mole fraction of the component liquids.

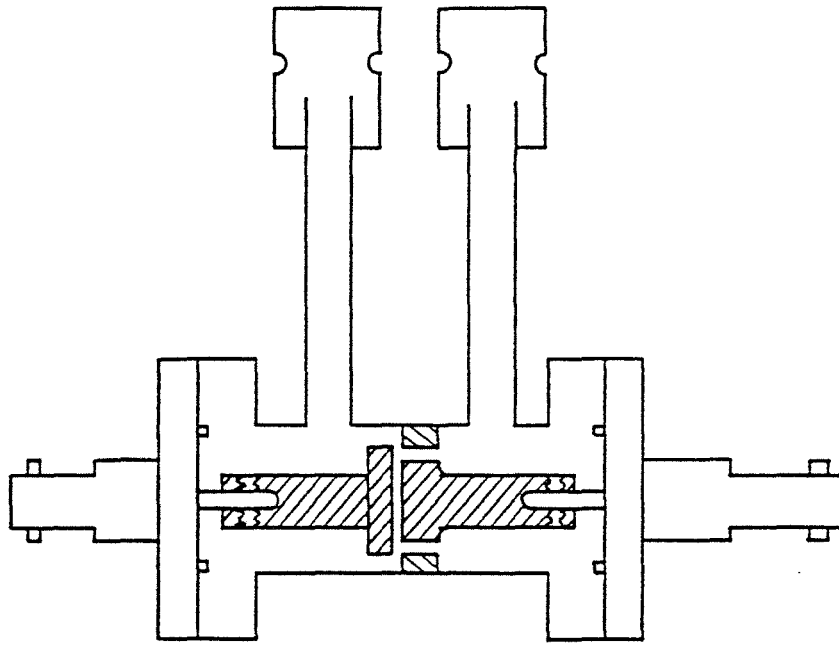

Figure 1

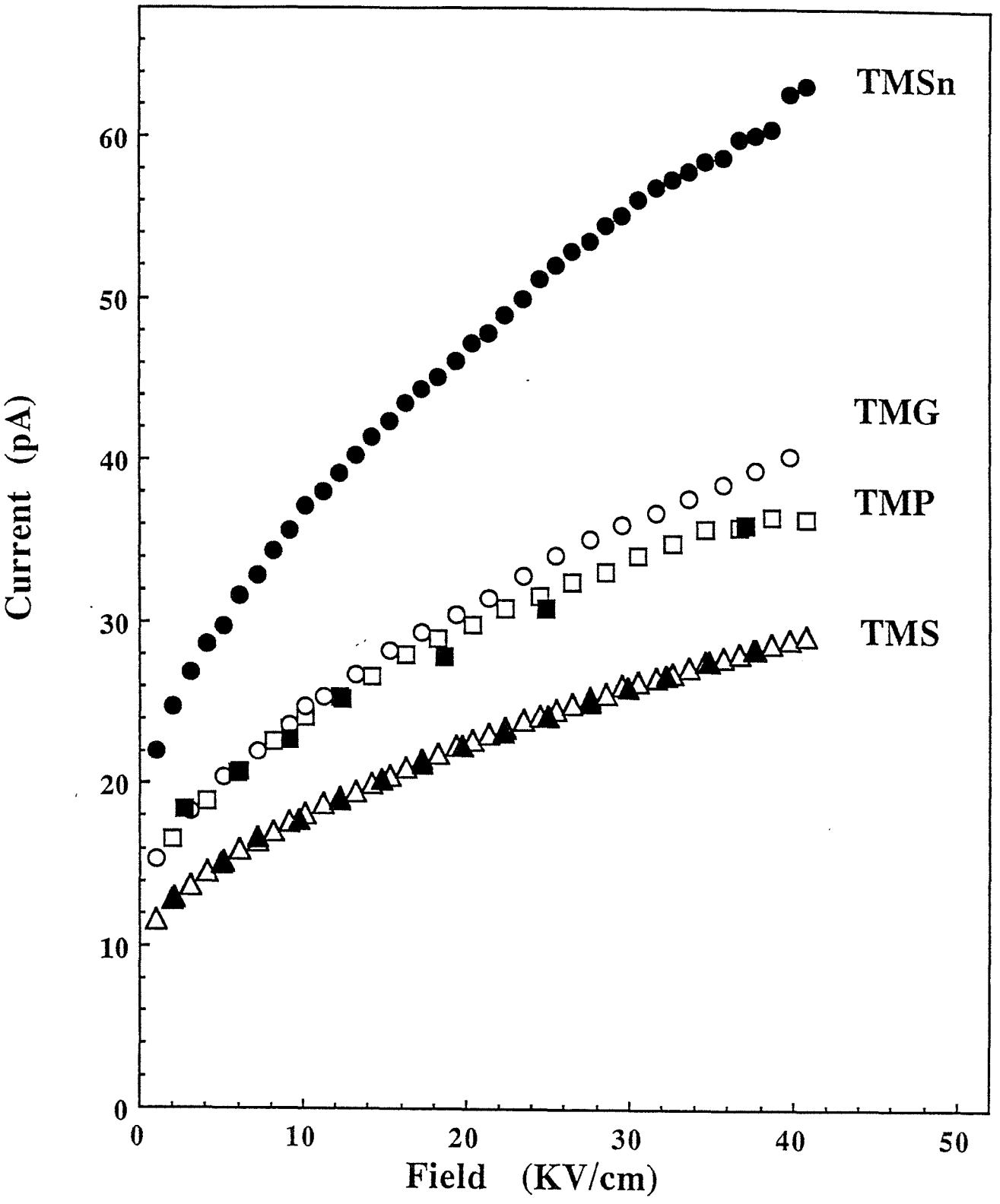

Figure 2

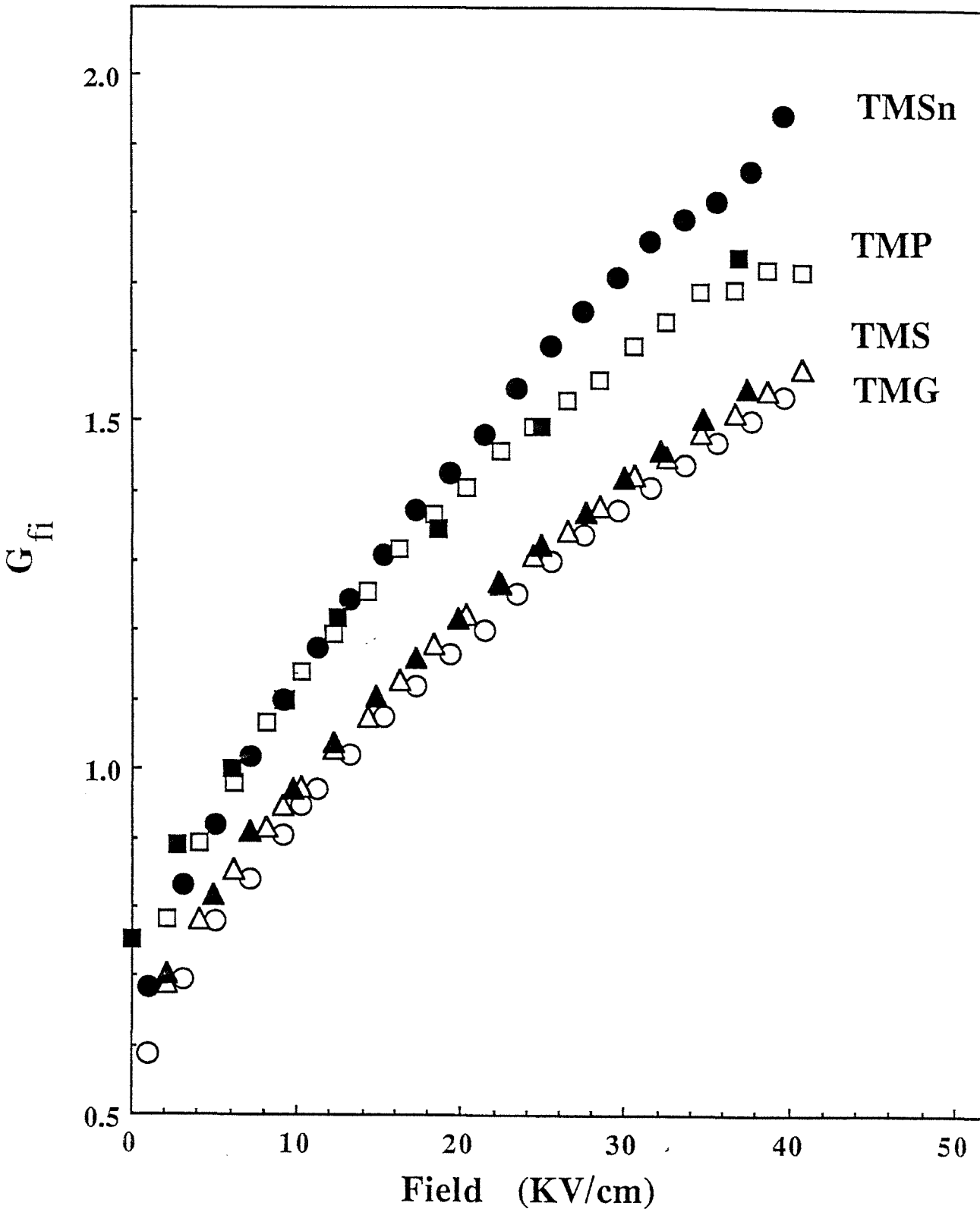

Figure 3

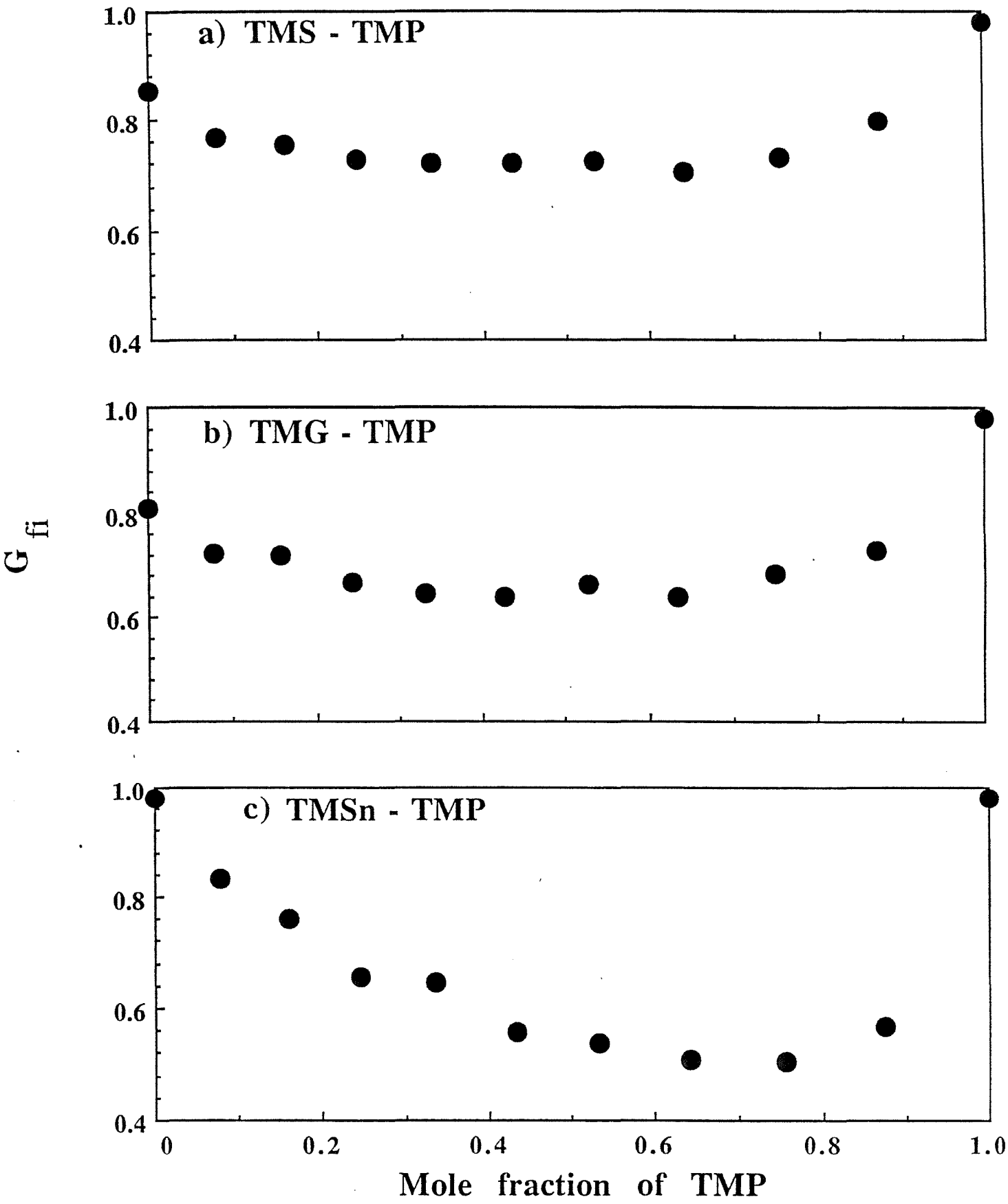
a) TMS - TMP
b) TMG - TMP
c) TMSn - TMP
1.0
0.8
0.6
0.4
0
0.2
0.4
0.6
0.8
1.0
G_{fi}
Mole fraction of TMP

Figure 4

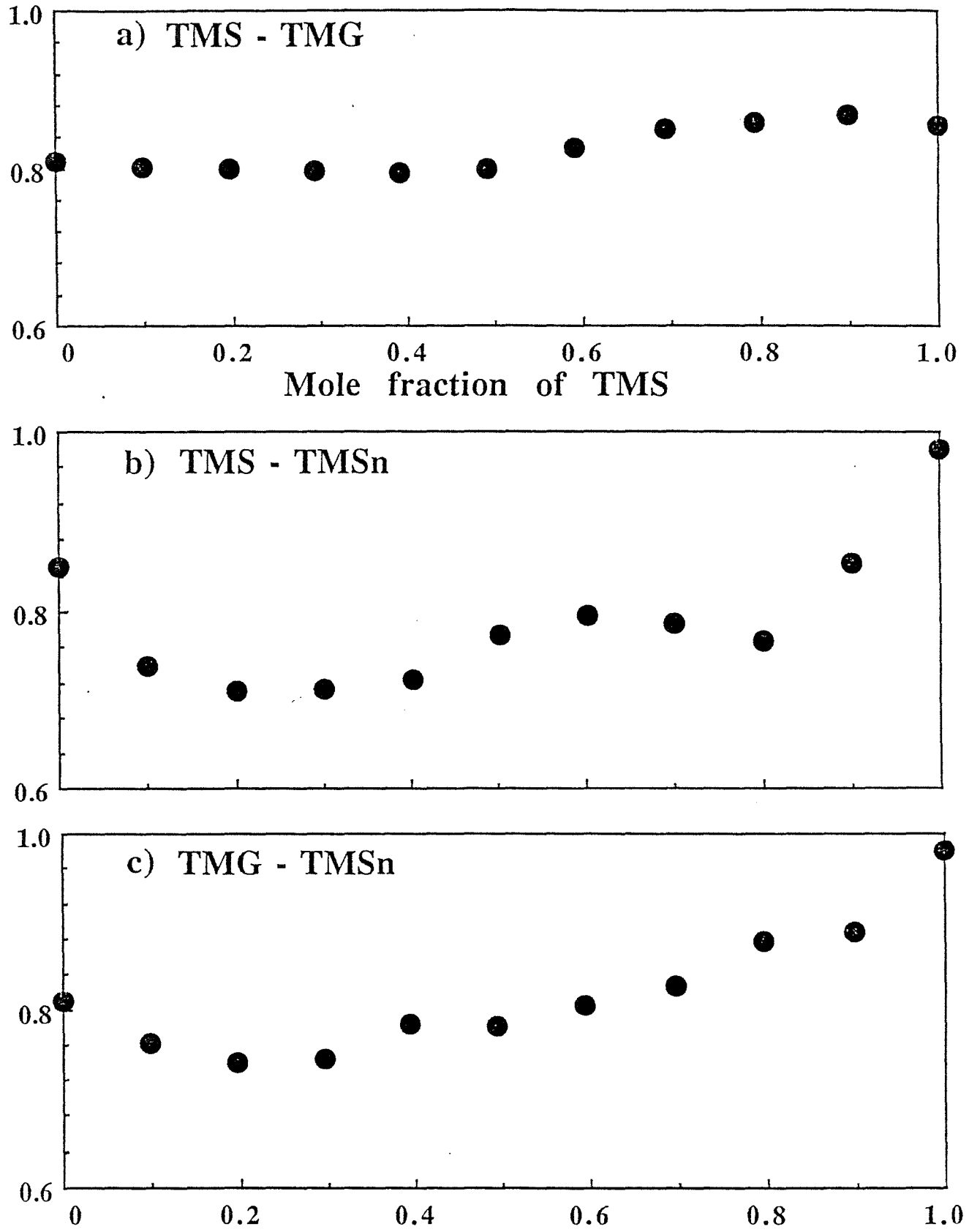
a) TMS - TMG
1.0
0.8
0.6
0
0.2
0.4
0.6
0.8
1.0
Mole fraction of TMS
b) TMS - TMSn
1.0
0.8
0.6
c) TMG - TMSn
1.0
0.8
0.6
0
0.2
0.4
0.6
0.8
1.0
Mole fraction of TMSn

Figure 5