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▶ **To cite this version:**

Romain Bodeux, Dominique Michau, Mario Maglione, Michaël Josse. Thin films sputtered from Ba2NdFeNb4O15 multiferroic targets on BaFe12O19 coated substrates. Materials Research Bulletin, 2016, 81, pp.49-54. 10.1016/j.materresbull.2016.04.027 . hal-01323323

HAL Id: hal-01323323
https://hal.science/hal-01323323

Submitted on 30 Jun 2016

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Thin films sputtered from $Ba_2NdFeNb_4O_{15}$ multiferroic targets on $BaFe_{12}O_{19}$ coated substrates

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1 **Abstract**

2

3 $Ba_2NdFeNb_4O_{15}$ tetragonal tungsten bronze (TTB) / $BaFe_{12}O_{19}$ (BaM) hexaferrite bilayers
4 have been grown by RF magnetron sputtering on $Pt/TiO_2/SiO_2/Si$ (PtS) substrates. The BaM
5 layer is textured along (00l) while the TTB layer is multioriented regardless of the PtS or
6 BaM/PtS substrate. Dielectric properties of TTB films are similar to those of bulk, *i.e.*, $\varepsilon \sim$
7 150 and a magnetic hysteresis loop is obtained from TTB/BaM bilayers, thanks to the BaM
8 component. This demonstrates the possibility of transferring to 2 dimensional structures the
9 composite multiferroic system TTB/BaM previously identified in 3 dimensional bulk
10 ceramics.

11

12

13 KEYWORDS: A. magnetic materials, A. multilayers, B. sputtering, C. impedance
14 spectroscopy, D. multiferroics

15

1 **1) Introduction**

2

3 "The world Multiferroic" was coined for materials in which ferroic orders, *i.e.* ferroelectricity
4 (FE), ferromagnetism (FM), ferroelasticity,… coexist in the same phase. These
5 multifunctional materials have recently attracted great scientific and technologic interests due
6 to their versatile properties which could lead to potential applications in memory devices,
7 actuator and sensors [1]. Multiferroism can be achieved when, in a single-phased material,
8 one cationic sublattice induces a structural distortion and a polarization, while a second
9 sublattice built of paramagnetic ion induces a magnetization [2]. However such materials are
10 relatively unusual and many efforts have been dedicated to "multiferroic composite
11 materials", which associate two ferroic phases [3]. Commercial magnetoplumbite type barium
12 ferrite $BaFe_{12}O_{19}$ (BaM) is a good candidate for the design of multiferroic composites because
13 it is a hard magnet, with a large uniaxial anisotropy which could be useful in micro- or
14 nanostructured materials. Different devices were recently investigated combining BaM with a
15 ferroelectric material, *i.e.* $SrBa_2Ta_2O_9$/BaM thin films heterostructures [4], multilayers
16 consisting of alternating BaM layers with $Pb(Zr,Ti)O_3$ (PZT) [5] or $(Ba,Sr)TiO_3$ (BST)
17 perovskite layers [6-11] and self-assembled BaM clusters embedded in a PZT or BST matrix
18 [12-14]. It was also shown that $BaTiO_3$/BaM ceramics exhibit magnetoelectric coupling at
19 room temperature [15,16].
20 The main aim of the present paper is to show that tetragonal tungsten bronze niobiates can be
21 alternative compounds for the processing of such composite multiferroics. TTB forms an
22 important family of dielectric materials. Their general formula $(A1)_4(A2)_2(C)_4(B1)_2(B2)_8O_{30}$
23 allows many cationic substitution that leads to tunability in their physical properties. Thus
24 TTB could be easily compatible with coexistance of ferroelectricity and ferromagnetism after
25 the introduction of paramagnetic cations. Original bulk multiferroics composites were already

1 identified in the $Ba_2LnFeNb_4O_{15}$ (Ln:TTB) family (Ln = Nd, Sm and Eu) [17,18].
2 Ferroelectric behaviour in this family is driven by the size of the rare earth ions and their
3 accommodation leads to the formation of a small amount of barium ferrite BaM. This
4 magnetic phase is generated *in situ* during the ceramic processing, which indicates a very high
5 chemical stability of both FE and FM phases in this composite system. Castel and al.
6 investigated the elaboration of Eu:TTB composite samples with an excess of hematite in the
7 nominal composition [19], while Roulland et al. tuned the Europium content in Eu:TTB [20].
8 They have shown that the magnetic response is modulated by the amount barium ferrite,
9 which can be increased with excess of hematite or reduced by diminishing the rare earth
10 content. More recently, Ta substitution for Nb was identified as another crystal chemical lever
11 to control these multiferroic composites [21]. Attempts to induce the formation of barium
12 ferrite in other TTB structured system proved unsuccessful, revealing how special the
13 $Ba_2LnFeNb_4O_{15}$ composite multiferroic ceramics are rare [22]. Thus multiferroic composites
14 based on these TTBs, in which ferroelectric properties and magnetization can be tuned, are of
15 interest. In this view, it is important to investigate Ln:TTB thin film deposited on BaM
16 coated substrate. Stability and chemical compatibility of BaM with Ln:TTB is a real
17 advantage for this heterostructure, although it seems particularly complex and requires
18 optimum growth for both TTB and BaM layers. Previous reports on individual BaM and TTB
19 films are encouraging for the development of such systems,. On the one hand, BaM thin films
20 deposited on PtS were widely studied because a Pt underlayer favors the growth along the
21 easy axis of magnetization, required for applications [23,24]. On the other hand, relaxor TTB
22 thin films were deposited by RF magnetron sputtering on Pt(111) and showed dielectric
23 properties very close to those of bulk ceramics [25,26].
24 In the present work, we compare growth orientation, microstructure and dielectric properties
25 of $Ba_2NdFeNb_4O_{15}$ films deposited on silicon with Pt or BaM/Pt intermediate layers. We find

1 that the ferromagnetic properties of BaM and the intrinsic TTB dielectric behaviour are kept
2 in such bi-layers. More interestingly, because of the good chemical and structural
3 compatibility between the two phases, possible magneto-dielectric coupling is anticipated.
4
5 **2) Experiments**
6
7 $Ba_2NdFeNb_4O_{15}$ tetragonal tungsten bronze (TTB) and $BaFe_{12}O_{19}$ (BaM) ceramic targets were
8 prepared through a solid state route. TTB target was sintered at 1300°C from stoichiometric
9 mixture of $BaCO_3$, Fe_2O_3, Nb_2O_5 and Nd_2O_3 [25] while BaM was sintered at 1000°C from
10 stoichiometric mixture of $BaCO_3$ and Fe_2O_3. The purity of the targets was checked by XRD
11 using small control pellets sintered with the targets.
12 TTB films were grown on Pt(111)/TiO_2/SiO_2/Si(100) (PtS) and BaM/PtS substrates by RF
13 magnetron sputtering (Plassys PM450 - 13.56 MHz) [19]. During all depositions, the
14 atmosphere pressure of 40 mTorr was a mixture of 10% O_2 and 90% Ar. PtS substrates were
15 purchased from INOSTEK and appropriate masking during the films deposition enabled the
16 use of the Pt underlayer as the bottom electrode. The deposition parameters of BaM films
17 were optimized regarding crystalline quality and following previous reports [23,24]. All BaM
18 films were deposited at 300 °C. Crystallization took place in a tubular furnace during the post
19 deposition annealing step at 850°C for 3 H in oxygen atmosphere. TTB films were deposited
20 at 300 °C and post annealed at 800°C for 1 H in oxygen atmosphere. Films thickness was
21 measured using a Veeco Dektak6M stylus profile mechanical profilometer. Various
22 thicknesses of layer were obtained by appropriately tuning the deposition time, the thickness
23 of TTB layers varying between 250 nm and 1000 nm, that of BaM layers one varying
24 between 0 and 200 nm (table 1). X-ray diffraction data in the θ–2θ Bragg–Brentano
25 geometry were recorded on a Philips XPert pro diffractometer (Cu $K\alpha_1$ radiation, λ = 1.54056

Å) with 10° < 2θ < 80° and step = 0.02°. Sample's surface morphology was examined by atomic force microscopy (Veeco dimension 3100). Magnetic hysteresis loops were measured on a SQUID magnetometer (MPMS, Quantum Design Inc.) on samples cut to small pieces compatible witht the sample holder. Pt Circular dots were deposited ex-situ at room temperature as a top electrode by dc sputtering through a metal mask at room temperature. The dot geometry defined the capacitors area: $S = 0.1\ mm^2$. Dielectric properties of capacitors were measured in the metal/insulator/metal configuration using a Hewlett-Packard 4194 impedance analyser as a function of frequency (100 Hz – 1 MHz) and temperature (on heating) with an ac excitation amplitude of 100 mV.

3) Results and discussion

After standard solid state processing of powders and dense ceramics, X-ray diffraction (XRD) evidenced the successful stabilisation of the tetragonal tungsten bronze structure for the compound $Ba_2NdFeNb_4O_{15}$ (TTB). The XRD pattern of the TTB ceramic target is shown in Fig.1a. Diffraction pattern matches with the reference data for homologous $Ba_2LaNb_5O_{15}$ TTB (JCPDS 46-0232). In a previous work [27], X-ray microprobe experiment revealed that traces of $BaFe_{12}O_{19}$ (BaM) barium ferrite should be present in the target ceramics, although it was not detected by XRD. In fact, chemical strain and distortions induced by smaller rare earths lead to the evolution of the TTB compositions and the occurrence of secondary phases, *i.e.* $LnNbO_4$ fergusonite and BaM.

Fig. 1b shows the XRD pattern of a sputtered films deposited at 300°C and of a film post-annealed at 800°C (sample A in table 1). After deposition the films were amorphous and post-annealing was required to trigger crystallisation. The annealed films displayed a diffraction pattern compatible with the TTB structure, with several peaks easily identified by comparison

with the ceramic target. The films were polycrystalline and the diffraction peaks exhibited a distribution of relative intensity rather similar to that of ceramics. A notable exception to these intensity ratio is found with (00l) peaks, the amplitude of which is significantly larger than in the ceramics. This suggests a partial texturation of the polycrystalline film along [001] . The overall multioriented growth of films is nonetheless in agreement with other post-annealed compounds of the same family [28]. The mismatch of peak positions with those of ceramic indicates that the films are compressive strained. Strain can be induced by the lattice misfits between TTB and Pt at the interface or produced by the intrinsic stress during the post-annealing. The diffraction peaks, which are relatively narrow, suggest a good quality of the films. The full width at half maximum (FWHM) of TTB(001) reflection is $\Delta 2\theta = 0.19°$ which is close to that of Pt(111) which is $\Delta 2\theta = 0.20°$. An average crystallite size of about 50 nm was obtained by the Scherrer's formula.

The use of $BaFe_{12}O_{19}$ (BaM) for high density magnetic recording applications requires thin films with perpendicular c-axis orientation, because of BaM large magnetic anisotropy. C-axis growth of BaM can be obtained by using a Pt underlayer [23,29-33]. However, high temperature deposition is required to produce BaM films with high quality by RF magnetron sputtering and post annealing was generally undertaken [24]. Our preliminary works are in agreement with these previous reports. XRD pattern are shown in Fig. 2 for the BaM films deposited on PtS(111) with optimized conditions (sample C) in table 1. The BaM films displayed intense (00l) peaks, indicating that the films are crystallized and suggesting a (00l) texture. From the crystallography view point, the (111) plane of packed Pt atoms is seen as a hexagonal structure with a lattice constant a = 5.547 Å, which is close enough from (001) plane of the hexagonal BaM structure with a = 5.892 Å. The relatively small mismatch between Pt(111) and BaM(001) favors the c-axis orientation of the BaM layer. The TTB films were deposited on BaM/PtS in the same deposition conditions than the ones on PtS, *i.e.* as-

1 deposited TTB films were amorphous and crystallization took place during the post
2 deposition annealing step at 800 °C (sample E). Fig.2 shows XRD pattern for the
3 TTB/BaM/PtS stack (sample E). TTB films are multioriented while BaM remains textured
4 (001), and this results demonstrates the successful deposition of TTB on the BaM phase to
5 form a bilayer. The peaks of the BaM layer are well defined indicating that the integrity of the
6 BaM phase was preserved through the second heat treatment. More interesting, the
7 introduction of a BaM buffer layer restores the random orientation of the grains in the TTB
8 film. Generally, when synthesizing bi- or multilayers, the structure is significantly affected by
9 the under-layer structure. The under-layer can lead to strains of the top layer crystal lattice
10 and partial texturing. However, the large mismatch of both Pt and BaM with the tetragonal
11 structure of TTB layer (a ~ 12.48 Å in the ceramic) should induce disorder and dislocations at
12 the interface. The interaction between BaM/Pt and TTB confines at the interface. Thus, the
13 structure within TTB layer remains unchanged and random orientations of grains take place.
14 Furthermore growth procedure can play a significant role on the TTB structure. In fact, we
15 observed similar results on ceramic composites, which are obtained from solid state route
16 consisting in a mixture followed by sintering at highest temperature ~1200°C, i.e. both TTB
17 and BaM phases were maintained. This shows that these two phases coexist without any
18 intermixing in these thermodynamical conditions.
19 Fig. 3a and Fig 3b show the typical morphology of the surface for TTB films deposited on PtS
20 and BaM/PtS substrates (sample A and sample E, respectively). The grains are distinctly
21 visible, assuming that the films are well crystallized, as confirmed by XRD. The grains are
22 densely and regularly packed without cracks or voids and the surfaces are relatively uniform
23 and smooth. The film shows a granular surface with a regular and fine grain size about 50 nm
24 which is comparable to the crystallite size obtained from XRD data. The granular surface of
25 films is typical of post-annealed films formed from amorphous film precursors. It was also

1 observed in others materials [34] and is relatively independent of the buffer layer [35,36]. The
2 root-mean-square (rms) roughness surface roughness is about 4.4 nm and 7.6 nm for PtS and
3 BaM/PtS substrates. The higher value for BaM/PtS has to be associated with the roughness of
4 BaM (about 13.3 nm) which leads to dark areas on the picture obtained from TTB/BaM/PtS
5 stacks. However, the peak-valley distance (50 nm) is clearly lower than the thickness of all
6 TTB films (>250 nm). Therefore, the TTB films are suitable for dielectric measurements in
7 metal/insulator/metal configuration.

8 Ferromagnetic response of $Ba_2NdFeNb_4O_{15}$ ceramics is related to the presence of
9 small amount of barium ferrite and this spontaneous magnetization reaches M_S ~ 1 emu/g at
10 10 kOe [27]. The smaller quantity of matter in the films (three orders of magnitude with
11 respect to ceramics), would yield traces of *in situ* formed BaM, which should not be sufficient
12 to overcome the diamagnetic response of PtS substrate, *i.e.* M_{dia} ~ 2 memu per gram of PtS
13 substrate at 10 kOe (not shown). However, the magnetic properties of TTB composites can be
14 controlled by inducing the formation of an excess of barium ferrite [19]. In this view, the
15 elaboration of TTB/BaM/PtS stacks should allow to investigate a layered architecture, which
16 could be used as a model system of the corresponding ceramic system. Magnetic hysteresis
17 loops of such a stack (sample E) at 300 K are shown in Fig. 4. The stack exhibits large
18 remanence, as expected for a single BaM layer. The loop is attributed to the prevailing easy-
19 axis of magnetization along the BaM(001) orientation [23]. Spontaneous magnetization
20 reaches ~7.10^{-3} emu per gram of PtS substrate while the coercive field is about 1 kOe, which
21 is consistent with the values reported for BaM films [37].

22 The capacitance C was extracted from impedance measurements Z* Z'-jZ''. The dielectric
23 losses were related to the complex impedance through the relation tan δ = Z'/Z''. In figure 5
24 is shown the frequency dependence of capacitance and the dielectric losses for the
25 TTB/BaM/PtS stacks with various thicknesses of BaM layer, 0, 100 et 200 nm (sample A, E

and B, respectively). Below 10 kHz, capacitance exhibits a constant value about 5 nF/mm^2 for the stack with a BaM layer thickness of 100 nm, similar to the one observed in a TTB single layer. The capacitance slightly decreases about 3.5 nF/mm^2 when the BaM layer thickness increases, as discussed below. Thus, the dielectric properties of TTB films are maintained when deposited on the BaM buffer layer. These results are consistent with the XRD and AFM measurements which show that the TTB layer is not affected by the BaM layer. To values greated than 10 kHz, a decrease of capacitance is observed for the TTB/BaM/PtS stack. The relaxation in capacitance is accompanied by a broad peak in tan δ at f = 300 kHz, which doe not appear in the TTB/PtS stack. For the TTB/PtS stack, the limited increase of dielectric losses at 1 MHz is an artefact arising from the stray inductance of the contacts. The appearance of a dielectric anomaly on TTB/BaM/PtS stacks suggests that it is related to the insertion of the BaM layer. BaM is extensively used in the microwaves devices because of its high resistivity which allows minimizing the losses. This dielectric response of the BaM layer may act as an additional impedance source on the impedance measurements connected in series to the TTB film [36]. From these results, the dielectric response can be modelled and analysed using two RC parallel electrical elements, one corresponding to TTB film and the others to an interface capacitance. The contribution of each of these elements to the dielectric response is then visible over characteristic frequency ranges, *i.e.* $\omega = (RC)^{-1}$. Thus, the dielectric response of the TTB layer dominates at low frequency while that of the BaM layer appears at high frequency.

To further analyse the size effect in stacks, the dielectric properties of four TTB/BaM stacks were studied for which the TTB layer were deposited at various thicknesses: 250, 500, 750 and 1000 nm (sample D, E, F and G, respectively). The frequency dependence of capacitance and tan δ are shown in Fig. 6a and 6b, respectively. The capacitance slightly

increases from 2.1 nF/mm^2 to 4.8 nF/mm^2 when the thickness of the TTB layer decreases for the high frequency range f < 100 kHz,

A large increase of capacitance is observed when the frequency decreases below 100 kHz, especially for the 250 nm TTB layer. It is accompanied by a large relaxation in tan δ. The largest capacitance is associated with the thinner TTB layer. In this analysis, the stack capacitance C_{stack} can be written as

$$\frac{1}{C_{stack}} = \frac{1}{C_{TTB}} + \frac{1}{C_{BaM}} = \frac{t_{TTB}}{\varepsilon_{TTB} S} + \frac{t_{BaM}}{\varepsilon_{BaM} S} \qquad [1]$$

where C_{TTB}, C_{BaM}, ε_{TTB}, ε_{BaM}, t_{TTB} and t_{BaM} denotes the capacitances, the permittivity and the thickness of the TTB and BaM layers, respectively. By plotting $1/C_{stack}$ vs t_{TTB} and t_{BaM} curves at low frequency, f = 100 Hz, and high frequency, f = 1 MHz, from the inset figures 5 and 6, we determine from their slopes the experimental values of ε_{TTB} ~ 150 and ε_{BaM} ~ 40 which are consistent with ones reported in the literature [26,38]

Capacitance and losses of a Pt/TTB/BaM/PtS stack (sample G) are plotted as a function of the temperature at various frequencies 10 kHz, 100 kHz and 1 MHz in Fig. 7. Capacitance exhibits a steplike feature from a low-temperature plateau value of 1.6 nF/mm^2 towards 2.5 nF/mm^2 at high temperatures. The step shifts to higher temperatures with increasing frequency. This typical signature is commonly reported as a Maxwell Wagner relaxation [39]. This relaxation is caused by the contribution of two different electrical elements evidenced in Figure 5 and 6, such as the TTB layer capacitance visible at high temperature and low frequency and the BaM layer capacitance observed at low temperature and high frequency. In addition, the step in C(T) is transferred into a peak in tan δ (T). The characteristic frequency (f_r) of this relaxation is governed by the resistance and capacitance of both TTB and BaM layers according to the relation [39]

$$fr = \frac{R_{TTB} + R_{BaM}}{R_{TTB} R_{BaM} \left(C_{TTB} + C_{BaM}\right)}$$

1 [2]
2
3
4 From Fig. 6 and Fig. 7, it appears that on the one hand R_{TTB} and R_{BaM}, and on the other hand
5 C_{TTB}, and C_{BaM} are the same order of magnitude and therefore each term contributes
6 significantly to the relaxation. The peak shifts to the highest temperatures as the frequency is
7 increased. The relaxation frequency was fitted with an Arrhenius law with a characteristic
8 activation energy E_a ~ 0.34 eV. In hexaferrites, the conductivity arises mainly with
9 temperature from electron hopping between Fe^{3+} and Fe^{2+} cations. caused by the presence of
10 oxygen vacancies. The activation energy of this mechanism is about 0.3 eV and could explain
11 the relaxation observed in our TTB/BaM stacks [40]. For a TTB single layer, a low-
12 temperature anomaly was also observed and was likely related to a relaxor behaviour [24]. A similar
13 value of activation energy of 0.3 eV was reported although the anomaly is shifted to lower
14 temperatures with respect to the Maxwell-Wagner relaxation observed in the stacks. Both mechanisms
15 are rather different, but one should keep in mind that Fe^{3+} cations are also present in TTB compound.
16 Moreover in TTB single crystals, $Fe^{0,\ 2+,\ 3+}$ ions were detected from XPS spectra and were associated
17 with reduced niobium ions ($Nb^{5+,\ 4+,\ 3+}$). Significant dielectric losses were clearly seen in these crystals
18 and may be related to electron hopping within the octahedral framework, as it is statistically occupied
19 by iron and nobium ions. [18]. Further investigation is needed to assess the mechanisms involved both
20 in TTB single layers and in TTB/BaM stacks. Anyhow, the results presented here demonstrate a good
21 potential for the coexistence of ferroelectricity and ferromagnetism in TTB/BaM bilayer thin films.
22 We plan to improve their quality by different synthesis route in order to look for a magneto-
23 capacitance effect.
24
25 **4) Conclusion**
26

Tetragonal tungsten bronze and barium ferrite films from $Ba_2NdFeNb_4O_{15}$ (TTB) and $BaFe_{12}O_{19}$ (BaM) homemade ceramic targets were obtained by RF magnetron sputtering on Pt and postannealed. It was found that growth orientation, microstructure and dielectric properties of the TTB layer were retained regardless of the underlayer (Pt bottom electrode or BaM). A dielectric relaxation appears on Pt/TTB/BaM/PtS stacks and suggests that the insertion of the BaM layer provides an additional impedance source. The capacitance of the TTB layer is visible at high temperature and low frequency whereas that of BaM layer is observed at low temperature and high frequency. The relaxation frequency follows an Arrhenius relationship with a characteristic activation energy E_a ~ 0.34 eV, and is likely related to a Maxwell-Wagner effect. Magnetic hysteresis loops were obtained from the stacks, and are consistent with the magnetic properties of the BaM layer. The loop is attributed to the prevailing easy-axis of magnetization along the BaM(001) orientation. This allows for further development of TTB/BaM/PtS bilayers and multilayers as multiferroic and possibly magnetoelectric composites.

Acknowledgments

This project was supported by the French Agency for Research under contract ANR CROCRODIEL(Grant ANR-09-JCJC-0079).

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1 Table 1 Thickness of TTB and BaM layers for TTB/BaM/PtS stacks
2

Parameter	A	B	C	D	E	F	G
TTB thickness (nm)	500	500	0	250	500	750	1000
BaM thickness (nm)	0	200	100	100	100	100	100

3

1 Fig1. XRD patterns for a) the TTB ceramic target and b) as deposited TTB/PtS film and
2 annealed TTB/PtS film (sample A)
3
4 Fig.2 XRD patterns for BaM/PtS film (sample C) and TTB/BaM/PtS (sample E)
5
6 Fig. 3 (Color online) Morphology image (AFM) of TTB films deposited on a) PtS (sample A)
7 and b) BaM/PtS (sample E)
8
9 Fig. 4 Magnetization per gram of PtS substrate for the TTB/BaM/PtS stack. Thickness of the
10 BaM layer is 100 nm (sample E).
11
12 Fig. 5 Frequency dependence of a) capacitance and (b) the dielectric losses for the
13 Pt/TTB/BaM/PtS with BaM thickness of 0 nm (sample A), 100 nm (sample E) and 200 nm
14 (sample B)
15
16 Fig. 6 Frequency dependence of (a) the capacitance and (b) the dielectric losses for the
17 Pt/TTB/BaM/PtS capacitors with various TTB thicknesses 250 nm (sample D), 500 nm
18 (sample E), 750 nm (sample F) and 1000 nm (sample G)
19
20 Fig. 7 Temperature dependence of a) the capacitance and b) dielectric loss for the
21 Pt/TTB/BaM/PtS (sample G) for various frequency 10 kHz, 100 kHz and 1 MHz
22