温泉科学（J. Hot Spring Sci.），63，317-327（2014）

原　　著

Effects of Magnesium Hydride as a Reductive Bath Additive on the Skin

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(Received Sept. 27, 2013, Accepted Jan. 22, 2014)

還元系水素入浴剤としての水素化マグネシウムの皮膚に及ぼす効果

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要　　旨

筆者らは，これまでに温泉水の本質的特性は還元系であることを提案し，その還元系を実現するには，水素発生剤である水素化マグネシウムの有効性を明らかにしてきた．また，水素は浴槽水を還元系にするだけでなく，電解で生成した水素を溶解させた水素浴槽水への継続的な入浴で皮膚の弾力性が向上することを報告した．そこで今回，実際の芦野温泉（アルカリ性単純温泉，栃木県）の源泉かけ流し浴槽に，水素化マグネシウムを添加し，電解での水素浴槽水で皮膚の弾力性向上効果が観察された水素濃度 50 ppb（μg/kg）以上にした水素化温泉水を準備した．実験は 2013 年 1 月中旬から真冬の 1 ヶ月間，ボランティアによる継続的入浴を行い，皮膚（前腕屈側）の弾力性を測定した．水素化マグネシウムを添加しない芦野温泉水および家庭浴槽水（シャワー浴も含む）への継続的入浴者との 3 グループで比較を行った．その結果，皮膚の弾力性では，家庭用浴槽入浴者では低下し，未処理の芦野温泉入浴者では変化は見られなかったが，水素化温泉水入浴者では弾力性向上効果が統計的有意差をもって観察された．今回の入浴実験開始直前の皮膚の弾力性データは，加齢により皮膚の弾力性が低下するとしたこれまでの文献データとよく一致したことから，水素化温泉水への継続的な入浴で，皮膚の老化抑制効果が期待できることが示された．

さらに，水素化浴槽水への入浴は，人工炭酸泉（二酸化炭素濃度 1,000 mg/dm^3）と同様に深部体温（鼓膜温）を上昇する効果も有することが確認できた．

キーワード：水素化マグネシウム，水素化温泉水，芦野温泉（栃木県），皮膚の弾力性，深部体温（鼓膜温）

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Abstract

We have proposed so far that natural hot spring source waters fresh out of wellheads have essentially a reductive characteristic, and that the use of magnesium hydride (MgH_2 ; hydrogen generating agent) is effective in preparing bathwater having a reductive characteristic equivalent to that of hot spring source waters. Moreover, in a series of our previous research papers, we have reported that, since hydrogen makes bathwater reductive, the elasticity of the human skin can be improved through habitual bathing in water hydrogenated by dissolution of electrolytically produced hydrogen therein (electrolyzed reductive water).

In the present study, magnesium hydride was added to non-circulated hot spring source water in bathtubs of Ashino Onsen (alkaline simple spring, Tochigi Prefecture) so as to prepare hydrogenated hot spring water having a hydrogen concentration exceeding 50 ppb (μg/kg), at which improvements in skin elasticity through habitual bathing in electrolyzed reductive water had been confirmed in our previous examinations. For a period of one midwinter month starting from the middle of January 2013, experiments of habitual bathing by volunteer subjects were carried out to measure the elasticity coefficients of the skin (flexural side of forearm) of each subject. With the purpose of comparison, the subjects were divided into three groups ; habitual bathing in hydrogenated Ashino hot spring water (prepared by addition of magnesium hydride), habitual bathing in intact Ashino hot spring water without the addition of magnesium hydride, and habitual bathing in home-use bathwater (including showering). The result data of the elasticity coefficients of the skin in the bathing experiments showed that there was a tendency to decrease in the group of subjects bathing in home-use bathwater, and no change was found in the group of subjects bathing in intact Ashino hot spring water, whereas a statistically significant improvement effect was observed in the group of subjects bathing in hydrogenated Ashino hot spring water. Further, skin elasticity data measured immediately before the start of the present bathing experiments were in good agreement with the hitherto reported literature data showing a tendency to decrease with age in skin elasticity. It is, therefore, suggested that habitual bathing in hot spring water hydrogenated by addition of magnesium hydride can contribute to providing an anti-aging effect on the skin. Besides, in an additional experiment of bathing in hot tap water hydrogenated by addition of magnesium hydride, an effective increase in deep body temperature (tympanic temperature) was confirmedly observed as in the case of artificial CO_2 hot spring water (CO_2 concentration : 1,000 mg/dm^3).

Key words : Magnesium hydride, Hydrogenated hot spring water, Ashino Onsen (Tochigi Prefecture), Skin elasticity, Deep body temperature (tympanic temperature)

1. Introduction

We have heretofore revealed that natural hot spring source waters fresh out of wellheads are essentially reductive in terms of oxidation-reduction potential (ORP), and that the ORP values of hot spring source waters increase due to oxidation over time after eruption thereof (Okouchi *et al*., 1998, 1999, 2000). Furthermore, through a series of our studies concerning the effects of hot spring waters on the skin, it has been elucidated that the skin, most susceptible to bathing conditions, is also essentially reductive and likely to oxidize with age as in the case of fresh hot spring source waters (Okouchi *et al*., 1999, 2000, 2002). Hence, we have proposed that habitual bathing in fresh reductive hot spring source water can suppress skin oxidation to contribute to anti-aging. Moreover, since skin lipid oxidizes with age to form 2-nonenal, causing an unpleasant aging body odor, habitual bathing in reductive hot spring source water is also recommendable for suppressing the aging body odor (Okouchi *et al.*, 1999, 2000, 2002). That is, reductiveness is of primary importance to hot spring water bathing and skin caring.

Thus, with our attention focused on means for artificially producing reductive bathwater, we have examined calcium-polysulfide-based bath additives (releasing hydrogen sulfide by reaction with water), and other hydrogenation treatments, i.e., magnesium hydride addition method as well as electrolysis method (Ohnami *et al.*, 2008a ; Okouchi, 2010 ; Kurita *et al.*, 2013). Unfortunately, however, the production of the calcium-polysulfide-based bath additives was discontinued under administrative direction for preventing the then-epidemic of suicide attempts using hydrogen sulfide. As for other hydrogenation treatments, we have been investigating electrolytic reduction of bathwater (electrolysis method) (Okouchi *et al.*, 2003, 2005), and addition of magnesium hydride (MgH_2 bath additive) into bathwater for hydrogen formation by reaction with water (magnesium hydride addition method) (Kurita *et al.*, 2013). With regard to investigations on the electrolysis method, experiments of habitual bathing in electrolyzed reductive water were conducted, resulting in confirmation of improvements in skin elasticity and hair sleekness/lustrousness (hydrogen penetration into skin and hair). With regard to investigations on the magnesium hydride addition method, it was confirmedly observed that skin elasticity can be improved even by habitually applying a magnesium-hydride-containing gel onto the skin, and that chlorinated oxidative tap water in a bathtub can be made reductive by adding magnesium hydride thereto. Further, in experiments at the spa facilities of Ashino Onsen (alkaline simple spring, Tochigi Prefecture), magnesium hydride was periodically added to non-circulated hot spring source water ($7.5\,m^3$), with the result that the ORP value thereof was decreased to indicate enhancement in the degree of reductiveness. Besides, as previously reported (Kurita *et al.*, 2013), hydrogenating conditions such as the amount of magnesium hydride to be added and the time interval of its addition were defined so as to satisfy a mean hydrogen concentration exceeding 50 ppb (= μg/kg), at which improvements in skin elasticity by the electrolysis method had been confirmed in our previous examinations.

In the present study in Ashino Onsen, to verify beneficial effects on skin elasticity in particular, experiments of volunteer subjects' habitual bathing were carried out using Ashino hot spring source water which was hydrogenated with magnesium hydride so that the mean hydrogen concentration noted above was provided.

Besides, according to the result data of a questionnaire survey regarding the experimental habitual bathing in the Ashino hot spring source water hydrogenated with magnesium hydride, most of the subjects remarked that a feeling of warmth after each bathing had persisted for a considerably long period. Then, for the purpose of further confirmation of the persistence of a feeling of warmth due to hydrogenation, an additional experiment of bathing in hot tap water hydrogenated with magnesium hydride was conducted to measure a possible increase in deep body temperature (tympanic temperature).

2. Experimental

2.1 Effects of reductive hydrogen bath additive on the skin

As a hydrogen generating agent, magnesium hydride (purity 90% ; Biocoke Lab. Co., Ltd., Japan) was used under the same conditions as those indicated in the previous report (Kurita *et al.*, 2013). More specifically, at the initial point of time and after a lapse of four hours during the bath open period of eight hours from 15 : 00 to 23 : 00, 15-gram magnesium hydride was added respectively into non-circulated hot spring source water running from No. 2 wellhead of Ashino Onsen (alkaline simple spring) for bathwater hydrogenation (7.5 m^3, 41℃). The ORP-pH relationships and hydrogen concentrations of the hydrogenated Ashino hot spring water were measured along with those of intact Ashino hot spring water and home-use bathwater (41℃) for confirmation in the same manner as reported previously. In determination of hydrogen concentration, a KM2100DH hydrogen concentration meter (manufactured by Kyoei Electronic Lab Co., Ltd., Japan) was used. For one month of the dry midwinter season starting from the middle of January, 2013, volunteer subjects were requested to do habitual bathing every day. With the purpose of comparison, the subjects were divided into the following three groups :

(1) habitual bathing (n = 16) in hydrogenated Ashino hot spring water (prepared by adding magnesium hydride into non-circulated hot spring source water running from No. 2 wellhead of Ashino Onsen),

(2) habitual bathing (n = 13) in intact Ashino hot spring water without addition of magnesium hydride (non-circulated hot spring source water running from No. 2 wellhead of Ashino Onsen), and

(3) habitual bathing (n = 13) in home-use bathwater, i.e., tap water (including showering).

Regarding the timing of hot spring water bathing, the subjects in the group of habitual bathing in hydrogenated Ashino hot spring water and in the group of habitual bathing in intact Ashino hot spring water were instructed to take a bath together with other hotel guests/visitors during the bath open hours from 15 : 00 to 23 : 00. The subjects in the group of habitual bathing in home-use bathwater (including showering) were instructed to take a bath at usual daily timing scheduled by each of them. The three groups of habitual bathing in hydrogenated Ashino hot spring water, in intact Ashino hot spring water, and in home-use bathwater consisted of 16 (male, 7 ; female, 9), 13 (male, 8 ; female, 5), and 13 (male, 6 ; female, 7) healthy subjects, respectively. The subjects in each group were in the age bracket of 40s to 70s.

With respect to effects of habitual bathing on the skin, the elasticity coefficients and water contents of the skin (flexural side of forearm) of each subject were measured for evaluation immediately before the start of the experiments of subjects' habitual bathing and the next day of one month thereafter (Okouchi *et al.*, 2005 ; Ohnami *et al.*, 2008b ; Kurita *et al.*, 2013). In determination of skin water contents, a MY-808S moisture checker (manufactured by Scaler, Japan) was used, and in determination of skin elasticity coefficients (skin elasticity determined as a ratio of a constricted skin state to a non-constricted skin state), a Cutometer SEM575 (manufactured by Courage & Khazaka Electronic GmbH, Germany) was employed, with which skin elasticity measurements were conducted in the following manner : Constant-negative-pressure suction was applied to the skin of each subject by using a probe having an opening of 2 mm in diameter, and the height of a skin level drawn into the probe was measured. Then, after unlocking the negative pressure, the height of a released skin level was measured, and the former height was compared with the latter to determine the elasticity of the skin.

Note that, prior to the implementation of the experiments mentioned above, the subjects were informed of the purposes of the present study and consented to participate therein.

2.2 Effects of reductive hydrogen bath additive on deep body temperature (tympanic temperature)

Magnesium hydride was added into hot tap bathwater (200 dm^3, 41℃) to prepare hydrogenated water, and using an artificial CO_2 balneotherapy device (manufactured by Mitsubishi Rayon Co., Ltd., Japan), artificial CO_2 hot spring water (CO_2 concentration : 1,000 mg/dm^3, 41℃) was prepared as control reference bathwater capable increasing a cutaneous blood flow to provide a thermotherapeutic effect. In experimental bathing in each of intact hot tap water, hydrogenated hot tap water, and artificial CO_2 hot spring water, healthy male subjects (5 persons, aged 22 to 25 years) bathed for a period of 15 minutes, and then during a period of 40 minutes thereafter, time-course measurements of a deep body temperature (tympanic temperature) of each subject were made in a resting state. The ambient resting-state conditions were maintained at a room temperature of 26℃ and a relative humidity of 50%. For tympanic temperature measurements, OMRON MC-510 thermometers (manufactured by OMRON Corp., Japan) were used.

3. Results and Discussion

3.1 Effects of reductive hydrogen bath additive on the skin

The hydrogen formation reaction of magnesium hydride (hydrogen generating agent) with water is expressed by Equation (1).

$$MgH_2 + 2H_2O \rightarrow 2H_2 + Mg(OH)_2 \tag{1}$$

Figure 1 shows an example of ORP-pH relationship of hydrogenated Ashino hot spring

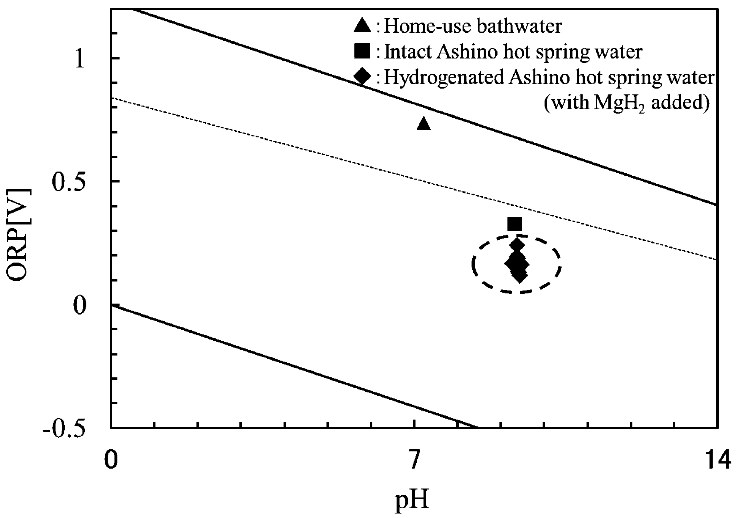

Fig. 1　ORP-pH relationships of hydrogenated Ashino hot spring water (with MgH_2 added), intact Ashino hot spring water, and home-use bathwater (41℃, respectively).

water, which was prepared by adding 15 grams of magnesium hydride into non-circulated hot spring source water (7.5 m^3, 41℃) at each of two time points during the bath open period of eight hours (◆). As shown also in this figure is exemplary ORP-pH relationships of intact Ashino hot spring water without addition of magnesium hydride (■) and of home-use bathwater (tap water, ▲). The upper and lower solid lines in Fig. 1 represent the boundaries of oxidative and reductive decompositions of water respectively, which are expressed by Equations (2) and (3).

$$\mathrm{ORP} = 1.23 - 0.059\,\mathrm{pH} \quad (2)$$

$$(O_2 + 4H^+ + 4e^- \rightleftarrows 2H_2O)$$

$$\mathrm{ORP} = -0.059\,\mathrm{pH} \quad (3)$$

$$(2H^+ + 2e^- \rightleftarrows H_2)$$

The broken line in this figure indicates the equilibrium ORP level (expressed by Equation (4)) that was empirically defined by us (Okouchi *et al*., 2002).

$$\mathrm{ORP} = 0.84 - 0.047\,\mathrm{pH} \quad (4)$$

More specifically, the oxidative region of water is located above the equilibrium ORP level indicated by the broken line in Fig. 1 (Equation (4)), and the reductive region of water is located below it. Note that “ORP” in Equations (2) to (4) represents a standard oxidation-reduction potential (V) with reference to a hydrogen electrode at a temperature of 25℃. As shown in Fig. 1, non-circulated hot spring source water of Ashino Onsen (intact Ashino hot spring water, ■) is in the reductive region, and hot spring water hydrogenated by addition of magnesium hydride (hydrogenated Ashino hot spring water, ◆) indicates lower ORP values, i.e., it is plotted at points shifted toward enhancement in reductiveness, similarly to the cases reported previously. In contrast, the ORP value of the home-use bathwater (▲) is in the oxidative region above the equilibrium ORP level due to residual chlorine (0.3～0.5 ppm) contained in tap water for disinfection.

Figure 2 shows two exemplary cases of changes with time in dissolved hydrogen concentration of hot spring water hydrogenated by addition of magnesium hydride during the bath open period of eight hours. In either case (●, First try on Jan. 22, 2013 ; ▲, Second try on Feb. 22, 2013), hydrogen concentrations exceeding 50 ppb can be confirmed during the bath open hours, with a few exceptions.

Figure 3 shows changes in the skin water contents of the subjects in the one-month experiments of habitual bathing in hydrogenated Ashino hot spring water (◇), intact Ashino hot spring water (□), and home-use bathwater (△). The skin water contents of the subjects in the three habitual bathing groups indicated a tendency toward slight decrease, but no statistically significant difference was observed in these groups. Note that there was no abnormal value in each group.

Figure 4 shows changes in the skin elasticity coefficients of the forearm flexural parts of the subjects in the one-month experiments of habitual bathing in hydrogenated Ashino hot spring water (◆), intact Ashino hot spring water (■), and home-use bathwater (▲). Regarding the skin elasticity coefficients, the subject group of habitual bathing in home-use bathwater (including showering) indicated a decrease corresponding to a statistically significant difference (++ : $p < 0.01$ in Fig. 4), whereas no change was observed in the subject group of habitual bathing in intact Ashino spring water. Contrastingly, the subject group of bathing in hydrogenated Ashino

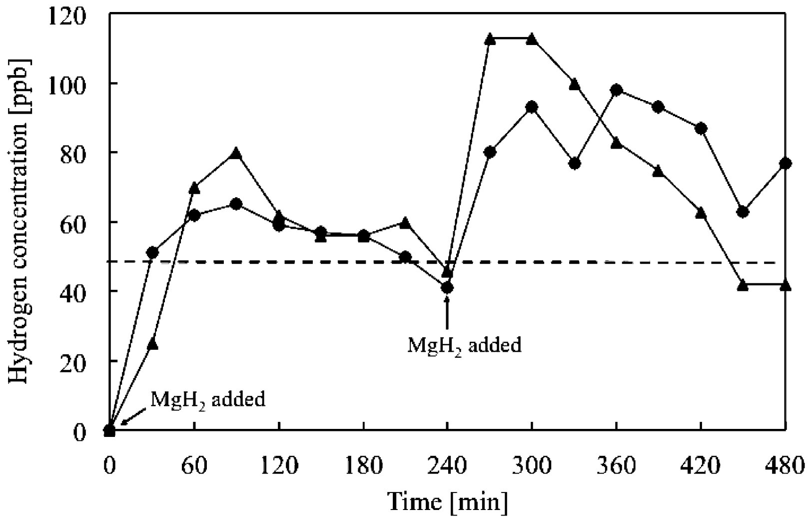

Fig. 2　Changes in dissolved hydrogen concentration with time of hot spring water hydrogenated by addition of MgH_2 (41℃).
●, First try on Jan. 22, 2013 ; ▲, Second try on Feb. 22, 2013.

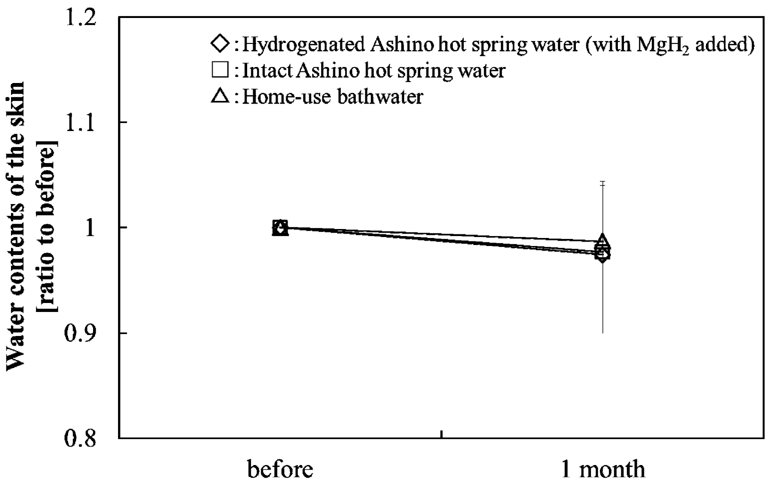

Fig. 3　Changes in the skin water contents of the forearm flexural parts of the subjects in the one-month experiments of habitual bathing in hydrogenated Ashino hot spring water (n=16), intact Ashino hot spring water (n=13), and home-use bathwater (n=13).

hot spring indicated a favorable increase (+ : $p<0.05$) in skin elasticity. That is, in comparison with the practice of habitual bathing in home-use bathwater incapable of preventing a decrease in skin elasticity, it was observed that both the practices of habitual bathing in intact Ashino spring water and in hydrogenated Ashino spring water were advantageous to provide a relatively higher level of skin elasticity in terms of statistical significance (** : $p<0.01$). More specifically, the practice of habitual bathing in hydrogenated Ashino spring water provided an

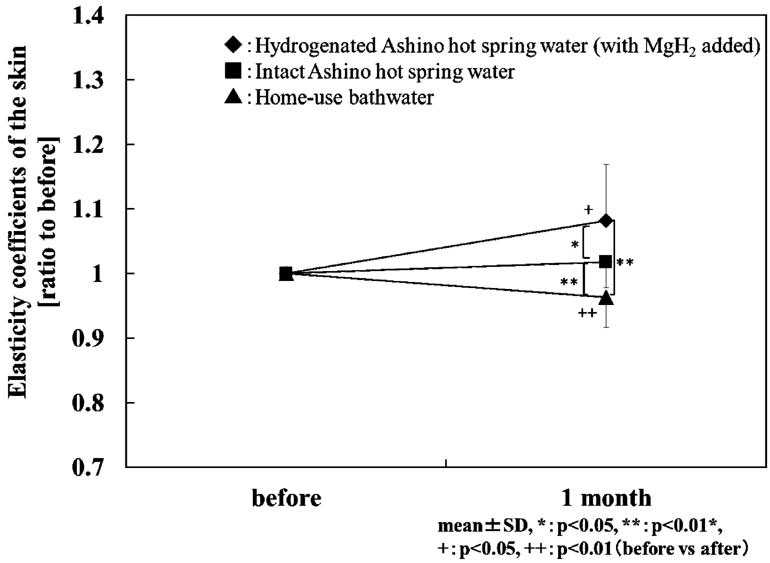

Fig. 4　Changes in the skin elasticity coefficients of the forearm flexural parts of the subjects in the one-month experiments of habitual bathing in hydrogenated Ashino hot spring water (n=16), intact Ashino hot spring water (n=13), and home-use bathwater (n=13).

improvement in skin elasticity (* : $p < 0.05$) over the practice of habitual bathing in intact Ashino hot spring water.

These experimental results demonstrate that a possible decrease in skin elasticity during a dry winter season can be suppressed by habitual bathing in intact hot spring water, and more noteworthily, that an effective improvement in skin elasticity can be attained by habitual bathing in hydrogenated hot spring water. Note that there was no abnormal value in each group.

Figure 5 shows the relationships between the skin elasticity constants (actual measured values) and ages of the subjects. Plotted in this figure are skin elasticity data measured immediately before the start of the present bathing experiments (○), and skin elasticity data measured immediately before the start of the hydrogen-gel application experiments reported previously (△ ; Kurita *et al.*, 2013), along with skin elasticity extracted from reference literature (the average skin elasticity constant and age of the subjects) authored by other researchers (◆ ; Cua *et al.*, 1990). These data, which indicate the skin elasticity coefficients of the forearm flexural parts of volunteer subjects, clarify a tendency to decrease with age in skin elasticity, giving good agreement with the reference literature data (reference values). That is, the data shown in Fig. 5 corroborate the preceding studies reporting a tendency to decrease with age in skin elasticity.

Hence, we suggest here that habitual bathing in hot spring water hydrogenated by addition of magnesium hydride can contribute to providing an anti-aging effect on the skin. Note that no skin damage/trouble due to the addition of hydrogen was observed in the present experiments, in which the spatial concentration of hydrogen released from 60 grams of magnesium hydride per day was estimated to be less than 1/300 of the limit of hydrogen explosion (4%) even under a closed bathroom condition. Moreover, hydrogen is used as a food additive that is safe in

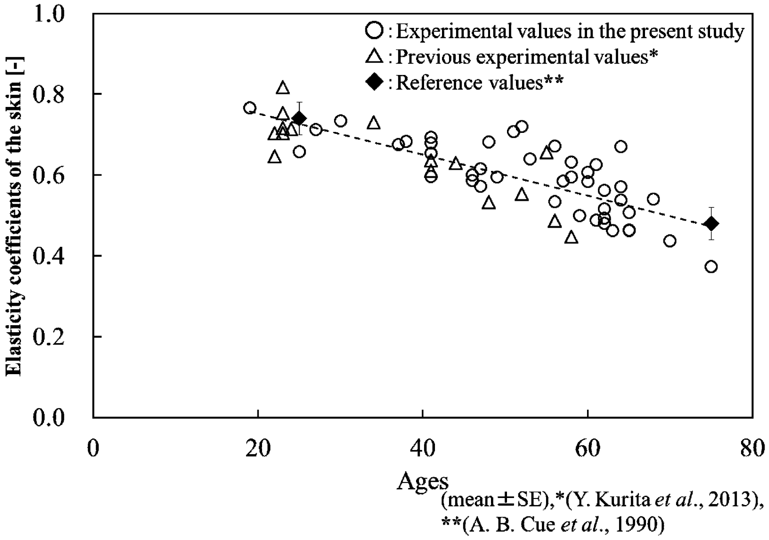

Fig. 5　Relationships between the skin elasticity coefficients (forearm flexural parts) and ages of the subjects.

health. Thus, safety is ensured in the use of magnesium hydride for hydrogenation of hot spring water.

3.2　Effects of reductive hydrogen bath additive on deep body temperature (tympanic temperature)

Figure 6 shows changes in deep body temperature with time (tympanic temperature) in experimental bathing in intact hot tap water (▲), hydrogenated hot tap water (with MgH_2 added ; ●), artificial CO_2 hot spring water (◆). In experimental bathing in hot tap water, an increase of 0.8℃ in deep body temperature was observed immediately after 15-minute bathing. In experimental bathing in hydrogenated hot tap water (with MgH_2 added) and artificial CO_2 hot spring water, an increase of 1.3℃ and an increase of 1.2℃ in deep body temperature were observed respectively, immediately after 15-minute bathing. The experimental results showed that, in comparison with the intact hot tap water, both of the hydrogenated hot tap water (with MgH_2 added) and artificial CO_2 hot spring water can provide a higher level of deep body temperature with statistically significant difference (* : $p<0.05$, ** : $p<0.01$, respectively). Hence, it has become apparent that an effective increase in deep body temperature is attainable by bathing in hot tap water hydrogenated with MgH_2 as in the case of artificial CO_2 hot spring water (Maeda *et al.*, 2001).

4.　Conclusions

In the present study, to examine possible effects of hydrogenated hot spring water (prepared by addition of magnesium hydride) on the skin, the experiments of habitual bathing by volunteer subjects were carried out for a period of one midwinter month. As a result of the

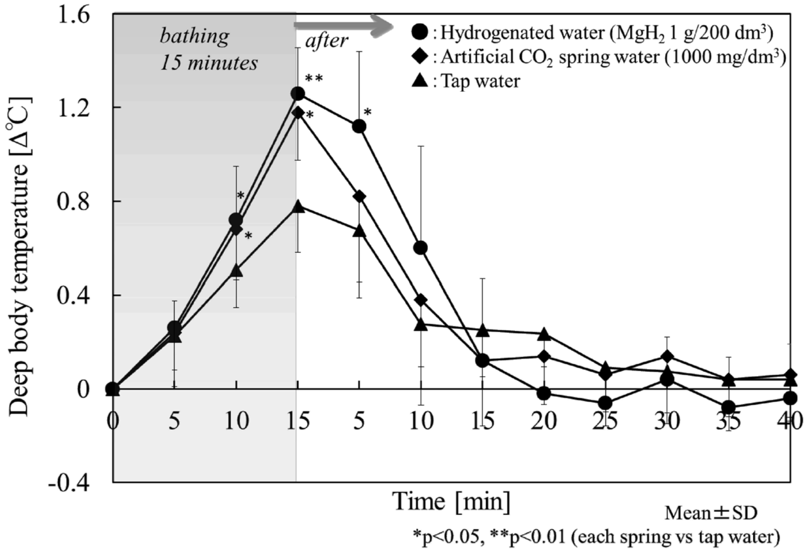

Fig. 6　Changes in deep body temperature (tympanic temperature) of bathing in hydrogenated water (41℃ with MgH_2 added ; n=5), artificial CO_2 spring water (n=5), and tap water (n=5).

experiments, it was confirmed that the addition of magnesium hydride into reductive non-circulated hot spring source water of Ashino Onsen enhanced the degree of reductiveness thereof to provide an improvement in skin elasticity with statistically significant difference. Contrastingly, in the experimental practice of habitual bathing in home-use bathwater (including showering), a decrease was found in skin elasticity, and in the experimental practice of habitual bathing in intact Ashino spring water without addition of magnesium hydride, no change was observed in skin elasticity. With respect to the water contents of the skin, although there was a tendency toward slight decrease, no statistically significant difference was found in any of the experimental practices of habitual bathing.

Through the experiments described herein, it has become definitely apparent that hydrogen is effective in enhancing skin elasticity. Nevertheless, skin-surface water content gives no significant effect to skin elasticity. In the preceding studies, the correlativity between skin elasticity and skin-surface water content has not yet been elucidated positively. It is therefore needed to investigate detailed conditions of interior skin tissue under the effect of hydrogen. Moreover, according to questionnaire result data obtained from the volunteer subjects who participated in the experimental habitual bathing in hydrogenated hot spring water, most of the subjects remarked that a feel of warmth after each bathing had persisted to cause continuous sweating for a considerably long period.

An additional experiment of bathing in hot tap water hydrogenated by addition of magnesium hydride was then conducted for further confirmation of the persistence of a feel of warmth due to hydrogenation. The results of this experiment showed that a higher level of deep body temperature (tympanic temperature) than that in bathing in intact hot tap water can be attained by bathing in hydrogenated hot tap water (with MgH_2 added) as in the case of artificial CO_2 hot spring water. Thus, as a physiological effect of bathing in hot tap water hydrogenated with MgH_2, an increase in deep body temperature has been confirmed.

Acknowledgments

The authors thank all the volunteers who made this investigation possible and employees of Ashino hot spring facility.

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