

• *Review article*

Indoor radon entry: 30 years later

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The paper discusses the results of 30 years of radon indoor entry research. While some progress has been made, many discrepancies between current theories and experimental data still remain. Misconceptions of radon transport mechanisms in homes are analyzed. The role of US Environmental Protection Agency in radon indoor entry research is examined and the agency's policies are criticized. The need to include thermotranspiration gas transport in the general model of radon indoor entry is substantiated. Iran. J. Radiat. Res., 2008; 6 (1): 1-6

Keywords: Radiation, radon, permeability, concrete, soil.

INTRODUCTION

Indoor radon entry (IRE) is a great public health concern. During the last three decades, intensive research has been done to clarify the mechanism of IRE. Numerous papers and scientific reports were devoted to these problems. US Environmental Protection Agency (EPA) and US Department of Energy (DOE) spent millions of dollars on researching the transport of radon into houses. Finally, US EPA declared that the pressure driven radon transport through cracks and openings in a concrete slab is the dominant mechanism of IRE. Since the pressure driven mechanism has been recognized as the dominant mechanism of IRE, US EPA and US DOE ceased sponsoring research on this topic. However, the understanding of the problem has not progressed much beyond the initial stage^(1, 2). Numerous misconceptions about radon transport in homes retard progress in

creating reliable, passive mitigation methods intended to reduce radon concentrations in homes. Active soil depressurization, as recommended by the EPA USA, is energy consuming, costly, and is not based on understanding of radon transport. Recognition of this fact would stimulate research on indoor radon transport and create effective and reliable radon prevention and mitigation techniques.

Criticism of indoor radon entry theories

Advective viscous pressure-driven mechanism of IRE through concrete openings is caused by indoor/outdoor temperature difference, which in turn causes indoor/outdoor pressure difference and air/radon bulk motion (stack effect). When indoor temperature is greater than outdoor temperature, anticipated indoor pressure is less than outdoor pressure and, therefore, viscous pressure-driven air flux through concrete cracks and openings from underground is expected. Although there are physical reasons for this effect, the calculation of indoor/outdoor pressure differential is based on oversimplified static model and the current theory does not allow for predicting the IRE in a specific dwelling. Review of numerous discrepancies and contradictions between the predictions of theory and experimental data is given by

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Minkin ^(1, 2). For example, stack effect should not take place while indoor temperature is less than outdoor temperature. In this case, mostly realized during summers, indoor pressure may exceed outdoor pressure, which should lead to large decrease in radon indoor flux. But a lot of measurements did not detect any significant change in radon indoor flow and, moreover, many scientists observed higher indoor radon concentration in the summer than in the winter. Also, the sealing of cracks and openings in concrete slab must be the direct passive mitigation of IRE, caused by the stack effect. However this mitigation is not effective and radon concentration often remains unchanged after this technique is applied. The contradiction between IRE model predictions and numerous experimental data is sufficient foundation to reject the hypothesis of prevalence of pressure-driven mechanism of IRE. Debates and skepticism about the understanding of the IRE has been continuing in the scientific community ^(3, 4), and it was premature for US EPA to establish the pressure-driven mechanism as the dominant mechanism of IRE.

The history of science and physics particularly, shows that discrepancies between the experimental facts and the leading theory were the driving force for emergence of powerful new ideas. It appears that new ideas have to be put forward to understand IRE transport. However, despite the evidence that pressure-driven mechanism is not able to explain numerous experimental data and cannot be considered as the primary mechanism in many practical cases, the recent trend in developing an IRE model has been aimed at improving and modernizing this pressure-driven model. The idea of scale dependence of soil permeability to adjust the pressure-driven model to experimental data was introduced ⁽⁵⁾. The paper is not helpful in explaining the presented uncertainties and is ineffective in explaining experimental facts which contradict the predictions of the viscous advective transport concept. The similitude theory states that no real laws of nature can depend on the choice of system of units or scale measurements; any real law can be

presented as a relationship between dimensionless quantities, the so-called similarity parameters (π -theorem). If "it is the structure (heterogeneity) of the soil that can produce the scale effect" ⁽⁵⁾, the effect should not be called scale effect. The authors also ignore the principle of dimension theory defining the dimension of two parameters (L', z') as $(\text{radian})^{\frac{1}{2}}$ (radian is dimensionless value). In addition, they provide all formulas in SI units making the odd exception for dynamic viscosity of soil gas (kg/ms).

Furthermore, authors consider soil air as an incompressible isothermal ideal gas. First of all, ideal gas is compressible and its isothermal compressibility (bulk modulus), B , can be easily calculated

$$B = -V \left(\frac{\partial P}{\partial V} \right)_T = P$$

Moreover, in incompressible fluid the speed of pressure signal propagation is infinite.

Also, the authors analyze the linear differential equation for pressure signal propagation in soil. However, it was shown ⁽⁶⁾ that the linearization of the equation of pressure propagation in soil is possible for constant soil permeability and specific time independent boundary conditions (not harmonic function as considered by the authors). Therefore, it was illegitimate to analyze the linear equation. As a phase shift of pressure signals is the key factor for the authors' method of measuring soil permeability, it must be the first priority to find phase transformation of the signal in non-linear system with variable propagated parameters (perhaps the one of the most difficult problems in physics ⁽⁷⁾). Definitely, in this case there is change of output signal spectrum (it will not be monochromatic for sinusoidal input signal). The simplified estimation shows that their results are doubtful. Indeed, from general theory of one dimensional stationary fluid flow through porous material, it follows that for two soil layers equal length with permeability K_1 and K_2 the total permeability K is:

$$\frac{1}{K} = \frac{1}{2K_1} + \frac{1}{2K_2}$$

From this formula one can see that

permeability K can be greater than K_1 or less than K_1 . Therefore, the conclusion that the permeability increases with the sample size is wrong (actually it depends on the values K_1 and K_2). Moreover, if the permeability of the soil K_2 , which is close to home, even ten thousand times greater than K_1 , the total permeability

$K \approx 2K_1$, and therefore it is not likely that permeability increases by factor 20 as a sampling size increases from 0.1m to 2 m ⁽⁵⁾. Actually, authors could not find a solid explanation of their result. To our knowledge, this so-called scale permeability effect is not confirmed by other researchers.

As there are obvious difficulties in explaining experimental facts, research of influence of different factors which could affect IRE has been done. Wind was the factor of investigation but no definite conclusion on this issue has been reached ⁽⁸⁾.

The hypothesis of temperature dependence of radon sorption as a substantial factor of radon transport was also put forward ⁽⁹⁾. Although the hypothesis is rational, the enthalpy of radon sorption on silicate is small and it is unlikely that temperature change can substantially influence radon concentration in the gas phase of porous materials ⁽¹⁰⁾.

Another explanation of high indoor radon concentrations is given in physics textbook ⁽¹¹⁾: the authors concluded that radon gas enters houses through cracks in the foundation, and tends to collect there since it is denser than air. In assumption of isothermal ideal gas equilibrium (that is successfully done often for gas distribution in atmosphere), Boltzman's distribution can be applied for radon vertical concentration distribution:

$$\frac{n}{n_0} = \exp\left(-\frac{\mu gh}{RT}\right)$$

Where n is radon concentration at height h , n_0 is radon concentration at $h=0$ m, g is acceleration due to gravity ($g=9.81$ m/s²), $\mu=2.22$ kg/mol is radon molar mass, $R=8.31$ J/(mol K) is gas constant, T is temperature in Kelvin scale. For $T=293$ K and $h=6.00$ m this ratio is 0.948. This means that radon concentration differs only by 5% between the

zero level and a height of 6.00 m; for air ($\mu=0.0290$ kg/mol) the ratio $n/n_0=0.999$ and air concentration at height difference of 6.00 m is about only 0.1%. Although the distribution of main atmospheric gases and radon differs with height and radon concentration in atmosphere decreases much faster with height than principle atmospheric gases, it is not a factor for radon to be concentrated in home foundations. For residential houses, radon concentration should not exceed 5% as a result of differences in height.

There is no also clear understanding of the mechanism of radon transfer through intact concrete. Concrete (and clay) has pores belonging mostly to the Knudsen region for which mean free path of molecules is much greater than pores radii. This means that the mechanism of gas transfer in these materials is free molecular. In this case there is practically no collision between molecules of gases; they collide primarily with the capillary walls and there is no diffusion or viscous fluxes of gases. Nevertheless, equation for one-component gas flow in free molecular region can be written in the form of Fick's diffusion law or Darcy's law: both laws formally can be used and both describe the same mechanism of gas transport. Therefore, in this case, there exists a simple relationship between diffusion and permeability coefficients. However there is no viscous flux in this case. It has been shown that by conveniently defining the diffusion coefficient and the permeability coefficient, Fick's law and Darcy's law can be made formally similar and either can be used to describe the transport of gases and vapors through porous materials ⁽¹²⁾. Concrete and many building materials have pored size distribution that mostly pertains to Knudsen's region and a transition zone (for which mean free path of molecules is the order of diameters of pores). Numerous papers devoted to air/radon transport, consider air advective transport through home concrete foundation using Darcy law:

$$j = K \nabla P = \frac{k}{\eta} \nabla P$$

Where j is gas flux in porous media, K is

permeability coefficient, η is gas viscosity, $k=K\eta$ is intrinsic permeability (which depends only the structure of porous material), and ∇ is a gradient operator. However, gas flux in concrete is not viscous (gas flow cannot be considered as motion of continuous media) and there is no much sense to introduce the viscosity of gases in such structure. η is not constant for given pressure and temperature but depends on pores radii. For a uniform cylindrical pore according Hagen-Poiseuille equation intrinsic permeability is $k = \frac{r^2}{8}$. For intact concrete, intrinsic permeability is approximately $k=1.0 \cdot 10^{-16} \text{ m}^2$ (13). Substituting this k in previous equation, one can find a radius of a capillary $r=2.8 \cdot 10^{-8} \text{ m}$. This radius is much less than the mean free path of molecules of air for atmospheric pressure which is approximately $0.1 \mu\text{m}$. This means that gas flow is free molecular. These calculations are very rough and should be modified slightly taking into account porosity of material, ϵ , and tortuosity factor τ . In this case $k = \frac{\epsilon \bar{r}^2}{8\tau}$, where \bar{r}^2 is the mean of r^2 . Concrete porosity is approximately 0.15 and tortuosity $\tau \approx 3$ is the upper limit (14). Substituting these parameters into the last equation, one can find root mean square radius of concrete pores $r_{\text{rms}} = \sqrt{\bar{r}^2} = \sqrt{\frac{8k\tau}{\epsilon}} = 0.073 \mu\text{m}$. Actually, for any pore size distribution, the mean radius, \bar{r} , is smaller than r_{rms} and therefore $\bar{r} < 0.073 \mu\text{m}$. This indicates that concrete pores belong mostly to the free molecular and transition regions and gas flow in this media is not viscous. The direct measurements of concrete pores using mercury and nitrogen adsorption methods confirm this statement (15). To our knowledge, in contrast to European general physics textbooks, none of US general physics textbooks include the kinetic theory of gas transfer phenomena (mass, impulse, energy) for Knudsen's and transition zones.

AIR/Radon thermotranspiration in concrete and soil

It was shown both theoretically (based on kinetic theory and irreversible thermodynamics) and experimentally that

thermodiffusion (thermotranspiration) can be a significant factor of radon transport in soils and concrete slabs (1, 2, 10). Conceptually, a thermogradient always induces mass transport in a multi-component system, such as a gas mixture, gas dissolved in solid materials, electronic gas in metals, admixtures alloys in metals, and gas in porous materials. Usually, thermogradient in houses' concrete slabs points up. Thermodiffusion air/radon flux has the same direction and increases radon infiltration in houses. This flux can be greater than diffusion radon flux (1, 2). The possibility of prevalence of thermodiffusion air flow through porcelain and clay was demonstrated experimentally (16-18). The thermodiffusion effect is also dominant for isotope separation in nuclear physics and essential for migration of alloys in metallurgy and the semiconductor industry. Apparently, thermodiffusion was underestimated in soil physics. Many soils have a large fraction of pores belonging to Knudsen's and transition zone (19). As there is temperature gradient in soils (temperature waves), thermodiffusion gas flow must arise. This effect will influence volatile organic contaminant transport, crop production, soil micro-organism transpiration, gaseous exchange between soil and atmosphere, and global air composition, as soil is a gigantic air filter.

CONCLUSION

The summary of 30 years of IRE research is not optimistic:

1. It should be recognized that 30 years of IRE research did not give the positive results. Radon transport is still a real problem and new approaches have to be explored for solving this thorny problem.
2. The US EPA's claim that pressure-driven mechanism is the dominant mechanism of IRE does not have enough scientific support since there are numerous experimental data contradicting this statement.
3. The declaration that radon collects in

- homes' foundations because it is a heavy gas is wrong.
4. The assertion that adsorption is an important factor of radon transport is doubtful.
 5. The hypothesis that soil permeability increases with the soil sample size is not justified.
 6. Passive mitigation of IRE is not reliable. Active Soil Depressurization and Building Pressurization, recommended by the US EPA of IRE mitigation, are costly and energy consuming. They are not based on the knowledge of the mechanisms of IRE and must work for any mechanism of radon transport, as it is obvious that suctioning of underground radon and diluting the radon concentration in homes will lead to a reduction of radon concentration in homes. It was not necessary to spend millions of taxpayer dollars to come to this self-evident conclusion.
 7. The theory of IRE is not adequate and it is practically impossible to predict IRE in a specific dwelling. Although numerical methods and computer programs were developed to solve complicated partial differential equation of radon transport in houses, the experimental data and calculations are frequently not in agreement. This indicates that the mechanism of IRE is not understood properly and model of IRE is not comprehensive.
 8. Gas (radon) flow through intact concrete is neither viscous nor diffusive. It is mostly free molecular flow. Direct measurements of air permeability of concrete confirm this statement.
 9. In many cases, gas (radon) thermotranspiration in soil and concrete may be an important and dominant transport of IRE. Thermotranspiration radon transport in soil may also explain diurnal and seasonal radon atmospheric fluctuations ⁽²⁰⁾. Further research of this gas transport mechanism is needed.
 10. Some physics textbooks ^(11, 21) and popular scientific journals ^(22, 23, 24) give the wrong

message that the mechanism of IRE is well understood and that pressure driven radon indoor flux through concrete cracks and openings is the primary mechanism of IRE. It should be point out that Cutnell and Johnson ⁽²⁵⁾ acknowledge in the last edition of their physics textbook that the mechanism of IRE is not properly understood.

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