Materials Research Express

PAPER

Liquid structure of aluminum binary alloys characterized by electrical parameters under electromagnetic field

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Keywords: thermoelectric power, electrical resistivity, electromagnetic field

Abstract
The electrical parameters of aluminum binary alloys (Al–Fe and Al–Si alloys) have been investigated in the presence of alternating current (AC) and direct current (DC) electromagnetic fields. The results show that both thermoelectric power and electrical resistivity of Al–Fe alloys increase in the AC/DC electromagnetic field, which indicates that the liquid structure is changed by the electromagnetic treatment. Study on Al–Fe alloys with different Fe content reveals that Al–1.99wt.% Fe alloy shows the sharp increase of transient variation ΔS (S: thermoelectric power) and inflection of residual variation ΔS0 at near eutectic composition in AC electromagnetic field, which is comparable with the electrical resistivity in DC electromagnetic field. In addition, the effects of electromagnetic parameters (setting current, frequency and duty cycle) on voltage/current waveform of power system and thermoelectric power have been further studied. After comparing the results of voltage and current waveforms, it shows that the output current can be regarded as a better evaluation index to the effects of electromagnetic parameters on the thermoelectric power. This quantitative method can clearly analyze the effects of electromagnetic parameters on the liquid structure, which is characterized by the thermoelectric power.

1. Introduction

The final properties of cast alloys depend on the microstructure during solidification. Therefore, some basic problems of controlling microstructure during solidification still present many scientific and technological challenges. External electromagnetic field has effects on grain refinement to minimize their disadvantages [1, 2]. Application of electromagnetic treatment has gradually received much attention in the area of material science. It is generally believed that improvement on solidification microstructure stems from structure variation in liquid metals affected by some physical phenomena in the electromagnetic field [3–6]. Diffraction is one of the methods to study the liquid structure, but it needs complex spectral line analyses which limit the further measurement of liquid structure under the electromagnetic field. As a function of structure factor Q, the change of electrical parameters (thermoelectric power or electrical resistivity) reveals the precise variation of liquid structure [7, 8]. Electrical parameters can also reveal the liquid structure during the external electromagnetic field, especially the treatment process [9–12]. Thus, it helps us to understand the structure variation of liquid metals due to the electromagnetic treatment.

The electrical parameters of liquid metals have been studied according to nearly free electron theory by Faber and Ziman [13]. Many researches have focused on the relationship between structure and electrical parameters. For example, thermoelectric power S and electrical resistivity ρ versus temperature have been measured at low-temperature region in magnetic fields up to 15 T [14, 15], as well as chemical element content dependency of microstructure and electrical transport properties under the external magnetic field [16]. Meanwhile, it suggests...
that thermoelectric power can be used to reveal the structural change of phase transition [17]. Therefore, electrical parameters should be discussed to help understand the relationship between liquid structure and solidification microstructures under the electromagnetic field.

Aluminum based alloys have been widely utilized in many fields such as construction, aerospace, electronic domains, and automotive applications [18]. Fe or Si presents as the most common impurity in cast aluminum alloys. Because Al–Fe or Si phases are in the form of irregular flake or bulk shapes which disperse the substrate, resulting in the deterioration of mechanical properties [19–21]. Some researchers have indicated that spherical or finer fibrous Si phases homogeneously distribute in α-Al matrix, which can improve mechanical properties including the tensile strength, ductility, etc [22–24]. Meanwhile, the refined Al–Fe phases can also increase temperature resistance of industrial aluminum alloys [25]. Therefore, effective ways like physical field treatment have been devoted to reduce the grain size and modify the morphology and distribution of second phases. The eutectic point compositions of Al–Fe and Al–Si alloys are 1.8 wt.% and 12.6 wt.% respectively [26, 27]. When Fe or Si content is less than 1.8 wt.% or 12.6 wt.%, it should be defined as Al–Fe or Al–Si hypoeutectic alloy, and its microstructure contains primary α-Al and eutectic phases. On the contrary, when Fe or Si content is more than the eutectic point composition, it should be defined as hypereutectic alloy, and its microstructure contains primary Si or Al–Fe phases and eutectic phases. In this study, we have studied the electrical parameters of liquid metals under alternating current (AC) and direct current (DC) electromagnetic field. Meanwhile, the effects of different electromagnetic parameters (setting current, frequency and duty cycle) on the electrical parameters have been further investigated.

2. Experimental section

2.1. Materials preparation
The materials in this study were Al–xFe (x = 0, 0.99, 1.99, 2.27, 2.89, 3.50, 4.52 wt.%) and Al–xSi (x = 10.06, 15.15 wt.%, corresponding to Al–Si hypoeutectic alloy and hypereutectic alloy respectively) alloys, which were prepared from commercial pure aluminum, iron and Al–Si master alloy. Inductive coupled plasma atomic emission spectroscopy (ICP–AES) measurement showed the chemical compositions of these alloys, which were listed in table 1. Samples were melted in cylindrical corundum crucible by resistance furnace under the atmosphere of high purity argon gas. Meanwhile, the electrical parameters of samples were measured during the heat keeping and electromagnetic treatment.

2.2. Thermoelectric power test
Thermoelectric power and electrical resistivity were measured by four-probe method. Stable current and voltage drop across the sample were detected by two probes respectively. The measurement precision of voltage drop reached ±0.01 μV. The temperature at both ends of sample was detected by HIOKI9334, and the temperature precision was ±0.1 °C. Thermoelectric power was given by calculating voltage drop ΔU and temperature difference ΔT across the samples. Electrical parameters were monitored during the whole process including the heat keeping and electromagnetic treatment, which were acquired at intervals of 1 s automatically to ensure the continuity.

2.3. Voltage and current waveforms under different electromagnetic parameters
Electromagnetic field acted on liquid metals by the energy intervention, which changed the structure of liquid metals. External electromagnetic field was generated by applying a transient current to the induction coils. And this transient current could be obtained by RLC circuit. (The principle of RLC circuit was briefly described as follows. The current was generated by the pulse low-frequency power supply system which was developed by the

| Alloy Type | Fe content/wt.% | Alloy |  |
|------------|-----------------|------|---|
| Al         | 0               | Pure Aluminum |
| Al-0.99Fe | 0.99            | Hypoeutectic alloy |
| —         | 1.80            | Eutectic point [22] |
| Al-1.99Fe | 1.99            | Near-eutectic alloy |
| Al-2.27Fe | 2.27            | Hypereutectic alloy |
| Al-2.89Fe | 2.89            | Hypereutectic alloy |
| Al-3.50Fe | 3.50            | Hypereutectic alloy |
| Al-4.52Fe | 4.52            | Hypereutectic alloy |

Table 1. Types of Al–Fe alloys (hypoeutectic alloy or hypereutectic alloy) investigated in this work.
research group independently. 380 V three-phase alternating current was converted into 200 ~ 300 V AC voltage through isolation transformer. Then the alternating voltage was transformed into unidirectional pulsating DC voltage by the thyristor composed of one single-way conductivity rectifying elements. The damped oscillating current in the load coil was obtained by IGBT component and controller, load coil and non-polarized capacitor. Similarly, the external electromagnetic field changed with the current periodically. The electromagnetic parameters supplied by this power system included setting current (I/A), frequency (f/Hz) and duty cycle (D/%). These parameters directly affected the voltage and current inputted to the induction coils. When the current passed through the coil, the induced electric field, magnetic field and induced current excited in liquid metals, of which the interaction generated corresponding electromagnetic force or Lorentz force. Electromagnetic force or Lorentz force agitated the liquid metal, resulting in the transportation of mass, momentum, and energy. The current inputted to the electromagnetic coils affected the magnitude, direction, and distribution of the electromagnetic field acting on the liquid metals. The voltage and current waveforms showed the actual current applied to the electromagnetic coils. And the actual voltage and current waveforms under different AC electromagnetic parameters (setting current, frequency and duty cycle) were detected by TBS2000 digital oscilloscope.

Taking the experimental condition of setting current 50 A, frequency 20 Hz and duty cycle 20% as an example, figure 1 showed the screenshot of TBS2000 digital oscilloscope to give the actual voltage and current waveforms. It showed that the voltage waveforms were rectangular and the current waveforms were saw-tooth, which were attributed to electromagnetic control system. The voltage in a single cycle included applied and intermittent stages, and the current was divided into rising and falling stages correspondingly. The waveform in a single cycle was extracted in order to present the curves of voltage and current clearly.

3. Results

3.1. Effects of electromagnetic field on the electrical parameters

As the changes of electrical parameters reveal the variation of liquid structure due to electromagnetic treatment. And the liquid structure treated by electromagnetic field should be closely related to the initial liquid structure. Further, the initial liquid structure is attributed to content and temperature. It can be inferred that the effects of electromagnetic field on the liquid structure also depend on Fe content and treatment temperature. The liquid metals with different Fe content are studied at different treatment temperatures in this paper. Thermoelectric power is measured when the alloys are kept at constant temperature. Taking Al-2.89 wt.% Fe alloy at 980 K as an example, thermoelectric power behaviors during heat keeping including AC electromagnetic treatment (II stage) are shown in figure 2. Thermoelectric power shows stable during the heat keeping before treatment (I stage), and the average of thermoelectric power during I stage is 3.14 μV/K. When thermoelectric power stabilizes for at least 1800 s (I stage), the electromagnetic treatment is applied to the liquid metals for 300 s (II stage). Thermoelectric power increases with oscillation during the treatment (II stage), and then reaches a
local peak 6.25 μV/K at about 2100 s when treatment stops. After stopping electromagnetic treatment, thermoelectric power decreases with the extension of holding time, showing a recovery process. The liquid metals continue to maintain at the same temperature, and thermoelectric power shows a new stable value 3.45 μV/K (III stage). Difference of thermoelectric power between the initial average value (3.14 μV/K) before treatment and this local peak value (6.25 μV/K) when treatment stops is named as transient thermoelectric power variation ΔS, which can be used to characterize maximum variation of liquid structure due to the electromagnetic treatment. Although there are some values larger than this local peak during oscillation under electromagnetic treatment, in fact, the electrical parameters during treatment are caused by two effects (thermopower/Seebeck effect and Nernst effect). Because in the presence of electromagnetic treatment, the diffusion of carriers produces both transverse electric field $E_x = -S_{xy} |\nabla T|$ (thermopower, Seebeck effect) and longitudinal electric field $E_y = S_{xy} |\nabla T|$ (Nernst effect) [28–31]. So the electrical parameters during treatment process are not suitable to characterize the liquid structure. Only after stopping electromagnetic treatment, thermoelectric power can be used to characterize the change of liquid structure. After stopping electromagnetic treatment, thermoelectric power decreases with the extension of holding time. So thermoelectric power shows a maximum at the moment of stopping electromagnetic field, which can be used to characterize maximum variation of liquid structure due to the electromagnetic treatment. Then it needs duration time Δt for thermoelectric power to recovery into a new stable value 3.45 μV/K (III stage). And this difference from the initial thermoelectric power 3.14 μV/K (I stage) is defined as residual thermoelectric power variation ΔS_0.

The transient and residual variation of thermoelectric power and duration time Δt of alloys with different Fe content at different temperatures are extracted to reveal the variation of structure in liquid metals due to the electromagnetic treatment, which is discussed by figure 3 in detail.

Figures 3(a)–(c) show the characteristic parameters (transient variation, residual variation and duration time) of thermoelectric power with the increasing Fe content. The transient variation of thermoelectric power ΔS due to electromagnetic treatment is $2 \sim 7 \mu V/K$, and the residual variation ΔS_0 is not more than 0.6 μV/K, as shown in figures 3(a), (b). Massardier et al. have found that the variation of thermoelectric power in 6061 Alloy during aging is not more than 0.25 μV/K, which corresponds to β phase transformation [17]. This comparison shows that the variation of thermoelectric power obviously reflects the changes of liquid structure due to electromagnetic treatment. The transient variation at higher temperature (1003 K) is larger than that at lower temperature (980 K), as shown in figure 3(a). Because when the liquid metals have the same composition, the number of thermally activated clusters increases and the size of these clusters decreases with the increasing temperature. Liquid metal shows its own thermal activity at higher temperature, and the structure of liquid metal at higher temperature is weakly changed after electromagnetic treatment. Correspondingly, the transient variation of thermoelectric power shows smaller value. After stopping the electromagnetic field, thermoelectric power needs duration time Δt to recovery into a new stable value, as shown in figure 3(c). And during time Δt shows a local peak when Fe content of Al-Fe alloys is close to the eutectic point. The sharp increase of transient variation in figure 3(a) and the inflection point of residual variation in figure 3(b) have been found at the near eutectic composition (Al-1.99 wt.% Fe alloy), and the Al-1.99 wt.% Fe alloy needs a longer time for the thermoelectric power to recover into a stable value. Because the thermoelectric power due to electromagnetic treatment is not only attributed to Fe content, but also related to treatment temperature. As an important parameter to characterize the maximum variation of liquid structure, the transient variations at different treatment temperatures are also studied in figure 3(d). The results indicate that the transient variation of Al-1.99 wt.% Fe alloy (near-eutectic alloy) is about 4.23 $\sim 5.56 \mu V/K$, as shown in red rectangle of figure 3(d).
When the treatment temperature is similar or the same, the transient variation of near-eutectic alloy is larger than that of other alloys with different Fe content like hypoeutectic and hypereutectic alloys. As a function of structure factors, electrical parameters (thermoelectric power or electrical resistivity) respond to structure variation. Electrical resistivity can also be used to reflect the structure of liquid metals during electromagnetic treatment. The sample of Al-2.89 wt.% Fe alloy is kept at constant temperature to measure electrical resistivity $\rho$ in direct current (DC) electromagnetic field. Taking electrical resistivity versus time at 965 K as an example, electrical resistivity sharply increases from $289 \times 10^{-3} \, \mu\Omega \cdot \text{m}$ to $295 \times 10^{-3} \, \mu\Omega \cdot \text{m}$ due to DC electromagnetic treatment, as shown in figure 4(a). The variation of electrical resistivity $\rho$ is $6 \times 10^{-3} \, \mu\Omega \cdot \text{m}$, which is about 2% of the untreated initial value. Electrical resistivity keeps constant value during electromagnetic treatment, and an instant recovery is observed after stopping treatment.

![Figure 3. Characteristic parameters obtained from the thermoelectric power in AC electromagnetic field. (a) $\Delta S$ with Fe content (b) $\Delta S_0$ with Fe content (c) $\Delta t$ with Fe content (d) $\Delta S$ with treatment temperature.](image3)

![Figure 4. Electrical resistivity of Al-Fe liquid metals in DC electromagnetic field. (a) Electrical resistivity (b) electrical resistivity versus temperature and electromagnetic field intensity.](image4)
The average electrical resistivity during heat keeping and electromagnetic treatment is extracted, as shown in figure 4(b). It shows that the initial electrical resistivity increases with the increasing temperature. When the alloys are at the same temperature, electrical resistivity increases with the increase of electromagnetic intensity. It indicates that the variation of electrical resistivity is attributed to the energy related to electromagnetic treatment. This increase of electrical resistivity in electromagnetic field is called the magnetoresistive effect.

According to the above analyses, the average electrical resistivity during heat keeping and electromagnetic treatment is also extracted, as shown in figure 5(a). The electrical resistivity of liquid metals during the heat keeping increases with the increasing Fe content, indicating the difference of initial liquid structure due to Fe content. The electrical resistivity also increases with the increasing temperature. The variation of electrical resistivity has a sharp increase point at the eutectic composition, as shown in figure 5(b). And this result is comparable with the one in AC electromagnetic field.

### 3.2. Effects of electromagnetic parameters on the voltage and current waveforms

Figure 6 shows the effects of setting current, frequency and duty cycle on voltage waveform. We add 0.05 s (figures 6(a), (c)) or 0.1 s (figure 6(b)) to keep each curve separate. The results show that the peak of voltage is about 1500 V under different electromagnetic parameters including setting current, frequency and duty cycle. The voltage in a single cycle includes the applied and intermittent stages. The setting current does not change the voltage duration of applied and intermittent stages in a single cycle, as shown in figure 6(a). With the increase of frequency, the total duration of applied and intermittent stages decreases. But the proportion of the applied stages to the total duration remains unchanged due to the constant duty cycle 20%. The duration of applied stage is equal to the duty cycle multiplied by the time of a single cycle. It shows that the duration of applied stages increases with the increasing duty cycle.

Figure 7 shows the effects of setting current on current waveform when power system frequency is 20 Hz and duty cycle is 20%, which is consistent with the external electromagnetic field conditions in figure 6(a). As shown in figure 7(a), with the increasing setting current, the maximum and minimum currents applied to the electromagnetic coils increase significantly. When the setting currents are 20 A, 40 A, 50 A, 60 A and 80 A, the maximum currents applied to the electromagnetic coils reach 82.5 A, 180 A, 227.5 A, 272.5 A, 357.5 A. In addition, the current increment ($\Delta I = I_{\text{max}} - I_{\text{min}}$), namely the difference between the maximum and minimum currents, increases by 52.5 A, 107.5 A, 137.5 A, 160 A, 215 A respectively. The results show that the current increment applied to the electromagnetic coils also increases obviously with the increasing setting current. Under the same frequency and duty cycle, the current applied to the electromagnetic coils needs the same time (0.01 s) to reach the peak. It can be inferred that the rising rate of current increases with the increasing maximum current applied to the electromagnetic coils.

After measurement and calculation, the maximum/average current and the current-time integral in a single cycle as well as the magnetic induction intensity of this power system are obtained in figure 7(b). The results show that the current-time integral shows a nearly linear relationship with the average current in a single cycle. Magnetic induction intensity has a linear positive relationship with the current according to Biot–Savart law. It is pointed out that the maximum current affects the magnetic induction intensity. Meanwhile, the relationships between setting current and the maximum current/magnetic induction intensity in a single cycle are obtained.
\[ I_{\text{max}} = -5.375 + 4.588f_{\text{setting}} \text{ and } B = -6.055 + 2.369f_{\text{setting}}. \] After comparing the results of voltage and current, the output current of this power system can be regarded as a better evaluation index to the effects of AC electromagnetic parameters on the liquid structure of alloys. This quantitative method can clearly analyze the effects of AC electromagnetic parameters on the structure of liquid metals, which is characterized by thermoelectric power.

Figure 8 shows the effects of frequency on current waveform. As shown in figure 8(a), with the increase of frequency, the time for the current to reach the peak decreases gradually, and the maximum and minimum currents applied to the electromagnetic coils increase significantly. The current increment \( \Delta I = I_{\text{max}} - I_{\text{min}} \) is 132.5 A, 125 A, 127.5 A, 122.5 A, 127.5 A, 120 A, 125 A respectively, which remains unchanged. After measurement and calculation, the maximum/average/minimum current and the current-time integral in a single cycle are obtained in figure 8(b). The maximum/minimum current in a single cycle increases linearly with the increasing frequency, and the relationship formulas are as follows, \( I_{\text{max}} = a_1 + 5.375f_{\text{setting}} \) and \( I_{\text{min}} = a_2 + 5.607f_{\text{setting}}. \) The current increment \( \Delta I = I_{\text{max}} - I_{\text{min}} \) remains unchanged. With the increase of frequency, the current-time integral in a single cycle gradually decreases, that is to say, the area enclosed by the maximum current, minimum current and time in a single cycle gradually decreases.
Figure 9 shows the effects of duty cycle on current waveform. As shown in figure 9(a), with the increase of duty cycle, the time for the current to reach the peak increases gradually, and the maximum and minimum currents applied to the electromagnetic coils increase significantly. The current increment $\Delta I = I_{\text{max}} - I_{\text{min}}$ remains unchanged. After measurement and calculation, the maximum/average/minimum current and the current-time integral in a single cycle are obtained, as shown in figure 9(b). The current-time integral has a nearly linear relationship with the duty cycle, and increases with the increasing duty cycle gradually, which is similar to the results of current.

3.3. Effects of electromagnetic parameters on the electrical parameters

Meanwhile, the liquid structures of Al-10.06 wt.% Si hypoeutectic alloy and Al-15.15 wt.% Si hypereutectic alloy are also studied by measuring the thermoelectric power under different AC electromagnetic parameters correspondingly. The treatment temperatures of these two alloys are 908 K and 933 K respectively in order to keep the same superheat.

Figure 10 shows the curves of transient thermoelectric power variation $\Delta S$ and duration time $\Delta t$ with setting current due to electromagnetic treatment when the frequency is 20 Hz and duty cycle is 20%. The results show that these two parameters ($\Delta S$ and $\Delta t$) show a similar trend. In addition, the transient thermoelectric power variation $\Delta S$ of Al-Si hypoeutectic alloy exhibits the linear trend with the increase of setting current, while the one of Al-Si hypereutectic alloy shows the nonlinear trend. It can be inferred that the increasing trend of transient thermoelectric power variation $\Delta S$ is affected by the Si content in liquid metals. Similar results that the effects of alloy composition on thermoelectric power are also found in Al-Fe alloys, as shown in figures 3 and 5.

Further, the transient thermoelectric power variation $\Delta S$ of Al-Si hypereutectic alloy is not sensitive to the increase of setting current, which reflects that the change of liquid structure is not as large as the one of Al-Si hypoeutectic alloy.

When the setting current is less than 50 A, the transient thermoelectric power variation $\Delta S$ does not change significantly with the increasing setting current, and when the setting current increases to 50 A, the transient variation increases rapidly. According to the effects of setting current on the voltage and current waveforms, the
transient variation $\Delta S$ shows positive relationship with the current, magnetic induction intensity and current-time integral in a single cycle.

Figure 11 shows the curves of transient thermoelectric power variation $\Delta S$ and duration time $\Delta t$ with frequency in electromagnetic field when the setting current is 50 A and duty cycle is 20%. The results show that transient thermoelectric power variation fluctuates in a certain range with the increase of frequency when the frequency is less than 40 Hz. The transient variation $\Delta S$ of Al-Si hypoeutectic alloy decreases generally, while the one of Al-Si hypereutectic alloy is not sensitive to the increase of frequency. It can be inferred that the liquid structure of Al-Si hypereutectic alloy changes slightly and weakly with the increase of frequency under the electromagnetic field. When the frequency increases to 40 Hz, both the transient thermoelectric power variation $\Delta S$ and duration time $\Delta t$ start to increase rapidly. In other words, the effects of frequency on the liquid structure will be reflected when the frequency reaches a higher value. According to the effects of frequency on the current waveform, frequency has few effects on the transient thermoelectric power variation $\Delta S$ of liquid metals, which may be contributed to the fact that $\Delta I = I_{\text{max}} - I_{\text{min}}$ remains almost stable with the increase of frequency.

Figure 12 shows the curves of transient thermoelectric power variation $\Delta S$ and duration time $\Delta t$ with duty cycle in the electromagnetic field when the setting current is 50 A and frequency is 20 Hz. The results show that the transient thermoelectric power variation $\Delta S$ and duration time $\Delta t$ show a similar trend with the duty cycle. With the increase of duty cycle, the transient thermoelectric power variation $\Delta S$ of these two alloys decreases slightly and then increases (U or V-shaped for each alloy respectively). Compared with the results of these two alloys, Al-Si hypereutectic alloy shows larger transient thermoelectric power variation $\Delta S$ and smaller duration time $\Delta t$. According to the effects of duty cycle on current waveform, it can be indicated that lower duty cycle brings larger transient thermoelectric power variation $\Delta S$. Because the current needs shorter time to increase to the peak. By contrast, higher duty cycle also brings larger transient thermoelectric power variation $\Delta S$, which may be contributed to the larger current-time integral in a single cycle. In general, in order to obtain larger
transient thermoelectric power variation $\Delta S$ by electromagnetic field, we should increase the setting current at 10 Hz or 40 Hz, 50% duty cycle.

4. Conclusion

Thermoelectric power and electrical resistivity of aluminum binary alloys in alternating current (AC) and direct current (DC) electromagnetic fields have been measured in this paper. The results show that the thermoelectric
power increases due to the AC electromagnetic treatment, and electrical resistivity increases due to the DC electromagnetic treatment, indicating that the above two electromagnetic treatments change the liquid structure. Study on Al-Fe alloys with different Fe content reveals that both thermoelectric power and electrical resistivity show the inflection points at the near eutectic composition. Then the effects of electromagnetic parameters on the voltage/current waveforms of power system and the electrical parameters of aluminum binary alloys have been further studied. After comparing the results of voltage and current waveforms, the output current of this power system can be regarded as a better evaluation index to the effects of electromagnetic parameters on the thermoelectric power. This quantitative method can clearly analyze the effects of electromagnetic parameters on the structure of liquid metals, which is characterized by thermoelectric power. Transient thermoelectric power variation $\Delta S$ shows positive relationship with the current, magnetic induction intensity and current-time integral in a single cycle. In addition, transient thermoelectric power variation $\Delta S$ shows obvious local peaks at some specific frequencies, and decreases and then increases with the increasing duty cycle.

Acknowledgments

This work is supported by The Science and Technology Planning Project of Inner Mongolia [2020GG0175].

Data availability statement

The data used to support the findings of this study are available from the corresponding author upon request.
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